

ANNUAL REPORT 2016

INSTITUTE OF ION BEAM PHYSICS
AND MATERIALS RESEARCH

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Annual Report 2016

**Institute of Ion Beam Physics
and Materials Research**

Editors

J. Fassbender, V. Heera,
M. Helm, P. Zahn

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Cover Picture

The cover picture shows a snapshot of the induced changes of the electron charge density in a graphene sheet while the incoming highly charged ion projectile traverses the layer from the top. The polarization of the layer due to the ion's electric field and the excitation of the graphene layer while and after the crossing of the ion are clearly visible.

The width of the figure covers a length of two nanometers.

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For further information see:

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Preface by the directors

After having allocated a tremendous amount of time to the integration into the Helmholtz Association, the initiation of collaborative research with other Helmholtz centers, and the Helmholtz program oriented funding evaluation, we could focus more on research again in 2016. This was very successful and we are particularly delighted in view of the numerous publications in high-impact journals, including ACS Nano, Advanced Materials, Applied Physics Review, Nano Letters, Nature Communications and Nature Nanotechnology, which have been published by scientists from the Institute. You will find a few of these highlight publications on the subsequent pages, demonstrating the broad range of science the Institute is addressing. Important results have been achieved in our already well established areas of interaction of highly charged ions with solids, THz spectroscopy and magnetization dynamics. The potential of our recently installed semiconductor molecular beam epitaxy system was demonstrated by growing epitaxial GaAs nanowires on Si at CMOS-compatible temperatures.

In addition, also users of our Ion Beam Centre (IBC) were very productive. Most notably users of our DREsden Accelerator Mass Spectrometry (DREAMS) facility within the IBC were able to publish their results in the most prestigious journals Science and Nature. We would like to congratulate them and would be happy to welcome more users from this area of science which is rather new to our Institute.

Excellent research is only possible, if there is a critical mass of people working cooperatively together. In order to further increase the number of scientists, the acquisition of external funds is inevitable. Also in this respect 2016 was a very successful year. In addition to the two already running EU-projects we have received funding for two new collaborative projects funded by the European Union. In one of them, Dr. Gregor Hlawacek further exploits the use of Helium Ion Microscopy for the characterization of nanoparticles. In the other, an FET-Open project which in total had only 4 % of funding probability, Dr. Alina Deac and her team will extend the communication characteristics of spin torque resonators to the Terahertz regime. Also three of our young investigators, Dr. Rantej Bali, Dr. Alexei Pashkin and Dr. Richard Wilhelm, successfully applied for projects from the Deutsche Forschungsgemeinschaft.

A major achievement in 2016 was to attract Prof. Dr. Olav Hellwig, who had been working for more than a decade in California on magnetic storage media for IBM, Hitachi GST and Western Digital, to a joint appointment between TU Chemnitz and HZDR. Prof. Hellwig started his professorship with TU Chemnitz and will, in addition to his Chemnitz group, be able to establish a second group at our Institute, working on functional magnetic materials due to a special grant he received from the Helmholtz Association. We wish him a quick start off and look forward to the new expertise we gain in magnetic thin film growth and characterization.

With the end of 2016 the division on structural materials led by Dr. Eberhard Altstadt and Dr. Cornelia Heintze left the Institute and is now part of the Institute of Resource Ecology. This move was necessary to better integrate their research into the Helmholtz programme "Nuclear Safety" which is mainly operated by the Institute of Resource Ecology. We thank all members of this division for their contributions to our Institute and wish them a good reception and working environment in their new Institute.

Last but not least, we recently received the information that our Emmy Noether group leader Dr. Helmut Schultheiß will receive the prestigious Walter Schottky Prize 2017 from the Deutsche Physikalische Gesellschaft for his contributions to the understanding of spin wave propagation phenomena in nanostructures and their application in new functional devices. We sincerely congratulate him on this excellent achievement.

Finally we would like to cordially thank all partners, friends, and organizations who supported our progress in 2016. Special thanks are due to the Executive Board of the Helmholtz-Zentrum Dresden-Rossendorf, the Minister of Science and Arts of the Free State of Saxony, and the Ministers of Education and Research, and of Economic Affairs and Energy of the Federal Government of Germany. Numerous partners from universities, industry and research institutes all around the world contributed essentially, and play a crucial role for the further development of the institute. Last but not least, the directors would like to thank again all IIM staff for their efforts and excellent contributions in 2016.



Prof. Manfred Helm



Prof. Jürgen Fassbender

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The background features a light gray field with a pattern of small, semi-transparent circles. A spiral of these circles converges towards the center, creating a sense of depth. In the lower half, a large, white, three-dimensional swirl or vortex shape is visible, with the text overlaid on it.

Selected Publications

Copyright remarks

The following journal articles are reprinted with kind permission from:

Wintz, S.; Tiberkevich, V.; Weigand, M.; Raabe, J.; Lindner, J.; Erbe, A.; Slavin, A.; Fassbender, J.

Magnetic vortex cores as tunable spin-wave emitters

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Slow Noncollinear Coulomb Scattering in the Vicinity of the Dirac Point in Graphene

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Magnetic vortex cores as tunable spin-wave emitters

Sebastian Wintz^{1,2,*}, Vasil Tiberkevich³, Markus Weigand⁴, Jörg Raabe⁵, Jürgen Lindner¹, Artur Erbe¹, Andrei Slavin³ and Jürgen Fassbender^{1,2}

The use of spin waves as information carriers in spintronic devices can substantially reduce energy losses by eliminating the ohmic heating associated with electron transport. Yet, the excitation of short-wavelength spin waves in nanoscale magnetic systems remains a significant challenge. Here, we propose a method for their coherent generation in a heterostructure composed of antiferromagnetically coupled magnetic layers. The driven dynamics of naturally formed nanosized stacked pairs of magnetic vortex cores is used to achieve this aim. The resulting spin-wave propagation is directly imaged by time-resolved scanning transmission X-ray microscopy. We show that the dipole-exchange spin waves excited in this system have a linear, non-reciprocal dispersion and that their wavelength can be tuned by changing the driving frequency.

Spin waves in ferromagnetic materials have been studied since the beginning of the twentieth century¹ (Fig. 1). The corresponding subfield of magnetism is now known as ‘magnonics’, named after the quasiparticle of spin-wave excitations, the magnon^{1–3}. In general, spin waves propagate much more slowly than electromagnetic waves, which makes their wavelengths λ up to five orders of magnitude smaller ($\sim 1 \times 10^{-6}$ m) than those of electromagnetic waves ($\sim 1 \times 10^{-1}$ m) of the same microwave frequency ($f \approx 1$ –10 GHz)⁴. This property stimulated interest in the application of spin waves as information carriers in microwave signal processing, which would allow for the development of miniature devices with large and easily controllable time delays.

Besides energy, spin waves also carry spin angular momentum. They can therefore be used in nanoscale spintronic signal-processing devices^{5–9}, where the information is coded with the spin orientation. Spin waves as information carriers are very flexible, because they can propagate in both magnetic conductors and insulators, and their dispersion relations can be ‘tailored to order’ by applying external magnetic bias fields⁴. Finally, their propagation is not associated with the motion of charge, so ohmic losses are avoided, and the power consumption for information processing can be reduced substantially.

A major challenge for the use of spin waves in nanosized spintronic devices is the requirement to excite propagating spin waves with nanoscale wavelengths. The traditional method of linear spin-wave excitation using spatially confined oscillating magnetic fields (with dimension on the order of the smallest achievable λ) is difficult to implement because of technological

restrictions in fabricating nanoscale transducers and matching their impedance to that of conventional transmission lines¹⁰. Only recently has the challenge of generating short spin waves been partly solved by reducing the wavelength after the generation process¹¹. The second well-known method of spin-wave excitation is parametric pumping, which involves the creation of spatially uniform double frequency oscillating magnetic fields^{4,12}. However, this method suffers from a relatively low excitation efficiency, non-locality and poor control of the excited wavelengths.

Another recently developed method of spin-wave excitation utilizes spin-transfer torques, which are induced via spin-polarized^{13–15} direct currents in magnetic nanocontacts. This method allows one to excite propagating spin waves, predominantly in the case

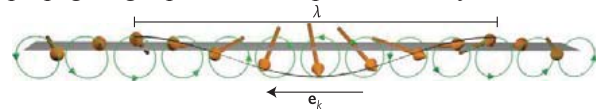


Figure 1 | Schematics of a spin wave propagating along \mathbf{e}_k . Magnetic moments (orange arrows) precess with a periodic spatial phase difference, determining the spin-wave wavelength λ .

where the oscillating magnetic layer is magnetized out of plane^{5,16–18}, which at the same time requires rather large external magnetic bias fields. For in-plane magnetization, which can be excited in smaller fields, spin waves resulting from spin-transfer torques are typically localized near the nanocontact^{5,18–20}. Therefore, such locally generated spin waves can only be used for the transmission of microwave signals when they are coupled into waveguides that provide a specific

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propagation channel for them²¹.

Vortex core spin-wave emitters

Here, we experimentally realize a new method for the excitation of short-wavelength propagating spin waves in layered magnetic structures. For this purpose we use the driven rotational dynamics of naturally formed nanosized magnetic vortex cores in an antiferromagnetically coupled trilayer without magnetic bias fields. The nanoscale size of the vortex cores ensures the excitation of spin waves with sufficiently small wavelengths. Meanwhile, the driven vortex core rotation, together with the rather linear spin-wave dispersion in the antiferromagnetically coupled trilayer, provides linear tunability of the excited wavelength by varying the driving frequency.

Magnetic thin-film elements in a certain size range exhibit a magnetization (**M**) ground state of a flux-closing, planar vortex with a nanoscale core of perpendicular magnetization in the centre (Fig. 2a)^{22–24}. Such magnetic vortices possess a non-trivial topological structure, characterized by their unique winding number of +1 and their binary core polarity of $p = \pm 1$ (ref. 25). Here, $p = +1$ corresponds to an upwards pointing core magnetization. Another vortex state parameter is the circulation $c = \pm 1$, where positive circulation $c = +1$ corresponds to an anticlockwise rotation of the static magnetization in the vortex plane. The magnetodynamic excitation spectrum of a vortex under the influence of laterally oscillating magnetic fields includes the gyration of the vortex core around its equilibrium position^{26–30}, as well as standing radial and rotating azimuthal dipolar spin-wave modes^{31–33}. A pair of stacked vortices can be confined into a trilayer element, in which two ferromagnetic layers are separated by a non-magnetic spacer (Fig. 2b,c). For such a vortex pair there may be four non-degenerate configurations with respect to the relative circulations and polarities^{35–36}.

We used vortex pairs with opposite circulations $c_1 c_2 = -1$ and parallel cores $p_1 p_2 = +1$ as emitters of propagating spin waves. In our first experimental sample (sample 1), the vortex pair was confined into a square-shaped element patterned with a lateral size of 4 μm , made of a Co/Ru/Ni₈₁Fe₁₉ (47.8/0.8/42.8 nm) trilayer stack. We chose two different ferromagnetic materials (Co and NiFe) to allow for layer-selective X-ray measurements. The Ru interlayer supports strong antiferromagnetic interlayer exchange coupling between these two ferromagnetic layers³⁷, which makes opposite vortex circulations energetically favourable³⁵. The pair configuration was verified by direct imaging of the magnetic orientation $\mathbf{m} = \mathbf{M}/M$ using scanning transmission X-ray microscopy^{38,39}. Figure 2 shows the planar circulations (Fig. 2d) and perpendicular cores (Fig. 2e) of both layers (Co and NiFe). Note that, instead of a continuous curl, square-shaped elements exhibit a discrete circulation via four uniform domains that are

intersected by 90° domain walls. From a topological point of view, however, both types of vortices are equivalent.

The vortex pair in sample 1 was excited by an in-plane sinusoidal magnetic field $H_y = H_0 \sin(\omega t)$. The excitation frequency $f = \omega/2\pi$ was varied between 1 and 4 GHz with amplitudes of the order of $\mu_0 H_0 \approx 1$ mT. Within this interval of driving frequencies it was possible to excite an ‘anti-phase’ gyration of the vortex cores^{36,40}, for which the eigenfrequency is strongly dependent on the inter-core distance^{40,41}.

We used time-resolved scanning transmission X-ray microscopy to directly image the magnetic response of the system. Figure 3 shows the resulting perpendicular magnetic deviation $\Delta m_z(t)$ from the average state $\langle m_z(t) \rangle_T$ at fixed times. The response to an excitation of $f = 1$ GHz is displayed separately for the NiFe layer (Fig. 3a) and the Co layer (Fig. 3b). Obviously, both layers exhibit a radially coherent wave

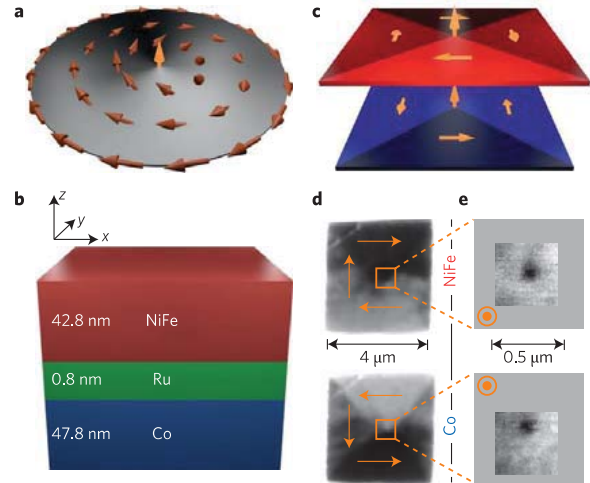


Figure 2 | Sketches and scanning transmission X-ray microscopy images of magnetic vortices. **a**, Disk with continuous planar magnetization circulation ($c = -1$, anticlockwise) and perpendicular core ($p = +1$, upwards along \mathbf{e}_z). **b**, Trilayer stack consisting of Co/Ru/NiFe layers. **c–e**, Vortex pair in a square-shaped trilayer element (sample 1), with discrete antiferromagnetic circulations (m_x -sensitivity; triangular domains) (**d**) and parallel cores (m_z -sensitivity; black dots) (**e**).

pattern with a characteristic wavelength of $\lambda = 365 \pm 20$ nm. Because this pattern is congruent between NiFe and Co ($m_z^{\text{NiFe}}(\mathbf{r}, t) \sim m_z^{\text{Co}}(\mathbf{r}, t)$), it represents a collective spin-wave mode of the trilayer stack.

The underlying wave propagation is depicted by Δm_z line profiles for different excitation times $t = 0, 250$ ps and 500 ps (excitation phases $\sim 0, \pi/2$ and π) in Fig. 3c. Subsequent profiles are shifted by $\lambda/4 \approx 90$ nm, which corresponds to a spin-wave phase shift of $\Phi = \pi/2$. The temporal evolution of the excited

wave pattern is best visualized by means of animated experimental image sequences (Supplementary Movies 1–4). These movies clearly show spin waves that are propagating from the centre to the edge of the sample in a spiralling manner.

Wave patterns corresponding to three different excitation frequencies (1, 2 and 4 GHz) are shown enlarged for the sample centre in Fig. 3d. All three panels show the same kind of spiralling spin-wave emission and propagation, but the resulting wavelength decreases when the excitation frequency is increased

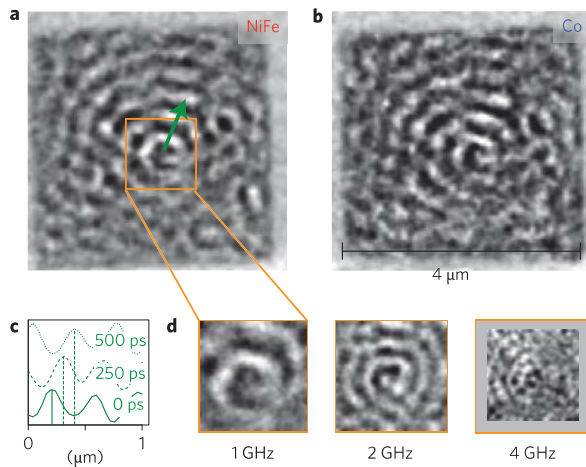


Figure 3 | Time-resolved scanning transmission X-ray microscopy images of spin-wave emission from magnetic vortex cores (sample 1). **a,b**, Normalized m_z contrast for NiFe (**a**) and Co (**b**) under lateral field excitation with $\mu_0 H_0 \approx 1$ mT at 1 GHz. **c**, Radial spin-wave line profile along the green arrow in **a**, for different response times of 0, 250 and 500 ps. **d**, Normalized NiFe m_z contrast of spin-wave emission at 1, 2 and 4 GHz in the central part of the sample, as indicated in **a** by the orange box.

(Supplementary Movies 5–10). Starting from $\lambda = 365 \pm 20$ nm at $f = 1$ GHz, a decrease to $\lambda = 200 \pm 20$ nm at $f = 2$ GHz and eventually to $\lambda = 125 \pm 10$ nm at $f = 4$ GHz is observed. We estimate the corresponding phase velocities $V(f)$ by $V = \omega/k$, with wavenumber $k = 2\pi/\lambda$, to be $V(1 \text{ GHz}) = 365 \text{ ms}^{-1}$, $V(2 \text{ GHz}) = 400 \text{ ms}^{-1}$ and $V(4 \text{ GHz}) = 500 \text{ ms}^{-1}$. The experiments clearly show that the wave-length of the excited spin-wave mode scales almost linearly with the excitation frequency. Therefore, stacked pairs of magnetic vortices confined into trilayer elements can be used as tunable emitters of propagating spin waves with nanoscale wavelengths.

Collective spin-wave dispersion relation

To obtain a clear understanding of the spin waves observed, we investigated their properties by means of analytic calculations. Our experiments show that we are looking at a layer-collective spin-wave mode propagating

perpendicularly to the circulating magnetization of an anti-ferromagnetically coupled vortex pair. This situation is similar to the propagation of a quasi-Damon–Eshbach mode⁴² in a trilayer where two ferromagnetic films,

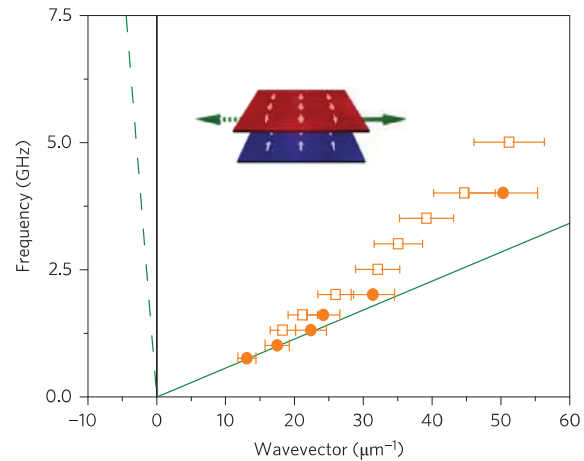


Figure 4 | Dispersion $f(k)$ of collective spin waves propagating perpendicularly to the direction of static magnetization in a trilayer where two in-plane ferromagnetic layers are antiferromagnetically coupled via a non-magnetic interlayer. Solid and dashed green lines correspond to the calculated dispersions for the ‘slow’ and ‘fast’ spin-wave branches, respectively. Inset: Schematic showing the directions of static magnetization in the two ferromagnetic layers (white arrows) and the propagation direction for the ‘slow’ and ‘fast’ spin-wave branches (solid and dashed green arrows, respectively). Experimental results obtained by exciting vortex pair sample 1 with laterally oscillating magnetic fields are shown by filled orange circles, and the results of micromagnetic modelling are shown by open orange squares. Error bars indicate the uncertainty of wavelength determination.

separated by a non-magnetic spacer, are magnetized into opposite in-plane directions. The dispersion of such magnetostatic waves (neglecting exchange) was calculated in the pioneering work of Grünberg⁴³. In our case, however, the exchange interaction cannot be ignored and thus we had to generalize the results of propagating perpendicularly to the circulating magnetization of an antiferromagnetically coupled vortex pair. This situation is similar to the propagation of a quasi-Damon–Eshbach mode⁴² in a trilayer where two ferromagnetic films, separated by a non-magnetic spacer, are magnetized into opposite in-plane directions. The dispersion of such magnetostatic waves (neglecting exchange) was calculated in the pioneering work of Grünberg⁴³. In our case, however, the exchange interaction cannot be ignored and thus we had to generalize the results of ref. 43 for the case of dipole-exchange spin waves. For this purpose, we start from a pair of coupled Landau–Lifshitz equations⁴⁴ for the

magnetizations \mathbf{M}_j ($i, j = 1, 2$) of the two ferromagnetic layers:

$$\frac{\partial \mathbf{M}_j}{\partial t} = \gamma [\mathbf{B}_{ji} \times \mathbf{M}_j] \quad (1)$$

where γ is the modulus of the gyromagnetic ratio and \mathbf{B}_{ji} is the effective magnetic field in the j th layer, which is assumed to contain only inhomogeneous exchange ($\propto \nabla^2 \mathbf{M}_j$) and magneto-dipolar ($\mu_0 \mathbf{H}_{ji}$) contributions:

$$\mathbf{B}_{ji} = \frac{2A}{M_s^2} \nabla^2 \mathbf{M}_j + \mu_0 \mathbf{H}_{ji} . \quad (2)$$

Here A is the exchange constant, M_s is the saturation magnetization, μ_0 is the permeability in vacuum, and \mathbf{H}_{ji} is the dipolar magnetic field that couples the magnetizations of the two ferromagnetic layers. It can be shown that the antiferromagnetic interlayer exchange

$$V(\phi_k) = \omega_M d \left[\sqrt{\frac{\lambda_{ex}^2}{d^2} + \frac{1}{12} \left[4 + 6 \frac{d_s}{d} - F \left(\frac{\lambda_{ex}}{d} \right) \right]} \sin^2 \phi_k - \frac{1}{2} \sin \phi_k \right] . \quad (4)$$

Here $\omega_M = \gamma \mu_0 M_s$, $\lambda_{ex}^2 = 2A/\mu_0 M_s^2$ (exchange length), d is the thickness of the ferromagnetic films, d_s is the separation between the films, and the function $F(u)$ is given by

$$F(u) = 1 - 12u^2 + 24u^3 \tanh \left(\frac{1}{2u} \right) . \quad (5)$$

The fact that the spin-wave dispersion relation (3) depends on the sign of $\sin \Phi_k$ means that this spin-wave mode is non-reciprocal; that is, the velocities of spin waves propagating in opposite directions are substantially different. In our experimental conditions, when $d_s \ll d$ and $\lambda_{ex}^2 \ll d^2$, expression (4) can be simplified to

$$V(\phi_k) \cong \omega_M d \left[\frac{1}{\sqrt{3}} + \frac{1}{2} \text{sign}(\sin \phi_k) \right] |\sin \phi_k| . \quad (6)$$

Thus, in our case of $\mathbf{k} \perp \mathbf{M}$, the velocities of spin waves propagating in opposite directions are much different— $V(\Phi_k = +\pi/2)/V(\Phi_k = -\pi/2) \approx 14$.

Using equation (6), one can estimate the velocity of the ‘slow’ spin-wave branch $V(\Phi_k = -\pi/2)$ for the conditions of our experiment, assuming $\omega_M d \approx (\omega_{M1} d_1 + \omega_{M2} d_2)/2$ to $V(\Phi_k = -\pi/2) \approx 360 \text{ ms}^{-1}$, which is rather close to the measured value at small wavenumbers. The wavelength of the ‘fast’ spin-wave branch with a velocity of $V(\Phi_k = +\pi/2) \approx 5,000 \text{ ms}^{-1}$, however, will be of the order of the lateral size of our sample, which would make it difficult to observe the ‘fast’ branch in our experiment. In particular, the

coupling between the ferromagnetic layers – which is ignored in equation (2) – does not noticeably change the dispersion of the considered spin-wave mode. A detailed analytical solution of the problem formulated by equations (1) and (2) is presented in Supplementary Section 7.

For the model case of identical ferromagnetic layers in the long-wavelength limit (when $|\mathbf{k}| = k \rightarrow 0$), we found that the dipole-exchange spectrum of an antiparallel bilayer contains a spin-wave mode with linear dispersion and no frequency gap:

$$\omega(\mathbf{k}) = V(\phi_k) k , \quad (3)$$

where Φ_k is the angle between the spin-wave wavevector \mathbf{k} and the in-plane direction of the static magnetization in the ferromagnetic layers (in our case $\Phi_k = \pm\pi/2$), while the velocity $V(\Phi_k)$ of the spin-wave mode is given by:

formation of standing wave patterns involving counter-propagating ‘slow’ and ‘fast’ wave branches is practically suppressed. For this reason, and because of the smaller amplitudes expected for the ‘fast’ branch (Supplementary Section 4), only the ‘slow’ spin-wave branch was observed in our experiment, as shown in Fig. 3. The corresponding experimental dispersion relation $f(k)$ is plotted in Fig. 4 as filled orange dots. In addition, the dispersion curves of the ‘slow’ and ‘fast’ spin waves, calculated using equation (6) are shown as solid and dashed green lines, respectively.

From Fig. 4 it is clear that the ‘slow’ spin-wave branch calculated from our planar symmetric model system in equation (6) essentially describes the experimentally

observed dispersion of a propagating mode in the vortex pair state with opposite circulations and asymmetric layers. Only for higher wavenumbers do the experimental frequencies of the ‘slow’ branch start to deviate from the theoretical line. This deviation can be attributed to an increasing effect of the exchange interaction, which presumably will eventually lead to a k -quadratic dispersion relation for high wavenumbers⁴⁴.

Spin-wave generation mechanism

To reveal the origin of the observed spin-wave generation process, we carried out micromagnetic simulations for a vortex-state magnetic trilayer⁴⁵. The results of these simulations, shown for the ‘slow’ spin-wave branch by open orange squares in Fig. 4, are in excellent agreement with the experimental data. The spiralling spin-wave emission patterns obtained from these simulations (Supplementary Section 5) are also very similar to the experimentally measured spin-wave profiles.

It also follows that the origin of the observed spin-wave emission is a field-driven, small-amplitude vortex core gyration around the equilibrium position of the parallel cores. This core gyration is accompanied by the formation of a similarly gyrating perpendicular magnetization ‘dip’ of opposite sign that has already been observed for single-layer vortices⁴⁶ (Supplementary Section 5). Cores and ‘dips’ then act as perpendicular magnetic perturbations travelling laterally on the gyration orbit. These perturbations thus give rise to a local m_z -collective magnetization precession, that is, a source for spin waves, with a continuous azimuthal phase shift along the gyration orbit. The azimuthal precession phase shift essentially leads to the formation of a spiralling wave pattern instead of a concentric one. In a certain way, this spin-wave excitation mechanism can also be seen as an analogue to an orbiting stirrer in a fluid system, which leads to a similar pattern of spiralling wave propagation. Note that the mechanism of coherent spin-wave generation identified here is fundamentally different from that of the incoherent spin-wave emission predicted for dynamic vortex core switching^{25,47,48}.

Non-reciprocal spin-wave propagation

As follows from both analytical theory and from our micromagnetic simulations, the spin-wave mode observed in our experiments belongs to the ‘slow’ branch of the dipole-exchange dispersion relation (3). For the vortex pair shown in Fig. 2d ($c_{1(\text{Co})} = +1$, $c_{2(\text{NiFe})} = -1$), this ‘slow’ branch corresponds to outwards-propagating spin waves. Obviously, when both circulations are reversed ($c_{1(\text{Co})} = -1$, $c_{2(\text{NiFe})} = +1$), the ‘slow’ branch should also experience a reversal of its propagation direction and should therefore become inwards-propagating. Note that this effect is independent of the mean circulation of the vortex pair given by the higher magnetic moment of the Co layer, and only depends on the handedness of the pair circulations (Supplementary Section 4).

To evaluate this property experimentally, we measured a further trilayer sample containing a vortex pair with $c_{1(\text{Co})} = -1$ and $c_{2(\text{NiFe})} = +1$ (sample 2). Sample 2 is a circular Co/Ru/Ni₈₁Fe₁₉ (47.8/0.8/42.8 nm) disk with a diameter of 2 μm (Supplementary Section 2). Given this sample, we were able to compare the excitation of spin waves in vortex pairs with $c_{1(\text{Co})} = +1$, $c_{2(\text{NiFe})} = -1$ (sample 1) and $c_{1(\text{Co})} = -1$, $c_{2(\text{NiFe})} = +1$ (sample 2) using time-resolved scanning transmission X-ray microscopy. To exclude any stroboscopic artefacts, the samples were now driven by in-plane magnetic field pulses ($\mu_0 H_y \approx 1$ mT) of 500 ps length at a repetition rate of 5 ns. The results from this experiment are presented in Fig. 5, where the image contrast corresponds to the perpendicular magnetic deviation $\Delta m_z(t)$ in the respective NiFe layers of sample 1 (left image series) and sample 2 (right image series). Each series of four images spans an observation time of $\Delta t = 1.43$ ns, which covers the significant temporal changes in the spin-wave patterns observed (see Supplementary Movies 11–16 for the full 5 ns interval).

In sample 1, following the application of the driving pulse, a spin-wave train is formed at the centre of the sample, which then successively propagates outwards to the sample periphery, eventually reaching the outer edge of the field of view after ~ 1.4 ns. This pulse-driven spin-wave generation and propagation is similar to that of spin waves excited by a sinusoidal driving signal (as shown in Fig. 3).

The spin-wave propagation following pulsed excitation in sample 2 is crucially different. Here, the spin-wave train first appears at the outer edge of the field of view, propagates inwards, and eventually disappears at the centre of the sample. This behaviour confirms our prediction of two strongly non-reciprocal spin-wave branches (‘slow’ and ‘fast’). The rotating vortex cores can only excite spin waves that propagate outwards from the centre. For sample 2, such outward-propagating spin waves correspond to the ‘fast’ branch, which cannot be directly observed in the experiment. However, once the ‘fast’ spin waves have reached the outer edge of the sample, they are reflected backwards, transforming into the observable inward-propagating ‘slow’ branch. Because there is only a very short delay (< 280 ps) between the application of the driving pulse and the appearance of the observable spin waves at the outer edge of the sample, we conclude from this experiment that the ‘fast’ spin-wave branch is propagating much faster (at least ten times) than the ‘slow’ spin-wave branch. Complementary micromagnetic simulations

(Supplementary Movie 19) were used to directly estimate the group velocity of the ‘fast’ branch as $\sim 4 \times 10^3 \text{ ms}^{-1}$, which is close to the calculated value of $\sim 5 \times 10^3 \text{ ms}^{-1}$. These findings therefore substantially support our theoretical prediction of the strongly non-reciprocal

character of spin-wave propagation in antiferromagnetically coupled trilayers.

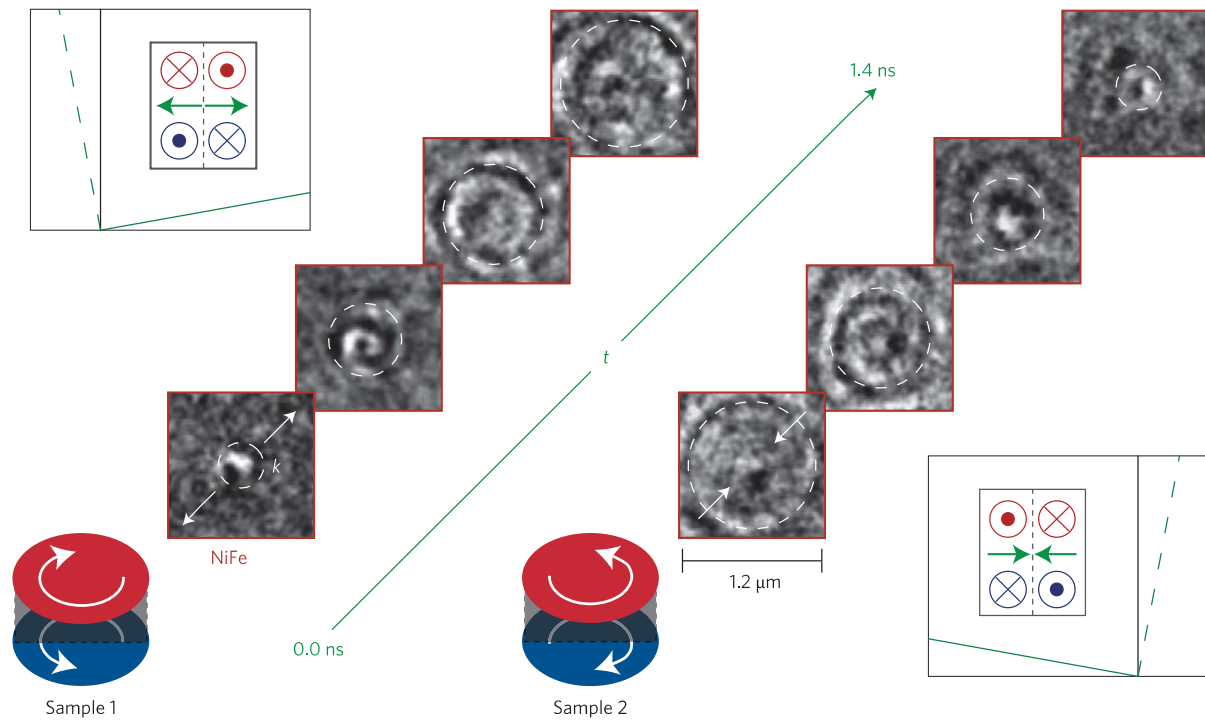


Figure 5 | Pulsed excitation of spin waves in the two samples containing magnetic vortex pairs with opposite circulations. The left series of images corresponds to sample 1 ($c_{1(\text{Co})} = +1$, $c_{2(\text{NiFe})} = -1$), supporting an outwards-propagating ‘slow’ spin-wave branch, and the right series corresponds to sample 2 ($c_{1(\text{Co})} = -1$, $c_{2(\text{NiFe})} = +1$), supporting an inwards-propagating ‘slow’ spin-wave branch. The image series consists of time-resolved scanning transmission X-ray microscopy images showing the Δm_z contrast of the NiFe layer for different delay times. The phase fronts of excited spin-wave trains are highlighted by dashed white circles. The vortex circulations in the two ferromagnetic layers (blue, Co; red, NiFe) are indicated by white arrows in the adjacent schematics. The upper left and lower right insets show the dispersion curves for the ‘slow’ (solid green lines) and ‘fast’ (dashed green lines) spin-wave branches in samples 1 and 2, respectively. In these insets, green arrows indicate the propagation directions of the ‘slow’ spin-wave branch.

Conclusions

We have found a highly agile mechanism for the coherent generation of propagating spin waves, whose nanoscale wavelengths are linearly tunable by the frequency of the driving signal. This generation mechanism overcomes the bandwidth limitations of conventional spin-wave generation methods, which are determined by the patterning size. In our case, spin-wave excitation stems from the driven gyration of naturally formed magnetic vortex cores that have sizes on the order of the exchange length of the sample material. We used time-resolved scanning transmission X-ray microscopy to directly image such propagating spin waves with wavelengths down to 100 nm and frequencies up to $\sim 4 \text{ GHz}$. We

found that – at relatively small wavenumbers – the excited spin-wave mode has a linear dispersion without a frequency gap. Moreover, this mode is strongly non-reciprocal; that is, waves travelling in opposite directions exhibit propagation velocities that are more than one order of magnitude different. Apart from the fundamental importance of the phenomena observed, we believe that the obtained results will open a way for the development of new and efficient bias-free non-reciprocal microwave signal-processing devices, such as isolators, circulators, unidirectional conduits, phase shifters and controlled switches.

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Author contributions

S. W., M. W. and J. R. performed the experiments. S. W. fabricated the samples and performed the simulations. V. T. and A. S. calculated the dispersion relations. S. W. and A. S. wrote the manuscript. All authors discussed the data and commented on the manuscript.

Additional information

Supplementary information is available in the online version of the paper. Reprints and permissions information is available online at www.nature.com/reprints. Correspondence and requests for materials should be addressed to S.W.

Competing financial interests

The authors declare no competing financial interests.

Methods

Sample fabrication. The samples were fabricated on X-ray-transparent silicon-nitride membrane substrates with a thickness of 100 nm. The patterning of microelements was achieved by a sequence of electron-beam lithography, multilayer deposition and liftoff processing. A positive poly(methyl methacrylate) (PMMA) resist was first spun onto the substrate. In the second step, circular and square-shaped elements with base sizes between 2 and 4 μm were exposed by means of electron-beam lithography and then subsequently developed for 30 s in a methyl isobutyl ketone:isopropyl alcohol (MIBK:IPA) mixture (1:3 volume ratio). A Co/Ru/Ni₈₁Fe₁₉/Al multilayer was then deposited by magnetron sputter deposition, with Al serving as a capping layer for oxidation protection. The resulting layer thicknesses were determined by transmission electron microscopy (TEM) to be 47.8, 0.8, 42.8 and 5 nm (Supplementary Section 1). For the chosen Ru thickness, antiferromagnetic interlayer exchange coupling is expected to occur between the two ferromagnetic layers; that is, antiparallel magnetic alignment between the two layers is energetically preferred. This coupling was quantitatively estimated by fitting an extended Stoner-Wolfarth model on the magnetization reversal signal measured by magneto-optic Kerr effect (Supplementary Section 1), resulting in a bilinear coupling constant of $J_L = -0.35 \text{ mJ m}^{-2}$ and biquadratic coupling constant of $J_Q = -0.07 \text{ mJ m}^{-2}$. This type of coupling leads to the formation of spin vortex pairs with antiparallel circulations in microelements of the given sizes and materials. Oscillating magnetic fields can be applied to the sample via electric currents flowing through a copper strip fabricated on top of the elements. The patterning of the strip was carried out analogously to the microelements, but a bilayer copolymer/polymer resist was used instead and the development time was increased to 120 s. The copper thin film was deposited with a nominal thickness of 200 nm by means of evaporation, together with a 5-nm-thick aluminium cap layer. The patterned microstrip had a width of 5 μm . Based on this, the resulting magnetic field was estimated to be $\mu_0 H = 4\pi \times 10^{-2} \text{ mT mA}^{-1}$.

Time-resolved scanning transmission X-ray microscopy. The magnetization distribution of the vortex pair structure was imaged by means of synchrotron-based scanning transmission X-ray microscopy^{38,39}. This method uses a Fresnel zone plate to focus a monochromatic X-ray beam onto the sample. The sample was then raster-scanned through this beam, and the transmitted intensity, collected by a point detector, yielded a two-dimensional image with $\sim 25 \text{ nm}$ lateral resolution. By the application of circularly polarized photons, X-ray magnetic circular dichroism (XMCD)⁴⁹ was used to

acquire a magnetic signal. Because XMCD is only relevant at the resonant absorption edges of a specific element, the two ferromagnetic layers of the Co/Ru/NiFe stack could be separately addressed by tuning the incident X-ray energy to different absorption edges, in this case the Ni L₃ edge ($E \approx 853 \text{ eV}$) and the Co L₃ edge ($E \approx 778 \text{ eV}$), respectively. The achieved magnetic contrast is proportional to the projection of the magnetic orientation $\mathbf{m} = \mathbf{M}/M$ onto the photon propagation direction \mathbf{e}_k . Therefore, a normal incidence set-up is sensitive to perpendicular magnetization components, whereas additional in-plane sensitivity can be gathered when the sample normal is inclined versus \mathbf{e}_k . The dynamic response of the vortex pair to an alternating magnetic field excitation was stroboscopically imaged by means of time-resolved scanning transmission X-ray microscopy. This method employs the specific time structure of the incoming synchrotron radiation, that is, X-ray flashes of an approximate temporal width of 100 ps at a 2 ns repetition rate. The transmitted intensity is on the order of a single photon per flash and is recorded by a fast avalanche photodiode. The result of each probing event (photon or no photon transmitted) is routed to a specific counting register of a periodic register set by a field-programmable gate array. Here, the integer total amount of registers has to be commensurable with the single-cycle (non-stroboscopic) measurement duration divided by 2 ns. Furthermore, the electronic excitation sources are required to be synchronized to the repetition rate of the X-ray flashes. The excitation signal (current in the stripline) is evaluated before and after the sample by means of two $\sim 20 \text{ dB}$ pick-off tees that provide pick-off signals to an oscilloscope.

Micromagnetic simulations. Micromagnetic simulations based on time integration of the Landau–Lifshitz–Gilbert equation⁵⁰ were carried out using the ‘LLG Micromagnetic Simulator’ environment⁴⁵. At first, the initial state of a Co/NiFe vortex pair with antiparallel circulations and parallel cores was simulated using an artificially high Gilbert damping constant of $\alpha = 1$, allowing for relatively fast relaxation of the system. Once the residual relative torque decreased below 1×10^{-6} in all simulation cells, the system was assumed to have reached its equilibrium state. The time evolution of the system was simulated with a damping constant of $\alpha = 0.01$ following excitation by a laterally alternating sinusoidal magnetic field. The simulated material parameters were set to the following values: $M_s^{\text{Co}} = 1,273 \text{ kAm}^{-1}$, $M_s^{\text{NiFe}} = 740 \text{ kAm}^{-1}$, $d^{\text{Co}} = 47.8 \text{ nm}$ and $d^{\text{NiFe}} = 42.8 \text{ nm}$ as measured by a superconducting quantum interference device (SQUID) and TEM³⁵, and $A^{\text{Co}} = 3.05 \times 10^{-11} \text{ Jm}^{-1}$, $A^{\text{NiFe}} = 1.05 \times 10^{-11} \text{ Jm}^{-1}$ and $d^{\text{IL}} = 0.8 \text{ nm}$, as taken from refs 35 and 45. For simplicity, we neglected the small intrinsic anisotropies of the ferromagnetic layers and we assumed that the interlayer possesses vacuum permeability. The interlayer exchange coupling constants were set to $J_L = -0.35 \text{ mJ m}^{-1}$ and $J_Q = -0.07 \text{ mJ m}^{-2}$ (ref. 35). The underlying simulation body was a square-shaped element with an edge length of 1 μm that was discretized into 128×128 uniform orthogonal cells, corresponding to lateral cell dimensions of approximately $(7.8 \text{ nm})^2$. For symmetry reasons, this structure was reduced to a circular element with a diameter of 1 μm . Vertically, a non-uniform discretization was chosen, attributing five cells each to the ferromagnetic layers (9.56 nm, Co; 8.56 nm, NiFe) and a single cell (0.8 nm) for the interlayer, respectively.

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Magnetic domain walls as reconfigurable spin-wave nano-channels

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In the research field of magnonics^{1–7}, spin waves are envisioned to be used as information carriers promoting operations based on their wave properties. However, the field still faces major challenges: To become fully competitive, novel schemes for an energy-efficient control of spin-wave propagation in 2D have to be realised on much smaller length scales than used before. In this letter, we address these challenges by the experimental realisation of a novel approach to guide spin waves in reconfigurable, nano-sized magnonic waveguides. For this purpose, we make use of two inherent characteristics of magnetism: the nonvolatility of magnetic remanence states and the nanometre dimensions of domain walls formed within these magnetic landscapes. We present the experimental observation and micromagnetic simulations of spin-wave propagation *inside* nano-sized domain walls and realise a first step towards a reconfigurable domain-wall based magnonic nano-circuitry.

Spin waves are the dynamic eigen-excitations of a magnetically ordered medium with their quanta being referred to as magnons. In the last years, the scientific community in the field of magnonics made huge efforts to realise concepts to use these spin waves for data processing. Quite recently, remarkable progress has been made by the demonstration of prototype building blocks of a future spin-wave based logic^{8–10}. Despite this progress, most concepts are based on very similar sample layouts that are limited regarding the further optimisation of existing devices. Previous studies on spin-wave propagation on the nanoscale relied on confinement in geometrically patterned waveguides¹¹ that lack any flexibility for manipulating the spin-wave propagation path as required for flexible, reprogrammable devices. On the microscale, this dynamic control of spin waves in two-dimensional structures has been realised but is based on the continuous application of external fields or currents and has been achieved at the cost of very high energy consumption. In the following, we will present a possible way to overcome these challenges.

Spin waves impinging on a domain wall are either reflected or experience a phase shift when transmitted, depending on the spin-wave wavelength and the type of domain wall^{12–16}. Additional studies were devoted to the investigation of the domain dynamics itself that were found in the very low MHz frequency range on the order of 10 MHz¹⁷. However, the possibility to send spin waves *along* a domain wall has only recently been ad-

ressed in a numerical study for a material with out-of-plane magnetisation that is difficult to handle in experimental scenarios¹⁸. Here, we experimentally explore the intrinsic spin-wave eigenmodes that are quantised across the width of a domain wall and yet possess a well-defined wave vector along the wall that enable information transport and processing. Targeting this class of still widely unknown spin-wave modes, we focus on the potential of utilising domain walls as nanometre-scaled spin-wave channels that open new perspectives for the energy-efficient control of spin-wave propagation in 2D.

The top panel in Fig. 1a shows a schematic illustration of a 180° Néel wall. The magnetic moments rotate within the sample plane, giving rise to magnetic volume charges – defined as the divergence of the magnetisation $\nabla \cdot \mathbf{M}$ – with opposite signs on the two sides of the domain-wall centre. The red curve in Fig. 1a illustrates these volume charges calculated via micromagnetic simulations. The charges generate a strong magnetostatic field $\mathbf{H}_{\text{demag}}$ (blue curve in Fig. 1a) oriented antiparallel to the magnetisation direction in the centre of the domain wall resulting in a locally decreased effective magnetic field. Since the spin-wave dispersion depends on this effective field, field gradients form potential wells for spin waves^{19,20}. In the actual case, a potential well for spin waves is created across the domain wall with a width of only a few ten nanometres. The exact width of a domain wall strongly depends on various parameters of magnetic materials and can be tuned towards even smaller sizes. In this letter, we demonstrate experimentally and numerically that such a potential well leads to spin-wave modes which are strongly localised to the domain-wall width and yet travel freely along the wall. Moreover, and in contrast to the vast majority of experimentally realised scenarios, no energy-consuming external bias fields are needed to promote spin-wave propagation in the preferred geometry¹⁰ along the domain wall within the magnetic remanence state.

Figure 1b shows a scanning electron micrograph of the structure that was used in the experiment. A 40-nm-thick $\text{Ni}_{81}\text{Fe}_{19}$ (Permalloy, Py) film is patterned into a spin-wave waveguide that is 5- μm wide at the top end, then gradually broadens until it reaches a constant width of 10 μm . The variable width stabilises the desired remanence state. A gold microwave antenna is positioned at the beginning of the 10- μm wide part of the waveguide. Oscillating magnetic fields h_{rf} generated by the application of microwave currents to this antenna allow for the local excitation of spin waves with well defined frequencies.

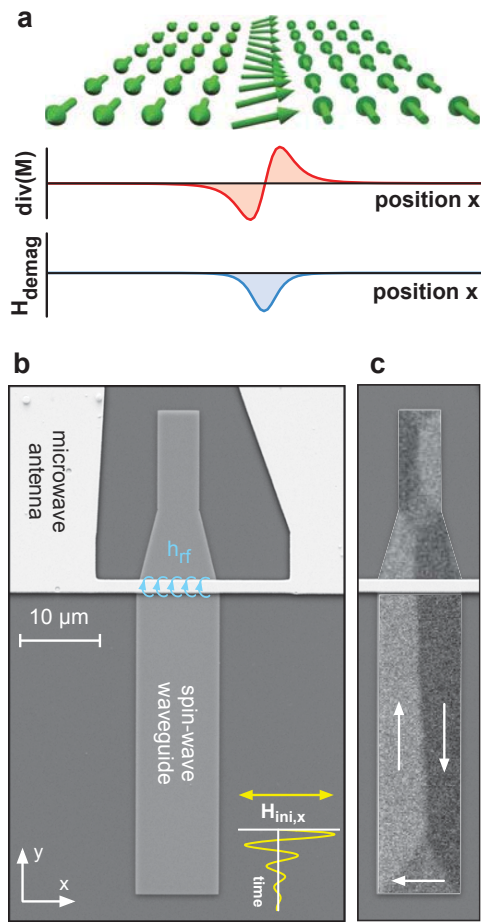


FIG. 1. Channelling principle, sample geometry and magnetisation configuration. **a**, Schematic illustration of a 180° Néel wall carrying a spin wave. The divergence of the magnetisation $\nabla \cdot \mathbf{M}$ across the width of the wall shows opposite sign on the two sides of the domain wall centre and results in a strong magnetostatic field $\mathbf{H}_{\text{demag}}$ oriented antiparallel to the magnetisation direction. This potential well confines the spin-wave propagation into the narrow region of the wall. **b**, Scanning electron micrograph of the investigated Py structure and the microwave antenna for the external excitation of spin dynamics. The inset on the lower right shows the magnetic field sweep that is used to initialize the domain configuration. **c**, The black-white contrast of the Kerr-microscopy image confirms the formation of a Landau-like domain pattern. The arrows indicate the magnetisation direction in the domains.

In order to reproducibly prepare the aforementioned remanence state with the domain wall parallel to the long axis of the waveguide, we applied a sinusoidal, exponentially decaying magnetic field $H_{\text{ini},x}$ parallel to the short axis of the waveguide as depicted by the inset in Fig. 1b. We used magneto-optical Kerr microscopy to confirm the magnetic state of the waveguide. The overlay in Fig. 1c shows an exemplary Kerr image with the black-white

colour code representing the magnetisation component along the y -direction. A Landau-like domain pattern is formed with a 180° Néel wall in the centre of the waveguide separating two domains with opposite magnetisation. The arrows indicate the magnetisation direction in the domains. Additional micromagnetic simulations (discussed below) confirm the Néel-type character of the domain wall with the magnetic moments rotating in the sample plane as is expected for the material parameters and thickness of the Py film²¹.

Experimentally, the propagation of spin waves was studied by micro-focus Brillouin light scattering (BLS) spectroscopy²². To analyse the spectrum in the different areas of the domain pattern, we measured the spin-wave intensity as a function of the excitation frequency at two different positions: inside the domain wall and in the domains. These measurements were performed in the 10- μm wide bottom part of the waveguide at a distance of 1- μm from the antenna. Figure 2a summarizes the results of the measurements and clearly shows two distinct spectra depending on the probing position: inside the domain wall (squares), the maximum spin-wave intensity was observed at the lowest possible detection frequency of 500 MHz whereas in the domains (triangles) highest intensities can be found at much higher frequencies around 2.8 GHz. The existence of a strongly localised mode inside the domain wall is a remarkable observation supporting our assumption of spin waves trapped in a potential well.

To analyse the nature of the different spin-wave modes within the wall and the domains, we detected their intensity profile across the width of the waveguide. These measurements were carried out at the excitation frequencies where maximum intensity was observed within the wall and the domain. Fig. 2b illustrates the results of these line scans. The detected modes show a clear spatial separation with one mode strongly confined in the centre of the waveguide at the position of the domain wall (squares). In contrast, the higher-frequency mode (triangles) is spread throughout the domains at both sides of the wall whereas it almost vanishes at the domain-wall position. Thus, the potential well formed by the domain wall can be used to confine spin waves in a certain frequency range on the nanoscale.

The slightly different intensity profiles detected at 2.8 GHz in the two domains are attributed to the fact that the domain wall is not perfectly centred in the waveguide. This asymmetry leads to different lateral confinement conditions in the two domains and, consequently, to a relative shift of the corresponding excitation spectra. However, quantisation due to confinement in micron-sized waveguide structures has already been studied extensively^{22–24}.

Here, we focus on the spin-wave mode inside the nanochannel formed by the domain wall that can be selectively excited by choosing the proper microwave frequency. The localisation becomes even more evident from the two-dimensional BLS measurement presented

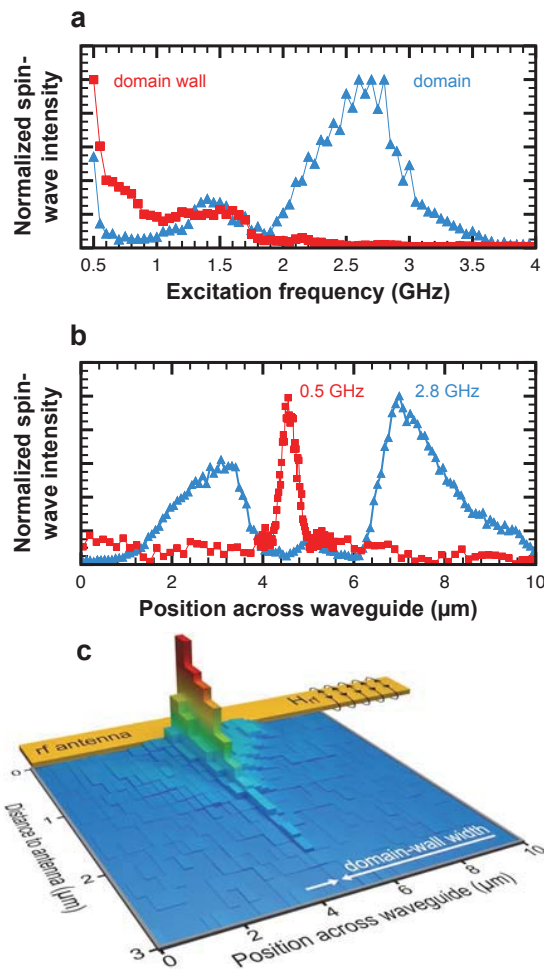


FIG. 2. Excitation spectra and spin-wave mode profiles. **a**, Spin-wave spectrum measured by BLS microscopy inside the domain wall (squares) and the domains (triangles), respectively. The measurement clearly shows different excitation spectra depending on the probing position. **b**, Line scans of the spin-wave intensity across the width of the Py waveguide for the two excitation frequencies that yielded maximum spin-wave intensity inside the domain wall (squares) and in the domain (triangles). The data exhibit two spatially separated spin-wave modes with the low-frequency mode strongly localised at the domain wall position. **c**, Two-dimensional intensity distribution of spin waves propagating along the nano-channel formed by the domain wall in the centre of the waveguide.

in Fig. 2c. Solely at the position of the domain wall, the signal of a spin-wave mode propagating away from the antenna is detected revealing the channelling character of the nano-sized domain wall.

To get a deeper insight in the propagation characteristics within the domain wall, micromagnetic simulations were carried out²⁵. The studied system was a 5-μm long, 1-μm wide and 10-nm thick Py rectangle and,

thus, smaller than the experimentally observed structure. This reduction of size allowed for a sufficiently fine discretisation required to simulate the nano-sized domain wall without affecting its character which is mainly determined by the material parameters and not by the different dimensions. Therefore, a direct comparison between experiment and simulation is still possible for the spin-wave modes within the domain wall. Figure 3a shows the static equilibrium magnetisation configuration. The red-blue colour codes the in-plane magnetisation component m_x parallel to the short axis of the rectangle and the arrows display the magnetisation direction. The microstructure exhibits a flux-closure Landau-like domain pattern with a 180° Néel wall separating two large domains with opposite magnetisation.

The magnitude of the effective magnetic field calculated from this remanence state is presented in Fig. 3b with the red-blue colour representing the in-plane field component H_x along the short axis of the rectangle. The data clearly show strong effective fields along the domain walls that are oriented anti-parallel to the magnetisation direction and, thus, form potential wells for spin waves.

Starting from the Landau-type magnetisation configuration, an out-of-plane field pulse is applied locally at the domain-wall position at 1.6-μm distance from the bottom edge to excite spin dynamics (see green dot in Fig. 3c). This pulsed excitation and subsequent analysis as discussed in the method section yields the spin-wave spectrum and dispersion relation, which is discussed in the next paragraph. For displaying the mode profiles of the spin-wave resonances we simulated the response to a continuous microwave excitation at four different frequencies. In Fig. 3c the normalized z-component of the magnetization (m_z) is plotted for a given time once the system reached a steady state. The time evolution of the spin-wave amplitudes showing their propagating nature are available as supplementary material for two distinct frequencies. For the lower frequencies of 0.52 GHz, 1.28 GHz and 2.16 GHz, the spin-wave modes are strongly localized inside the domain wall. However, a general trend in Fig. 3c is that the strong localisation within the domain wall gets weaker for increasing frequency. For modes with high frequencies, *e.g.* 5.68 GHz, the clearly visible radiation from the wall indicates the onset of the spin-wave band in the domains and, thus, the loss of the channelling effect. While the strong localisation of the low-frequency spin waves inside the domain wall is in good accordance to the experiment, the domain modes appear at higher frequencies in the simulation. In general, the spin-wave eigenmodes shift to higher frequencies when reducing the domain size which, therefore, allows guiding of spin waves inside the wall even for higher frequencies and smaller wavelength.

In addition to the spatial and spectral characteristics of the investigated spin-wave modes, the micromagnetic simulations also shed light on the spin-wave dispersion. The spin-wave wavelength for a given frequency - as illustrated by the green bars in Fig. 3c - can be determined

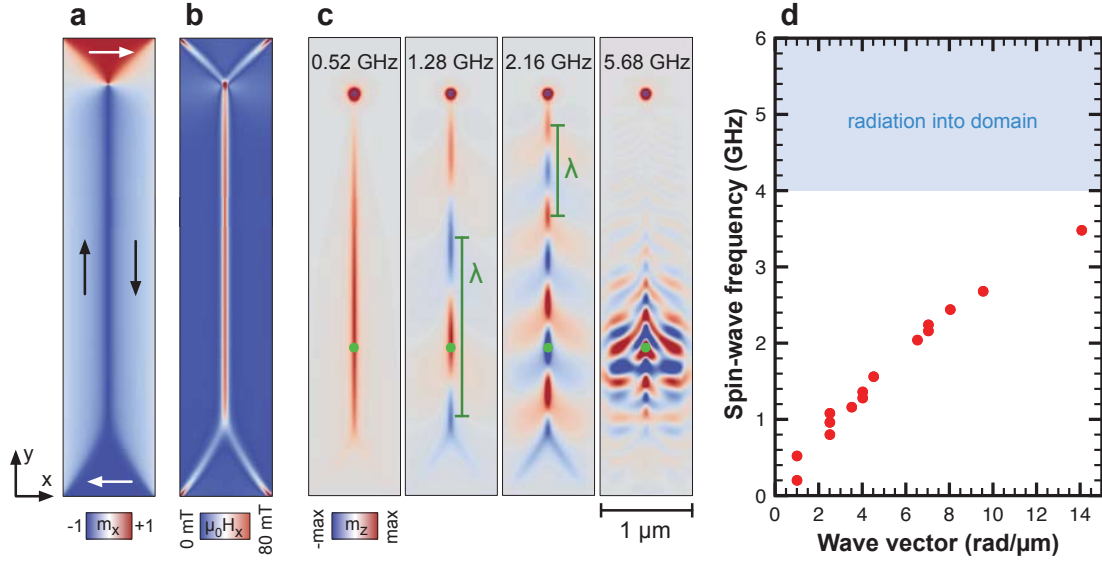


FIG. 3. **Magnetisation, effective magnetic field, spin-wave modes and dispersion.** **a**, Simulated domain configuration of the rectangular Py thin-film element. The red-blue colour represents the in-plane magnetisation component m_x parallel to the short axis of the rectangle, the arrows display the net magnetisation direction in the domains. **b**, x -component of the resulting effective field. The image clearly shows that strong fields are present along the domain walls. **c**, Amplitude profiles of different spin-wave modes excited locally at the position indicated by the green dot. The red-blue colour represents the out-of-plane component of the magnetisation m_z . The green bars indicate the spin-wave wavelengths that can be extracted from the data. **d**, Dispersion relation for spin waves mostly confined to the domain wall.

by a Fourier analysis of the dynamic magnetisation along the domain wall. Figure 3d shows the resulting dispersion relation and reveals a positive dispersion that enables the transport of information via spin waves propagating within the domain wall. **Even though the spin waves are confined transverse to the wall on a length scale given by the domain wall width, the dispersion is mainly dominated by dipolar energy. For the first three modes shown in Fig. 3c the dynamic part of the dipolar energy originating from the spin waves is by a factor 3-5 larger than the dynamic exchange energy.** The observation of well-defined wave vectors along the propagation path is a crucial pre-condition for numerous concepts that rely on the interference of spin waves^{12,26,27}. Therefore, this observation highlights the potential of utilising domain walls in sophisticated magnonic circuits for information transport and data processing.

To evaluate the localisation of the numerically calculated spin-wave modes, we extract a line profile of the spin-wave intensity excited at 0.52 GHz across the domain wall. The result is shown by the black dashed line in Fig. 4 yielding a signal with a full width at half maximum (FWHM) of about 40 nm. Additionally, we also plot the line profile of the effective magnetic field H_{eff} (blue solid line) extracted for the same position as the dynamic magnetisation. The direct comparison of the two curves shows the strong confinement of the spin precession to the area of the potential well and, hence, to

the position of the domain wall.

Furthermore, we analysed the lateral spin-wave localisation from the experimental results shown in Fig. 2. The experimental data is plotted by red squares in Fig. 4 and is well reproduced by a Gaussian fit (red dotted line) with a FWHM of 340 nm. To compare this value to the simulation, we have to take into account that the detected BLS signal does not directly reflect the spatial distribution of the observed spin-wave mode but is the convolution of the actual spin-wave signal with the shape of the probing laser spot. In fact, the FWHM extracted from the data is already on the order of the focal spot size that is expected for the confocal microscope used in our study and, thus, confirms the strong lateral confinement revealed by micromagnetic simulations.

In a final step we go beyond these mere observations and demonstrate the intriguing flexibility of spin-wave nano-channels based on domain walls by actually controlling the spin-wave propagation path. In contrast to waveguides based on geometric confinement that cannot be modified after patterning, a domain wall can be easily manipulated by several means like magnetic fields, charge or spin currents. These manipulations of domain walls open a new perspective to control spin-wave transport in two-dimensional nanostructures and towards reconfigurable magnonic circuits and logic devices.

In the actual experimental scenario, we controlled the position of the domain wall by applying small external magnetic fields along the long axis of the waveguide. De-

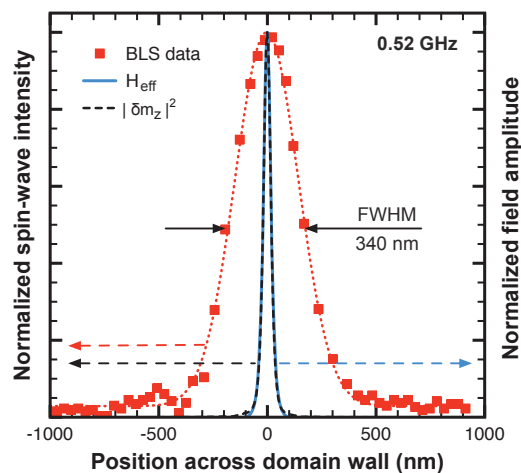


FIG. 4. **Spin-wave localisation.** Spin-wave intensity across the domain-wall width determined from experiment (squares) and simulation (dashed line) for 0.52 GHz. The dotted line represents a Gaussian fit to the measured data that is the convolution of the actual spin-wave signal with the focal spot size of the BLS microscope. This fit with a FWHM of 340 nm mainly reflects the spot size that is expected to be on the same order for confocal microscopes and, thus, suggest a confinement of several tens of nanometres. In accordance to this consideration, the micromagnetic simulation shows a strong lateral confinement of 40 nm for the spin-wave modes within the wall. The solid blue line is the magnitude of the effective magnetic field indicating the width of the potential well for the simulated magnetisation configuration.

pending on the polarity of the applied field, the growth of either the left or the right domain is favoured and results in a shift of the domain wall. Figure 5 shows BLS line scans across the width of the waveguide at four different small magnetic fields of -0.15 mT, -0.05 mT, 0.05 mT, and 0.23 mT. The measurements clearly show that the detected spin-wave mode is shifted together with the domain wall by the applied field. The observed left-right asymmetry of the domain-wall shift versus the field is attributed to the initial displacement of the domain wall even in the absence of any external magnetic fields.

In fact, the position of the domain wall and, thus, the spin-wave propagation path can be moved with nanometre precision over a distance of about $2 \mu\text{m}$ within a field range of only $\Delta H = 0.38 \text{ mT}$. The inset in Fig. 5 illustrates this displacement as a function of the applied field and yields a proportionality constant of $5.57 \mu\text{m/mT}$. These data already establish a novel mechanism to control spin-wave transport in 2D. Moreover, they pave the way for a potential reconfigurable but non-volatile magnonic nano-circuitry. In future scenarios, artificial pinning centres - for instance induced by ion implantation^{28,29} - might be used to enable switching between several stable remanence states with differ-

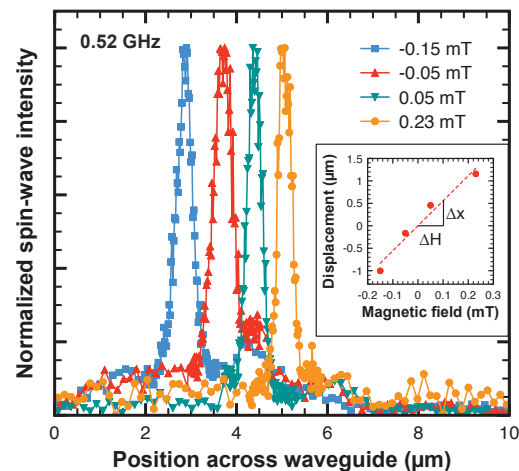


FIG. 5. **Steering spin waves with small fields.** Spin-wave intensity measured across the waveguide for different external magnetic fields applied parallel to the long axis of the waveguide for 0.52 GHz. The measurements show that the domain wall can be easily shifted with small applied fields over μm distances in both directions, therefore allowing for a fine control of the spin-wave channel position. The inset illustrates the displacement of the spin-wave propagation path as a function of the applied field and yields a proportionality constant of $5.57 \mu\text{m/mT}$.

ent domain-wall configurations that act as pre-defined nano-channels to guide spin-wave propagation in logic devices. Furthermore, multiple domain walls can form in materials with a perpendicular magnetic anisotropy with a separation of a few tens of nanometers¹⁸, which allows for an even higher integration density of these spin-wave nano channels.

In summary, we experimentally demonstrated the channelling of spin-waves in nanometre-wide magnetic domain walls with a width of about 40 nm. Micromagnetic simulations allowed for further analysis of the propagation characteristics. The spin-wave modes propagating inside domain walls exhibit a well-defined wave vector along their propagation path and a positive dispersion enabling information transport as well as data processing based on the interference of waves. Finally, we demonstrated a major advantage of domain-wall based magnonic waveguides over geometric patterning: manipulating the domain configurations with tiny, external fields well below 1 mT allowed us to actually shift the spin-wave propagation path over the distance of several micrometres. These observations pave the way for the realisation of reconfigurable, yet non-volatile magnonic circuitry by switching between different remanence states and, thus, for the realisation of energy-efficient and programmable spin-wave logic devices on the nanoscale.

METHODS

Sample Preparation. A bilayer of 3 nm Ti for adhesion and 40 nm Py was deposited on a SiO₂ substrate using electron-beam evaporation. Employing electron-beam lithography and lift-off techniques, micron-sized spin-wave waveguides were patterned. Their major part has a width of 10 μm which is gradually reduced to 5 μm towards one side to form a nucleation pad for a magnetic domain wall. In a second step, a 100-nm thick and 1.6- μm wide gold antenna is positioned at the beginning of the 10- μm wide part of the waveguide utilising electron-beam lithography, electron-beam evaporation and subsequent lift-off. A microwave current flowing through the antenna generates an oscillating magnetic field that couples to the magnetization and allows for the excitation of spin waves with well defined frequencies.

BLS microscopy. The spin-wave intensity is locally recorded by means of BLS microscopy at room temperature. BLS is the inelastic scattering of photons and magnons. Light from a continuous wave, single-frequency 532-nm solid-state laser is focused on the sample surface using a high numerical aperture microscope lens yielding a spot that can be approximated by a Gaussian profile with a FWHM of about 320 ± 50 nm.

The frequency shift of the inelastically scattered light is analysed using a six-pass Fabry-Perot interferometer TFP-2 (JRS Scientific Instruments) providing an accessible frequency range from 500 MHz to a few hundred gigahertz.

Micromagnetic simulations. The magnetisation dynamics is described by the Landau-Lifshitz-Gilbert equation of motion:

$$\frac{1}{\gamma} \frac{d\mathbf{m}}{dt} = -\frac{1}{1+\alpha^2} (\mathbf{m} \times \mathbf{H}_{\text{eff}}) - \frac{\alpha}{1+\alpha^2} \mathbf{m} \times (\mathbf{m} \times \mathbf{H}_{\text{eff}}), \quad (1)$$

where γ is the gyromagnetic ratio, $\mathbf{m} = \mathbf{M}/M_S$ the normalised magnetisation vector, α the Gilbert damping parameter, and \mathbf{H}_{eff} the total effective field comprising the exchange, anisotropy, magnetostatic, and external magnetic fields. The simulations are performed by numerically solving equation 1 using the GPU accelerated code MuMax3²⁵.

Simulations were performed for a rectangular element that is 5- μm long, 1- μm wide and 10-nm thick. This structure is discretised by cells with edge lengths $\Delta x = 2.44$ nm, $\Delta y = 1.90$ nm, and $\Delta z = 10$ nm, respectively. The following material parameters for Py are used: saturation magnetisation $M_S = 830$ kA/m, exchange constant $A = 1.3 \times 10^{-11}$ Jm⁻¹, and zero crystalline anisotropy $K = 0$ Jm⁻³. The Gilbert damping parameter is set to $\alpha = 0.007$.

The magnetisation dynamics is excited by an out-of-plane field pulse of Gaussian shape with 20-nm FWHM that is applied inside the domain wall at 1.6 μm distance from the bottom edge of the rectangular element. The time evolution of the field pulse is a *sinc* function chosen such that the cut-off frequency in the Fourier transform

is 40 GHz. The Fourier transform of the magnetisation dynamics was calculated over a period of 25 ns for each cell. The sum over all nodes provides the integral spectrum of the given configuration. The mode profiles of the individual resonances are determined by a backward windowed Fourier transform to deduce the dispersion relation.

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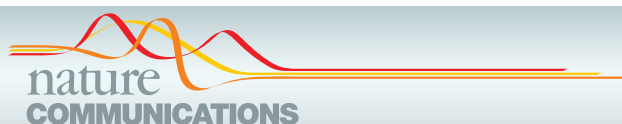
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AUTHOR CONTRIBUTIONS

K.W., T.S., A.H. and H.S. designed the experiment. K.W. and T.S. prepared the samples. K.W. performed the BLS microscopy measurements and analysed the experimental. K.W. and A.K. performed and evaluated the micromagnetic simulations. All authors interpreted and discussed the results and co-wrote the manuscript.

ADDITIONAL INFORMATION

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ARTICLE

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Ultrafast electronic response of graphene to a strong and localized electric field

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The way conduction electrons respond to ultrafast external perturbations in low dimensional materials is at the core of the design of future devices for (opto)electronics, photodetection and spintronics. Highly charged ions provide a tool for probing the electronic response of solids to extremely strong electric fields localized down to nanometre-sized areas. With ion transmission times in the order of femtoseconds, we can directly probe the local electronic dynamics of an ultrathin foil on this timescale. Here we report on the ability of freestanding single layer graphene to provide tens of electrons for charge neutralization of a slow highly charged ion within a few femtoseconds. With values higher than $10^{12} \text{ A cm}^{-2}$, the resulting local current density in graphene exceeds previously measured breakdown currents by three orders of magnitude. Surprisingly, the passing ion does not tear nanometre-sized holes into the single layer graphene. We use time-dependent density functional theory to gain insight into the multielectron dynamics.

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ts exceptionally high carrier mobility¹ makes graphene a promising material for future electronic applications. The linear Dirac-like dispersion and the associated constant high carrier velocity promise the realization of ultrafast devices in electronics², optics³ or even q-bits based on nitrogen vacancies⁴. The ultra-short timescales involved, <1 ps, provide stringent requirements on material properties. Direct current measurements on supported single layer graphene (SLG) reveal breakdown currents due to Joule heating larger than in copper, with densities of about 10^8 – 10^9 A cm⁻² (refs 5–8). Note that these measurements include substrate and finite size effects, which may increase the breakdown current as compared with pristine freestanding graphene. Indeed, heat dissipation via the substrate seems essential to achieve these numbers^{9,10}. Photoexcitation measurements reveal efficient excited carrier relaxation within a few hundred femtoseconds^{11,12} yet probe the lattice on the length scale of the optical wavelength. Moreover, large electric fields^{13,14} and collisions with energetic particles^{15–28}, which allow for tuning the properties of graphene, further modify the response of the material. A reliable tool to locally probe the timescales of the electronic response of pristine graphene to large fields is thus urgently needed.

One way to measure the short-time response of materials is the irradiation with highly charged ions (HCIs), which results in an extremely large, local external field: the Coulomb field of an approaching HCI. A charge state of $q_{\text{in}} = 35$ implies a local electric field strength of 1.8×10^{11} V m⁻¹ at 5 Å distance from it. Achieving the same local field strength using laser fields would require power densities above 10^{17} W cm⁻², a field strength where non-destructive measurements become challenging. Previous work on scattering of HCI from solid surfaces or their transmission through freestanding carbon membranes reported unexpectedly large charge capture within 5–30 fs (refs 29–34).

In this contribution, we take the final step and present the results for the ultimately thin carbon target, a freestanding single layer of graphene. Measurements of the charge state and energy of the transmitted ions and time-dependent density functional theory (TDDFT) calculations show that large number of electrons are extracted from a very small surface area, which implies a high local surface current density. We identify a multielectron process and estimate the relevant timescales for charge transfer along the graphene layer. For example, while passing through SLG, the HCI with initial charge state $q_{\text{in}} = 30$ captures ~25 electrons on a timescale of a few fs. Based on the experimental data, a lower bound for the current densities reached locally within a nm² area is at least 10^{12} A cm⁻² exceeding the breakdown current densities reported so far^{5–8} by three orders of magnitude.

Results

Exit charge state analysis. The amount of charge transferred to the HCI can be estimated by measuring the distribution of exit charge states q_{out} and the energy of highly charged Xe ions after transmission through SLG (for details of sample preparation, see Supplementary Note 1). Initial ion charge states of $10 \leq q_{\text{in}} \leq 35$ and velocities below 0.5 nm fs⁻¹ were used. Ions are transmitted through SLG under normal incidence and analysed with respect to their charge state and kinetic energy by an electrostatic analyser (Fig. 1c). Typical transmission spectra (Fig. 1a) show a distribution of different exit charge states, with a mean value \bar{q}_{out} shifted towards smaller exit charge states for slower ions. To extract the abundances and widths of every single peak, the spectra have to be deconvoluted with the analyser function, since the spectra are broadened by the design of the electrostatic analyser (details on the data evaluation can be found in ref. 35). The corrected abundances follow a symmetric Gaussian function with

a full width at half maximum of three to five electrons as a result of final de-excitation processes. From these Gaussian fits the mean value \bar{q}_{out} is extracted. Even for the smallest velocities (largest interaction times) used in our experiment ($v_{\text{min}} = 0.13$ nm fs⁻¹), \bar{q}_{out} remains considerably larger than the equilibrium charge state of a Xe ion in a solid target ($q_{\text{eq}} \sim 1$)³⁶, indicating incomplete neutralization.

Depending on the initial charge state the HCI captures and stabilizes between 20 and 30 electrons during its transmission through graphene (Fig. 1e). The experimentally observed average electron capture is extracted from the mean exit charge state of each transmission spectrum. The transmission time through graphene can be defined by $\tau = d_{\text{eff}}/v$, where v is the ion velocity. The effective interaction length d_{eff} corresponds to the projectile-surface distance where the electron transfer processes between the HCI and graphene take place. The values of d_{eff} can be obtained from the TDDFT calculations which yield for $q_{\text{in}} = 20$ a value of $d_{\text{eff}} \simeq 9$ Å. This is in good agreement with predictions of the classical over the barrier model³⁷.

The number of captured and stabilized electrons (they are not reemitted due to autoionization processes) as function of interaction time is shown in Fig. 1e. The data can be well fitted by the simple expression $\bar{q}_{\text{out}} = q_{\text{in}} \left(1 - e^{-\tau/\tau_n^{\text{exp}}}\right)$ (1)

with an effective q_{in} -neutralization time constant τ_n^{exp} . Using $d_{\text{eff}} = 9$ Å we obtain $\tau_n^{\text{exp}} = 2.1$ fs for $q_{\text{in}} = 35$, and $\tau_n^{\text{exp}} = 1.4$ fs for $q_{\text{in}} = 20$, respectively. The performed TDDFT study (for details, see Supplementary Note 2 and Supplementary Fig. 4) shows a multielectron character of the charge transfer that can explain the experimentally observed strong reduction of the charge state of the HCI. We calculate that HCIs with initial charge states $q_{\text{in}} = 10, 20$ and 40 capture ~9, ~17 and ~34 electrons during the passage through the graphene layer, respectively.

Since at least $q_{\text{in}} - \bar{q}_{\text{out}}$ electrons are transferred from the graphene sheet to the HCI during the interaction, the lower bound for the electron transfer rate is given by $(q_{\text{in}} - \bar{q}_{\text{out}})/\tau$, which corresponds to 10^{16} electrons per second as can be estimated from the experimental data. This corresponds to a local current of $I \gtrsim 1.5$ mA. The TDDFT study shows on the one hand that the charge is extracted from the graphene area with lateral radius $R = 5$ Å around the impact point that we can define as an interaction region (Figs 1b and 2), and on the other hand that the electron flow along the graphene layer compensates the electron extraction by the HCI on the timescale of the collision (fs), otherwise the neutralization of the projectile would be stopped by the local charging of the target. The latter is in accord with experimental data that shows the absence of the post-collisional defects that would result from Coulomb explosion (see the section Transmission electron microscopy results). The electrons moving along graphene enter the interaction region crossing the surface $S = 2\pi Rh$ ($h = 3.4$ Å is the width of the graphene layer that we estimate from the interlayer spacing of graphite³⁸). Thus, a time averaged electron current density $J = I/S$ in the graphene plane exceeding $\sim 6 \times 10^{11}$ A cm⁻² is reached. This value, however, is just a lower limit because it only accounts for the electrons captured by the projectile. Indeed, because of the interaction with HCI N_{vac} electrons will also be emitted into vacuum. We calculate that for each captured electron approximately one additional electron is emitted. N_{vac} can be even larger if one accounts for Auger processes involving tightly bound electronic shells of the projectile. Estimations as high as $N_{\text{vac}} \approx 3 \times q_{\text{in}}$ (ref. 39) have been reported.

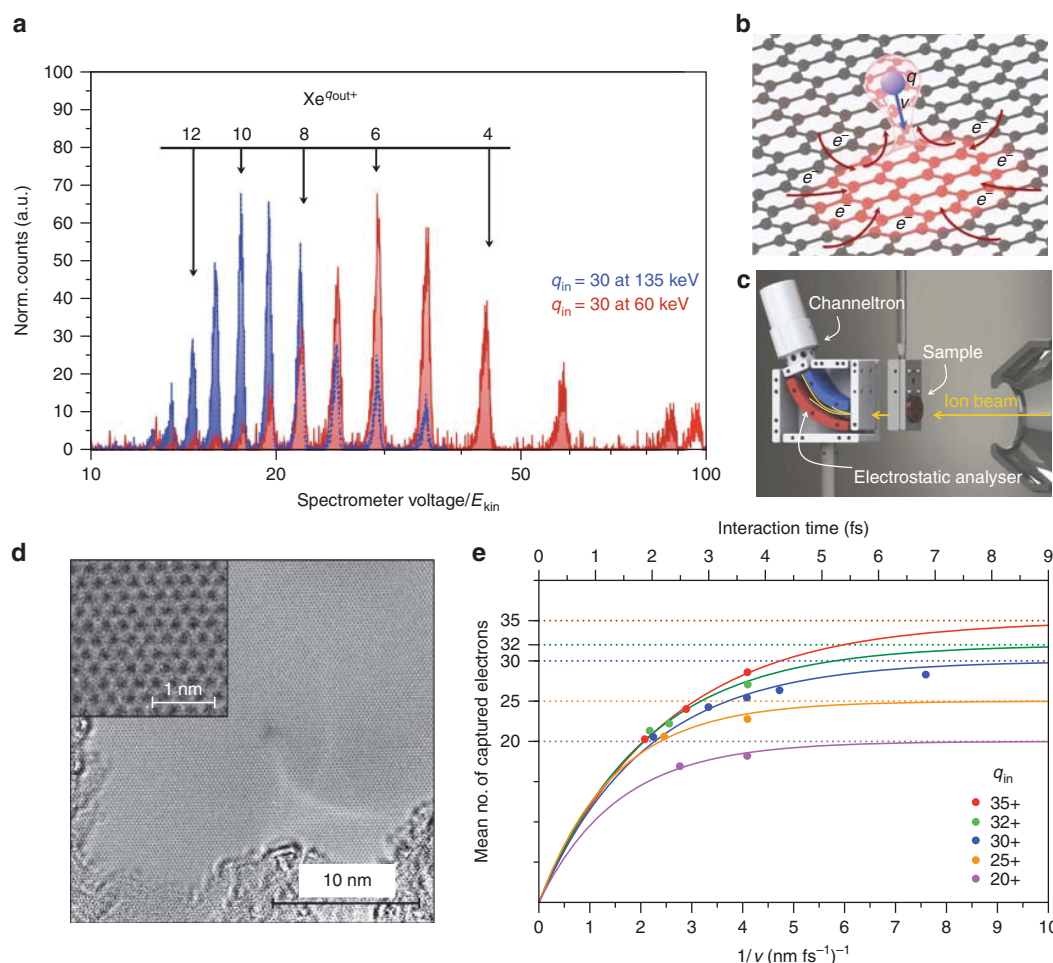


Figure 1 | Experimental scheme and results. (a) Measured spectra of a Xe^{30+} beam at kinetic energies of 135 and 60 keV (blue and red, respectively) transmitted through a freestanding SLG sheet. Exit charge states q_{out} are calculated from the spectrometer voltage of the electrostatic analyser. The exit charge state distribution shifts towards smaller average exit charge q_{out} for slower ions. (b) Schematic of the interaction process between freestanding SLG and an approaching highly charged ion (HCI). The HCI extracts a lot of charge from a very limited area on the femtosecond time scale leading to a temporary charge-up of the impact region. (c) Sketch of the experimental set-up with the target holder and electrostatic analyser. (d) TEM image of a freestanding monolayer of graphene after irradiation with Xe^{40+} ions at 180 keV with an applied fluence of 10^{12} ions per cm^2 (about six impacts on the shown scale). No holes or nanosized topographic defects could be observed. The inset shows the intact hexagonal structure of graphene. (e) Average number of captured and stabilized electrons ($q_{in} - q_{out}$) after transmission of $Xe^{q_{in}+}$ ions through a single layer of graphene as a function of the inverse projectile velocity for different incident charge states. Fits to the experimental data points assume a continuous neutralisation following an exponential function. Neutralisation time constants of a few femtoseconds can be extracted.

Transmission electron microscopy results. Failure to sufficiently resupply the lost charge and to dissipate the absorbed energy on a timescale small compared with lattice vibrations would result in Coulomb explosion tearing large holes (of the order of 10 nm) into the SLG, as we have observed for carbon nanomembranes^{35,40}. Despite the possible self-healing of localized defects in graphene⁴¹, such extended structural modifications should be detectable using transmission electron microscopy (TEM) or scanning transmission electron microscopy (STEM), yet careful investigation of the irradiated SLG does not reveal any nanometre-sized defect structures. Note that for freestanding SLG used in our study, in contrast to supported graphene layers, the defect formation because of a collision cascade in the substrate⁴² is not operative. In our case, elastic collisions (nuclear stopping) may cause direct knockout of carbon atoms, but less than one carbon atom is sputtered on average by a 10–100 keV Xe ions according to Lehtinen *et al.*⁴³. Even if point defects are produced, they will likely disappear due to dissociation of ubiquitous

hydrocarbon molecules⁴¹. In Fig. 1d, a typical TEM image of a freestanding monolayer of graphene after irradiation with Xe^{40+} ions with a kinetic energy of 180 keV is shown. The applied fluence of 10^{12} ions per cm^2 corresponds to approximately six ion impacts within this $25\ nm \times 25\ nm$ frame. No rupture could be detected. This is in strong contrast to ultrathin polymeric carbon nanomembranes, where, because of low electron mobility, creation of pores with diameters of up to a few nanometre was observed after exposure to HCl₃^{35,40}. The absence of any traces of large-scale lattice deformations thus confirms the intrinsic ability of suspended SLG to locally sustain exceptionally high current densities, even though it cannot efficiently diffuse heat to a substrate^{9,10}.

Local current density. To illustrate the way the electronic processes take place, in Fig. 2, we show snapshots of the current density for a $q_{in} = 20$ projectile incident at graphene with velocity $v = 0.87\ nm\ fs^{-1}$. Results of the TDDFT calculations are

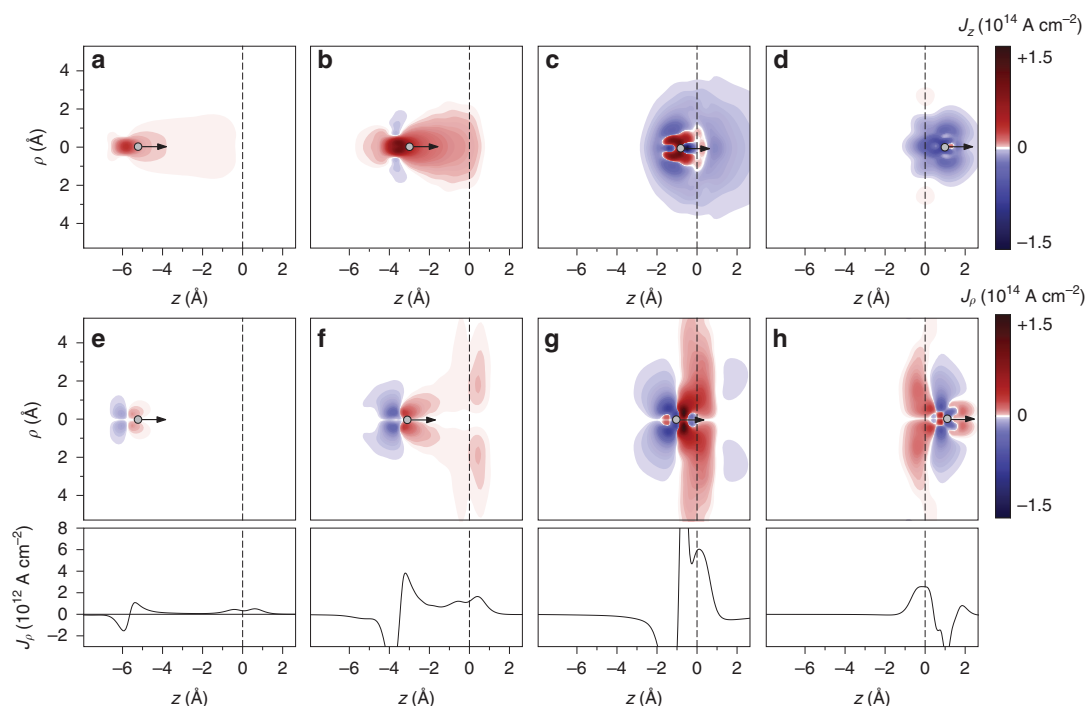


Figure 2 | Perpendicular and radial current density obtained by TDDFT calculations. Snapshots of the perpendicular J_z (a–d) and radial J_ρ (e–h) components of the current density for $q_{in}=20$ at four different HCl–graphene distances obtained from TDDFT calculations performed in cylindrical (ρ, z) coordinates with z -axis set along the projectile trajectory perpendicular to the target surface. The figures show that already above the graphene layer electrons are transferred to the approaching HCl and the current density along the direction of motion explains the charge exchange of the HCl. Extremely high transverse current density (f–h) along the graphene layer is obtained. The corresponding profiles (lower panels) show the z -dependent transverse current density averaged over a circle of 10 Å in radius. Values exceeding $10^{12} \text{ A cm}^{-2}$ are obtained. The position of the graphene layer by the vertical dashed line.

presented for four different HCl–graphene distances z_{ion} along the ingoing trajectory path. We use cylindrical (ρ, z) coordinates, with the z axis set along the projectile trajectory assuming normal incidence geometry. Note that because of the cylindrical symmetry in our calculations (see Supplementary Note 2 for details), there is no azimuthal angular dependence. Already at $z_{ion} \sim -6 \text{ Å}$ charge transfer from graphene to the HCl is sizable. For $z_{ion} \sim -3 \text{ Å}$, the J_z component of the current density along the direction of motion is further increased and a significant transverse current density along the graphene layer (located at $z=0$) can be observed.

The current density along the direction of motion J_z determines the charge exchange and energy loss of the HCl (see the section Energy loss), while the current density along the transverse direction J_ρ shows the fast local response of graphene to the strong HCl perturbation. In fact, our calculations reproduce our experimental estimate since the transverse current densities in the proximity of the graphene layer reaches values as high as $10^{12} \text{ A cm}^{-2}$. This means that, transiently and locally, in the femtosecond and nanometre scale, graphene is able to sustain extremely high current densities. The positive charges created by electron capture and electron emission are spread over the entire layer⁴⁴.

Energy loss. As neutralization is incomplete in our measurements charge state effects on the energy loss become experimentally accessible. As observed earlier with thicker foils^{31,34,45}, the energy loss of a HCl passing through thin sheets strongly depends on the number of electrons transferred to the ion. The energy loss as

experimentally deduced from the positions of the peaks for exit charge states $q_{out}=2$ and $q_{out}=4$ increases quadratically with the incident charge state (Fig. 3a). We observe keV energy losses, which are more than an order of magnitude larger than the result from a TRIM simulation (nuclear and electronic stopping) for neutral Xe transmitted through a layer with areal density of $3.82 \times 10^{15} \text{ at cm}^{-2}$ representing the target⁴⁶ (dashed line in Fig. 3a). This simulation predicts an energy loss of 228 eV taking into account our detector acceptance angle. The TRIM value is close to the energy loss expected from an extrapolation of our fit functions through our experimental data at $q_{in}=0$ representing equilibrium stopping.

According to our TDDFT calculations, the non-adiabaticity of the charge-exchange and ionization processes introduced by the ion motion translates into an electronic energy loss of similar magnitude and charge state dependence as that observed in the measurements, although somewhat underestimated. The lower values in the theory as compared with the experiment are due to the fact that the used pseudo-potential description of the HCl does not include the full Coulomb singularity at distances r smaller than a cutoff radius R . As discussed in the Supplementary Note 2 and shown in Supplementary Fig. 5, a simple change in the cutoff radius from $R=0.53 \text{ Å}$ to $R=0.26 \text{ Å}$ increases the value of the energy loss by a factor of two. Since the actual HCl would correspond to the value $R=0$ and our aim is not to reproduce the data but to explain them, we consider our results rather satisfactory. In addition, the overestimation of the minimum excitation energy of graphene valence electrons in the jellium model is another reason for the underestimation of the energy loss, as it has been explicitly checked for helium

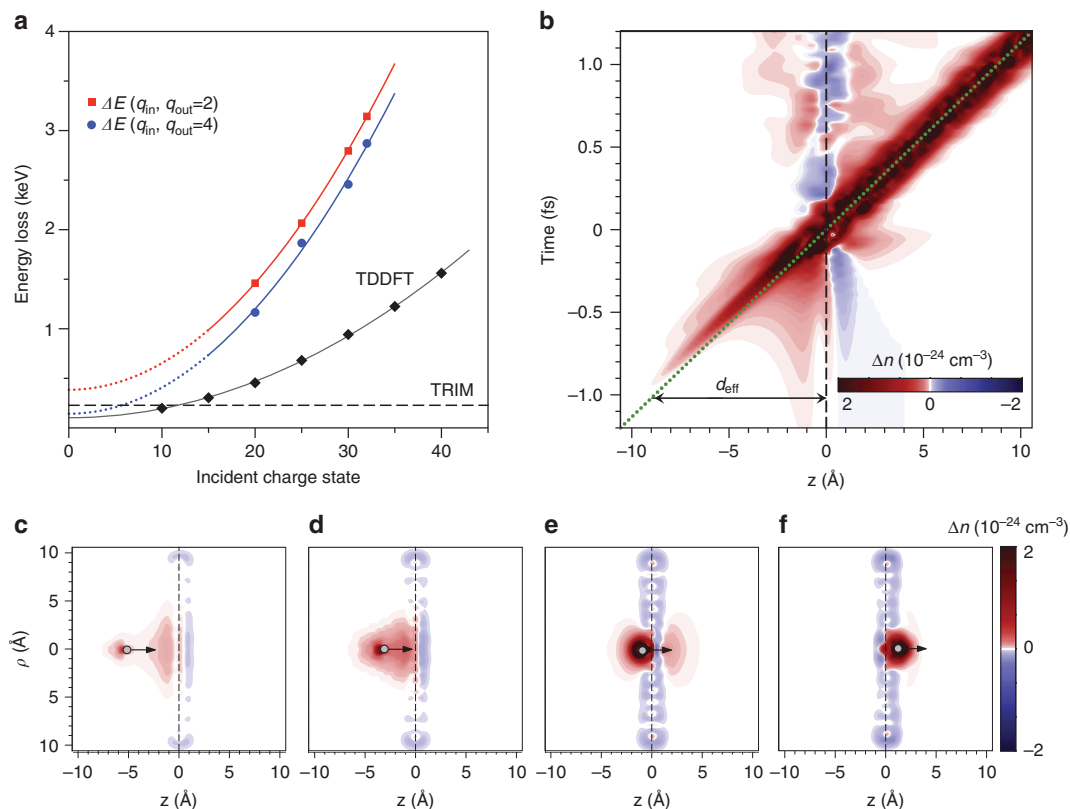


Figure 3 | Ion energy loss results and a detailed view of the neutralization dynamics. (a) Experimentally determined energy loss for ions with exit charge state $q_{\text{out}} = 2$ and $q_{\text{out}} = 4$ as a function of the incident charge state q_{in} . The energy of the projectiles was kept constant for all q_{in} at $E = 40$ keV. The data points are fitted by a quadratic function and the dashed line shows the result from a TRIM simulation (nuclear and electronic stopping) for a graphite layer of 3 Å thickness. The experimental data are compared with results from TDDFT calculations that reproduce the parabolic dependence with the initial charge state and the order of magnitude of the energy loss. (b) The induced charge density along the z -axis perpendicular to the surface and passing through the ion centre as function of time for $q_{\text{in}} = 20$ at $v = 0.87$ nm fs $^{-1}$ using the TDDFT description. It shows that already ~ 9 Å above the graphene layer the HCl starts to capture electrons. It is also visible that the induced charge is not centred at the HCl position but lags behind (see green dashed line). By approaching the surface more and more electrons are captured. (c–f) Snapshots of the induced charge density in cylindrical (ρ, z) coordinates for four different position of the incoming HCl projectile. (Supplementary Movie 1). The polarization of the surface due to the approaching HCl and the excitation of the graphene layer while and after the crossing of the ion are clearly visible as well as the HCl neutralization.

ions (Supplementary Note 2; Supplementary Figs 2 and 3). Reproducing the q_{in}^2 dependence and the order of magnitude of measured energy losses by our TDDFT calculations shows again that electronic response of the simulated system is well described.

The fact that the measured projectile energy loss can be assigned to the electronic excitations agrees with results obtained in ab initio studies for low projectile charges³⁸. It provides a consistent link between charge transfer and energy loss processes and allows to explain the absence of the induced damage despite the large energy deposition. Indeed, owing to the high electron mobility of graphene the positive charges created in the local surface area by electron capture and electron emission into vacuum are promptly screened thus reducing the local electronic temperature.

A detailed view on the neutralization dynamics is given in Fig. 3b, where we show a two-dimensional plot of the induced density along the direction of the ion motion at different ion positions from a simulation done for $q_{\text{in}} = 20$ using our TDDFT description. The horizontal axis represents the distance to the graphene layer located at $z = 0$ and the vertical axis corresponds to the timescale. The HCl starts capturing electrons from graphene already at 9 Å. The strong attractive potential accelerates electrons towards the HCl and, approximately, half

of them end up captured by it along the incoming path before penetration into the graphene sheet. The induced electronic charge density as the HCl approaches the graphene layer has two components (Fig. 3c–f): one is formed by the convoy electrons around the HCl position, forming an asymmetric wake potential that slows down the ion, and the other one is located at the graphene layer due to the target polarization. Both components merge as the HCl gets closer to the target (Fig. 3e) and forward electron emission starts. Finally, after crossing the layer (Fig. 3f) the projectile is nearly neutralized and the corresponding induced electronic charge is centred around the HCl along its outgoing path. The actual HCl is probably not fully relaxed at the instant of crossing the graphene layer and, therefore, it still suffers a number of autoionization processes (not described in TDDFT) and subsequent de-excitation without a significant energy loss.

Discussion

We have studied the electronic response of SLG to a large external field of an approaching HCl. We find an ultrafast neutralization within a few femtoseconds timescale leading to the capture and stabilization of almost (90%) all the missing electrons in the projectile. Our experiments and TDDFT calculations, both

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suggest local current densities in the graphene plane exceeding $10^{12} \text{ A cm}^{-2}$, at least three orders of magnitude higher than previously established local breakdown currents, however, on a timescale of a few femtoseconds only. The exceptional electronic properties of graphene allow for a resupply of charge and distribution of the impact energy promptly enough to prevent Coulomb explosion in the electron-depleted region. In addition, the surprisingly large energy loss of the ion of a few keV, which is strongly connected with the charge-exchange process and depends on the incident and exit charge state, could be successfully explained. Our study revealed how graphene responds to extremely high fields and our results underline the exceptional properties of graphene for ultrafast electronic applications at high current densities.

Methods

Experimental set-up. The measurements are performed at the Ion Beam Center of the Helmholtz-Zentrum Dresden-Rossendorf. Highly charged Xe ions are produced in a room temperature electron beam ion trap, charge state separated by an analysing magnet and then guided by several electrostatic lenses into the target chamber. Due to an electrostatic deceleration system, the kinetic energy of the extracted Xe ions can be varied between 0.1 and 4.4 keV corresponding to velocities between 0.13 and 0.5 nm fs⁻¹. The pressure in the experimental chamber is kept below 5×10^{-9} mbar during measurements to prevent charge-exchange processes of the ions before interaction with the target.

The freestanding SLG sheets, which span over a regular array of holes in a TEM grid, are produced at the University Duisburg-Essen and transferred without the use of polymer coating (see Supplementary Note 1 and Supplementary Fig. 1 for details on the sample preparation and characterization). Before performing the transmission measurements, the graphene sheets are inspected by STEM to check the sample coverage and the grade of residual contamination. Contamination of the surface by water was either not present or does not affect the charge-exchange processes as the results of the experiments did not change when heating the samples up to 200 °C in ultra-high vacuum before and during the measurements. A heatable target holder and an electrostatic analyser are mounted in the target chamber (Fig. 1c). The electrostatic analyser is equipped with two channeltrons to analyse the charge state and energy of the transmitted ions and to count neutralized particles in forward direction. The analyser has an acceptance angle of 1.6°, the energy resolution was determined to be $\Delta E/E \approx 1.5 \times 10^{-3}$. The analyser's maximum operation voltage of 5,000 V limits the range of measurable charge states for projectiles with larger kinetic energies.

TDDFT calculations. Our TDDFT simulations are done following the time evolution of the Kohn-Sham orbitals (see Supplementary Note 2 for details) of the system defined by the constant velocity approach of a model pseudo-potential HCl (cutoff radius $R = 0.26 \text{ Å}$, Coulomb tail Q/r) and a planar jellium disk with the correct work function value (4.6 eV)^{13,47} representing the graphene layer. A non-uniform real space grid in cylindrical coordinates is used to treat properly the Coulomb singularity close to $r = R$. Finite size effects have been checked using jellium disks of different sizes containing 500, 1,004 and 2,000 electrons (Supplementary Figs 6 and 7). The Gunnarson and Lundqvist approximation⁴⁸ for the exchange correlation Kernel was used.

Data availability. The data that support the findings of this study are available from the corresponding author upon request.

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Author contributions

E.G., R.A.W. and V.S. performed the measurements, F.A. and S.F. were involved in planning and supervised the work, E.G. and R.A.W. processed the experimental data, performed the analysis, drafted the manuscript and designed the figures. R.P., I.A., A.K.K., A.A. and A.G.B. performed the TDDFT calculations. R.K., A.H. and M.S. manufactured the samples and characterized them with Raman spectroscopy and TEM, B.C.B. performed the STEM characterization. F.L. and A.V.K. aided in interpreting the results and worked on the manuscript. All authors discussed the results and commented on the manuscript.

Additional information

Supplementary Information accompanies this paper at <http://www.nature.com/naturecommunications>

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Droplet-Confined Alternate Pulsed Epitaxy of GaAs Nanowires on Si Substrates down to CMOS-Compatible Temperatures

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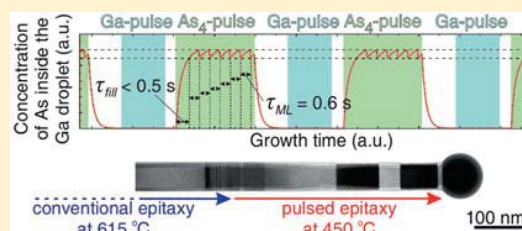
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Supporting Information

ABSTRACT: We introduce droplet-confined alternate pulsed epitaxy for the self-catalyzed growth of GaAs nanowires on Si(111) substrates in the temperature range from 550 °C down to 450 °C. This unconventional growth mode is a modification of the migration-enhanced epitaxy, where alternating pulses of Ga and As₄ are employed instead of a continuous supply. The enhancement of the diffusion length of Ga adatoms on the {110} nanowire sidewalls allows for their targeted delivery to the Ga droplets at the top of the nanowires and, thus, for a highly directional growth along the nanowire axis even at temperatures as low as 450 °C. We demonstrate that the axial growth can be simply and abruptly interrupted at any time without the formation of any defects, whereas the growth rate can be controlled with high accuracy down to the monolayer scale, being limited only by the stochastic nature of nucleation. Taking advantage of these unique possibilities, we were able to probe and describe quantitatively the population dynamics of As inside the Ga droplets in specially designed experiments. After all, our growth method combines all necessary elements for precise growth control, in-depth investigation of the growth mechanisms and compatibility with fully processed Si-CMOS substrates.

KEYWORDS: nanowire, self-catalyzed, pulsed epitaxy, GaAs, Si substrate



The self-catalyzed (or Ga-induced) growth of GaAs nanowires (NWs) on Si(111) by molecular beam epitaxy (MBE) has offered the opportunity to obtain vertical NWs with fairly good control on the structural polytypism (especially in favor of the zincblende phase),^{1–5} without the risk of contamination by foreign elements like Au.⁶ On those grounds, the plethora of exciting nanoscale phenomena already discovered in GaAs NWs^{7–12} could eventually be integrated monolithically on Si circuit platforms.^{13–15} The growth of NWs is typically performed close to the congruent sublimation temperature of GaAs (580–630 °C) in combination with relatively high V/III flux ratios.¹⁶ These conditions ensure sufficiently high surface diffusion of Ga adatoms from the substrate and the NW sidewalls to the apex of the NWs, without thermal decomposition of the {110} sidewalls. Nevertheless, the specific growth conditions impose several limitations: (1) the growth temperature exceeds largely the thermal budget limit of fully processed Si complementary metal–oxide–semiconductor (CMOS) circuits,¹⁷ rendering the future integration of the two material technologies impossible; (2) the interruption of axial growth cannot be atomically abrupt due to the long shut-off transient of the As flux, which typically leads to an unintentional prolongation of the growth with a disturbed crystal phase being formed just below the Ga droplet;^{3,18,19} (3) the composition or doping profiles in axial heterostructures cannot be atomically abrupt due to effects like interdiffusion, surface segregation, phase separation, or the

reservoir effect, which are more prominent at high temperatures (where kinetic limitations are weak and solubility in liquid phase is higher).^{20–23} Alternatively, metal–organic chemical vapor deposition (MOCVD) has been used for the self-catalyzed growth of GaAs NWs on Si(111) at 420–435 °C, but there is poor control of the shape (strong tapering) and the structural purity (rotational twin planes perpendicular to the growth direction) of the NWs.²⁴

Aiming to surmount the aforementioned high-temperature limitations of the conventional MBE, we introduce an unconventional growth scheme called *droplet-confined alternate pulsed epitaxy* (DCAPE), where the Ga and As₄ beams are not supplied simultaneously, but alternately in short pulses. This mode resembles the so-called migration-enhanced epitaxy, which has been used in the past for the layer-by-layer growth of GaAs-based planar heterostructures at relatively low temperatures.²⁵ The success of the migration-enhanced epitaxy relies on the enhancement of the diffusion of Ga adatoms along the GaAs(001) surface when doses of Ga (As₄) comparable to the atom sheet-density of the growth interface are supplied in the absence of As₄ (Ga). In our case, though, the aim is not the layer-by-layer growth mode, but the minimization of the radial

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NW growth and the enhancement of the axial one by transferring all the Ga adatoms from the sidewalls into the Ga droplet at the NW apex. This is possible due to the particular arrangement of the surface atoms on the GaAs{110} sidewalls, where the diffusivity of Ga adatoms is high and the sticking coefficient of As₄ is low.^{26,27} The alternate supply of Ga and As₄ has been used more recently for selective area growth of GaAs(111) pillars with {110} sidewalls at a typical high temperature (590 °C),²⁸ whereas only one report deals with the self-catalyzed growth of GaAs(111) NWs with {110} sidewalls in the temperature range of 540–580 °C.²⁹ The structural information, however, was limited to NWs grown at 580 °C, where a zincblende phase with a high density of (111) twin planes was observed, whereas the NW dimensions were found to be insensitive to the growth temperature in the investigated range.

In this work, we investigate the DCAPE of self-catalyzed GaAs NWs on Si(111) substrates in the temperature range of 450–550 °C. We study the dependence of the Ga adatom surface diffusivity on the growth conditions (namely, the growth temperature (T_{gr}) and the V/III flux ratio) and show that the directional character of the growth can be maintained even at the lowest temperature investigated here, in clear contrast to conventional MBE. Furthermore, we describe the correlation of the crystal structure of the NWs with the growth conditions used and demonstrate the possibility to obtain defect-free zincblende NWs. Finally, the possibility to employ very low growth rates and defect-free growth interruptions gave us a unique opportunity to design and conduct experiments dedicated to an insightful quantitative description of the growth mechanisms.

As this work did not concern the nucleation stage, GaAs NWs grown in the conventional mode at 615 °C were used as templates for all experiments reported here. The template NWs were 2 or 3 μm long and 25 or 40 nm thick, respectively, and a representative side-view (perpendicular to the substrate normal) scanning electron microscopy (SEM) image is shown in Figure 1a. The important role of the surface diffusivity of Ga adatoms is manifested by the fast axial growth rate of those NWs, which was approximately 50 times higher than what would be expected only from the direct Ga flux (i.e., the flux of Ga that impinged directly onto the Ga droplets at the apex of the NWs). That is, the amount of Ga adatoms that diffused from the substrate surface and the NW sidewalls to the Ga droplets was 50 times larger than the amount of Ga atoms that were deposited directly onto the Ga droplets. DCAPE follows the template growth and typically consists of a pulse sequence of Ga pulse/interruption/As₄ pulse/interruption, which is repeated numerous times. Comparing the dimensions and morphology of the NWs grown in DCAPE mode with those of the template by SEM (see Methods), we were able to study the growth mechanisms and to identify the optimal growth parameters.

First, we studied the DCAPE of self-catalyzed GaAs NWs at $T_{gr} = 550$ °C, which was the highest temperature in the investigated range (DCAPE at 615 °C resulted in complete thermal decomposition of the GaAs template NWs, demonstrating the difficulty to interrupt the growth at such high temperatures). A series of samples grown with different V/III ratios was used for that purpose (the Ga flux or pulse duration was varied, while the As₄ flux and pulse duration were kept constant; see Methods). For DCAPE, we define V/III as the ratio between the supplied amount of As₄ per As₄ pulse and the

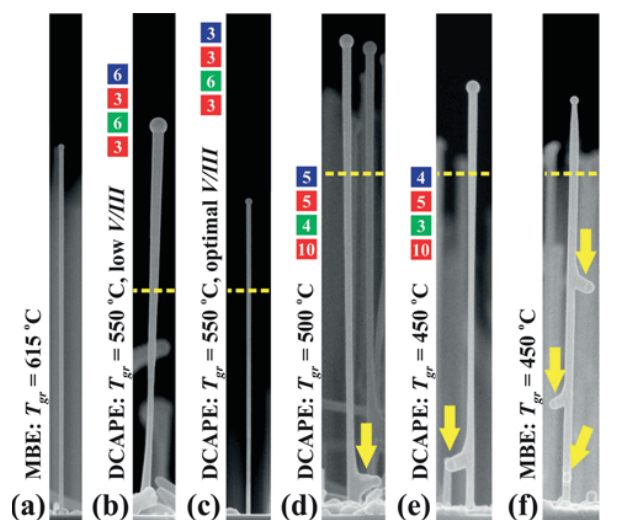


Figure 1. Side-view SEM images of GaAs NWs grown on Si(111). (a) Template NW grown in the conventional MBE mode at 615 °C. (b)–(e) NWs grown in DCAPE mode (on templates) at various V/III ratios or temperatures (T_{gr}) as indicated. The dashed yellow lines indicate the DCAPE/MBE interface. The yellow arrows indicate secondary Ga droplets on the NW sidewalls that formed during DCAPE. The pulse sequence of DCAPE is described with the colored boxes: the blue and green boxes indicate the pulse duration in seconds for Ga and As₄, respectively, while the red ones indicate the interruption time in seconds. The sequence reads from top to bottom and represents one period of an iterative process. (f) NW grown in the conventional MBE mode (on a template) at 450 °C. The yellow arrows indicate secondary Ga droplets on the NW sidewalls that formed during the growth at 450 °C. The scale bar corresponds to 1 μm for all images.

supplied amount of Ga per Ga pulse (each one is given by the product of beam flux × sticking coefficient at the given growth temperature × pulse duration). As depicted in Figure 1b, the NWs grow thicker at the top when a relatively low V/III ratio is used. The dashed yellow line indicates the position of the DCAPE/MBE interface, that is, the interface between the MBE template and the DCAPE segment. The NW morphology remains identical to that of the template NWs only when a high enough V/III ratio (11.5 in this case) is used as shown in Figure 1c. A further increase of the V/III ratio resulted in complete consumption of the Ga droplets (not shown here). The dependence of the NW morphology on the V/III ratio is attributed to the different arrival fluxes of Ga and As at the Ga droplets of the NWs (we refer to the amount of effective As rather than the total amount of As inside the Ga droplet). Those fluxes need to match in order to maintain a stable size of the Ga droplets and, thus, a homogeneous diameter along the NW axis. If this precondition is not fulfilled, but more Ga than As atoms arrive at the Ga droplets ($V/III < 11.5$), then the Ga droplets will grow larger and, consequently, the NWs will grow thicker than the template underneath. On the contrary, if less Ga than As atoms arrive at the droplets ($V/III > 11.5$), eventually the Ga droplets will be consumed and the self-catalyzed growth will be discontinued.

For a quantitative description of our observations, we measured the axial growth rate of the NWs during DCAPE (GR_{NW} ; see Methods) and compared it with what would be expected only from the direct flux of Ga (GR_{Ga}) or As₄ (GR_{As_4}) at the Ga droplets (the contribution of the direct fluxes to the

axial growth rate was calculated following ref 30 for a droplet contact angle of 130°). This is plotted in Figure 2a as a function

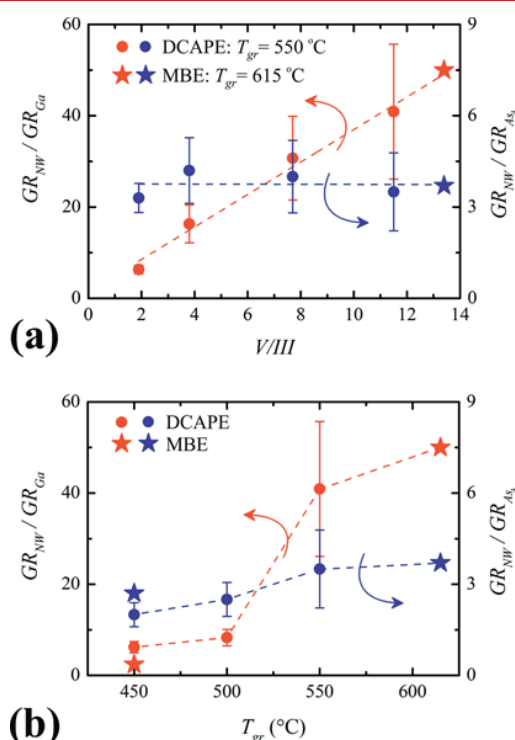


Figure 2. Plot of the GR_{NW}/GR_{Ga} (left axis/red dots) and the GR_{NW}/GR_{As_4} (right axis/blue dots) ratio for GaAs NWs grown in DCAPE mode as a function of (a) V/III ratio ($T_{gr} = 550^\circ C$) and (b) T_{gr} (with an optimized V/III at each T_{gr}). The star symbols correspond to NWs grown exclusively in the conventional MBE mode either at 615 or at 450 °C.

of V/III ratio for a constant As_4 flux (only Ga flux was varied). As expected for the self-catalyzed growth of GaAs NWs, where the axial growth rate is limited by the As_4 flux, the GR_{NW}/GR_{As_4} ratio was found to be constant for $V/III \leq 11.5$. In fact, it was found equal to 3.8 ± 0.5 , suggesting that each Ga droplet is an efficient collector of the As_4 rebound flux, that is, the flux of As_4 that re-evaporates from the surrounding substrate surface or the sidewalls of the neighboring NWs.³¹ Consequently, the linear decrease of the GR_{NW}/GR_{Ga} ratio with decreasing the V/III ratio (or with increasing the Ga flux) must reflect the increasing deviation from the matching condition of the arriving Ga and As at the Ga droplet. Comparing the DCAPE growth under optimal conditions ($V/III = 11.5$) with the template NWs (star symbols in Figure 2a), we found very similar GR_{NW}/GR_{As_4} and GR_{NW}/GR_{Ga} values. This means that we were able to grow NWs by DCAPE at 65 °C lower than the conventional MBE without any significant change in their morphological characteristics. However, the DCAPE NWs have superior crystal quality as will be presented in a following section.

Next, we investigated the DCAPE of GaAs NWs at even lower T_{gr} , namely at 500 and 450 °C. Figure 1d,e illustrate representative SEM images from single NWs grown at the given temperatures. In both cases, a V/III ratio of approximately 3 was employed in order to achieve a good matching of the Ga and As arrival fluxes at the specific T_{gr} . Small deviations from a perfect matching were still present (this

is a matter of fine-tuning of the V/III ratio), as evidenced by the slight increase of the diameter close to the top of the NWs, but this does not affect our analysis. Clear differences between the DCAPE NWs at 450 or 500 °C and those at 550 °C are the lower optimal value of V/III ratio, the formation of a secondary Ga droplet on the sidewalls near the base of the NWs (indicated by yellow arrows in Figure 1d,e), as well as the pronounced growth of polycrystalline GaAs islands on the substrate. All these findings suggest that the surface diffusivity of Ga adatoms is significantly reduced in the T_{gr} range of 450–500 °C and less Ga adatoms arrive at the NW apex. The formation of secondary Ga droplets, in particular, implies that the diffusion length of Ga adatoms on the NW sidewalls is shorter than the length of the NWs, and thus, the Ga adatoms at the bottom of the NWs cannot diffuse to the NW apex. Nevertheless, the NW growth remained fairly directional for the particular total length of the NWs. For example, the axial growth rate was measured to be approximately 60 times faster than the radial growth rate at 450 °C (see Methods). For comparison, the corresponding factor for the conventional MBE at 615 °C was 170.

For a quantitative description of DCAPE at low temperatures, we plotted the ratios of GR_{NW}/GR_{As_4} and GR_{NW}/GR_{Ga} as a function of T_{gr} , as shown in Figure 2b. The monotonic decrease of GR_{NW}/GR_{As_4} from 3.5 at 550 °C to 2.0 at 450 °C suggests that the rebound flux of As_4 is decreased at lower T_{gr} . Apparently, this is because more As_4 is incorporated into the polycrystalline GaAs islands that cover a larger surface area of the substrate at lower T_{gr} . If we assume a perfect matching of arriving Ga and As at the top NW droplets, then we can use the GR_{NW}/GR_{Ga} ratio as an indicator of the surface diffusivity of Ga adatoms on the NW sidewalls (the higher the ratio, the higher the diffusivity). As it is implied from Figure 2b, the Ga adatom diffusivity is reduced at 450 or 500 °C by a factor of 7 or 5, respectively, as compared to 550 °C. The diffusion length of Ga adatoms was estimated by measuring the distance between the top and the secondary Ga droplet on every NW. In particular, the diffusion length was found to be approximately equal to 1.6 μm at 450 °C and 2.4 μm at 500 °C. Thus, the diffusion length at 550 °C is estimated to be approximately 12 μm . The aforementioned values for T_{gr} of 450 and 500 °C correspond to the surface diffusivity of Ga adatoms along the NW {110} sidewalls (because the Ga adatoms on the substrate cannot diffuse to the NW apex), whereas the one at 550 °C corresponds to an overall value that includes both the NW sidewalls and the substrate surface.

The advantages of DCAPE are better highlighted by direct comparison with NWs grown in the conventional MBE mode at 450 °C. The conventional growth was performed on NW templates (like DCAPE) with very low fluxes of Ga and As_4 , which maximized the surface diffusivity of Ga adatoms (see Methods). A V/III ratio as low as 0.9 had to be used to maintain the size of the Ga droplet at the NW apex (any higher value resulted in complete consumption of the Ga droplet), which is indicative of a reduced arrival rate of Ga adatoms diffusing from the NW sidewalls to the Ga droplet at the NW apex. The reduced diffusivity is also evidenced from the analysis of side-view SEM images as the one in Figure 1f. Namely, the axial growth rate was measured to be only 25 times faster than the radial one, while several secondary Ga droplets were formed along the sidewalls of each NW. The regular droplet spacing of 0.6 μm indicates a 3 times shorter diffusion length of Ga

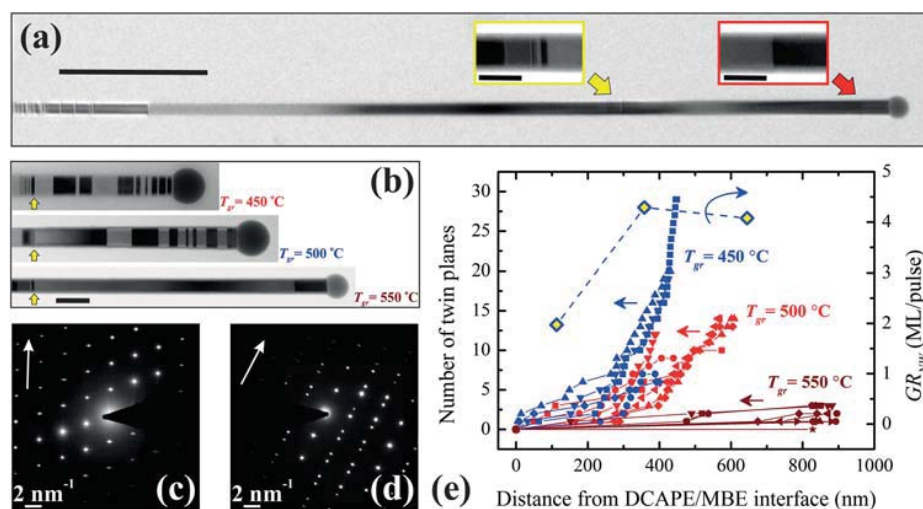


Figure 3. (a) Bright-field side-view TEM image of a GaAs NW grown in DCAPE mode at 550 °C. The yellow arrow indicates the defective segment at the DCAPE/MBE interface (shown magnified in the corresponding inset). The red arrow indicates the only twin plane found inside the DCAPE segment of the particular NW (shown magnified in the corresponding inset). The scale bars correspond to 0.5 μm (main image) and 50 nm (insets). (b) Bright-field side-view TEM images of GaAs NWs grown at three different temperatures in DCAPE mode. The images are close-ups of the DCAPE segment. The yellow arrows indicate the DCAPE/MBE interface. The scale bar corresponds to 100 nm. (c), (d) Representative SAED patterns from (c) pure and (d) twinned zincblende segments of GaAs NWs grown in the DCAPE mode. Both patterns were acquired along the [111] zone axis. The arrows indicate the growth direction. (e) Plot (left axis/single-colored symbols) of the total number of twin planes inside the DCAPE segment of GaAs NWs grown at three different temperatures as a function of the distance from the DCAPE/MBE interface. The axial growth rate of DCAPE GaAs NWs grown at 450 °C is also plotted (right axis/double-colored rhombus) as a function of the length of the DCAPE segment.

adatoms as compared to the DCAPE mode at the same temperature. In agreement with that, the $\text{GR}_{\text{NW}}/\text{GR}_{\text{Ga}}$ ratio was found to be 3 times lower than for DCAPE (Figure 2b).

The structural quality of the DCAPE NWs was evaluated by transmission electron microscopy (TEM). In Figure 3b, representative bright-field images are shown from the DCAPE segment of single NWs grown at different temperatures. The defective segment that is present on the left side of each image is attributed to a transient in the size of Ga droplets at the end of the template growth and the beginning of DCAPE (see Supporting Information for a detailed explanation). Thus, the defective segment serves as a marker of the DCAPE/MBE interface. The actual position of the interface is conventionally set at the top of the defective segment, as indicated by the yellow arrows. For completeness, the full length of one of the NWs ($T_{\text{gr}} = 550$ °C) is shown in Figure 3a, where the yellow arrow shows the DCAPE/MBE interface (magnified in the corresponding inset), whereas the red arrow indicates the only twin plane inside the DCAPE segment (magnified in the corresponding inset) of the particular NW. The defective segment at the bottom of the NW is commonly found in conventionally grown NWs and attributed to local transients of the growth conditions in the beginning of the growth. The crystal structure of the DCAPE NW segments at any T_{gr} was found to be zincblende, but twin planes were also present as evidenced by the sharp changes in the diffraction contrast along the NWs in Figure 3b (each contrast reversal corresponds to one twin plane). Representative selected area electron diffraction (SAED) patterns are shown in Figure 3c,d from DCAPE segments without and with twin planes, respectively.

In Figure 3b, we observe that the DCAPE NWs grown at 550 °C are almost defect-free and their top segment near the Ga droplet does not contain a distinctively higher number of defects, in contrast to what is typically found in conventional

MBE. We also observe that the lower T_{gr} , the higher the total number of twin planes contained in each NW, but again, a distinctively higher number of defects was never detected near the Ga droplets. The spacing of the twin planes is irregular and, in any case, much larger than the NW elongation per As_4 pulse. Thus, the formation of twin planes cannot be attributed to the periodic variation of the growth conditions due to the alternate beam pulsing, which then would have been an inherent limitation of DCAPE.

The total number of twin planes was measured as a function of the distance from the DCAPE/MBE interface for several NWs grown at each temperature and the results are plotted in Figure 3e. The different symbols of the same color correspond to different NWs grown at the same temperature (from the same sample). As found for $T_{\text{gr}} = 450$ and 500 °C, the total number of twin planes increased with the distance from the DCAPE/MBE interface in two modes, that is, with a slow rate during the first 300–400 nm of DCAPE and with a much faster rate afterward. This behavior suggests that the local conditions at the growth interface were not stable throughout the DCAPE stage, but some continuous or transient changes accelerated the twin plane formation. Interestingly, the two-mode increase of the twin plane number density at 450 °C correlates well with the evolution of GR_{NW} at the same temperature, which is also plotted in Figure 3e (GR_{NW} was measured for NWs grown under identical conditions at 450 °C, but with a different total number of identical pulses). The low formation rate of twin planes within the first 300 nm is accompanied by a relatively low GR_{NW} that increases gradually, whereas the higher formation rate for the rest of the growth is accompanied by a higher GR_{NW} that is stabilized at approximately 4 monolayers (MLs) per As_4 pulse (or equivalently 1.4 ML/s). The correlation between the twin plane formation rate and the axial growth rate was also confirmed for NWs grown at $T_{\text{gr}} =$

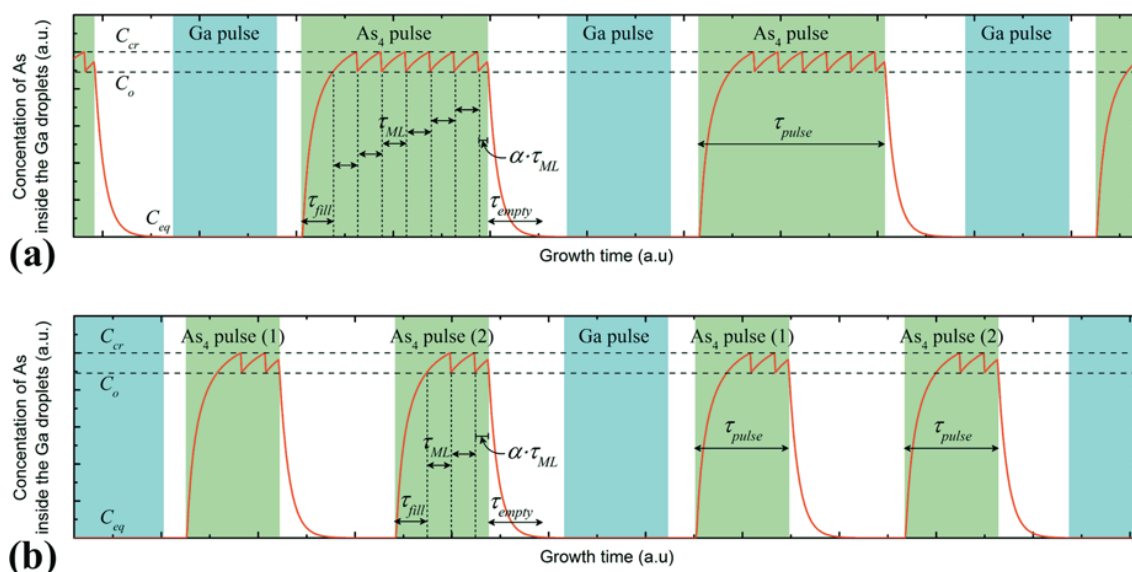


Figure 4. (a) Schematic plot of the As concentration inside the Ga droplets during the DCAPE pulse sequence. The characteristic time constants are indicated. (b) Same plot for an alternative pulse sequence, where each As₄ pulse in (a) has been split into two equal pulses of half duration. Constant α depends on the As₄ pulse duration (τ_{pulse}); thus, it is different in (a) and (b).

500 °C, where the onset of the higher formation rate was shifted from 400 nm to more than 500 nm after decreasing the GR_{NW} deliberately from 1.7 ML/s to 0.6 ML/s (see [Supporting Information](#)).

The initial increase and the subsequent stabilization of GR_{NW} during the DCAPE stage ([Figure 3e](#)) can be understood as a result of the imperfect matching of the Ga and As arrival fluxes at the Ga droplet. That is, more Ga than As arrives at the Ga droplet, causing a gradual increase of the Ga droplet size with time. Being proportional to the droplet size (for fixed NW diameter and growth conditions), the axial growth rate also increases with time correspondingly. Eventually, the size of the Ga droplet and the axial growth rate will both reach a new equilibrium due to either the increasing diameter (tapering) of the NW or the increasing collection of the rebound flux of As₄ from the growing Ga droplet.^{32,33} At this point, it is not conclusive whether the formation rate of twin planes is directly controlled by the axial growth rate or the Ga droplet size (or contact angle), but further investigations are currently in progress and the results will be published elsewhere. After all, the small number of twin planes at the beginning of the DCAPE clearly manifests the possibility to grow defect-free NWs at temperatures as low as 450 °C, if the Ga and As₄ fluxes are well-tuned.

The ability in DCAPE for defect-free growth interruptions in combination with very low axial growth rates (in the range of 1–10 MLs of GaAs per As₄ pulse) gave us the opportunity to design experiments dedicated to the investigation of the growth mechanisms. In specific, we designed a series of experiments to probe the dynamics of the As population inside the Ga droplets. As discussed in detail by ref 34, the concentration of As inside the Ga droplet fluctuates periodically during growth between two characteristic values. We adapt this picture for the case of DCAPE, as it is schematically plotted in [Figure 4a](#). At this stage, we use it only as a deterministic approximation of the stochastic nucleation process. The high value C_{cr} is the critical concentration that is needed to overcome the nucleation barrier at the top facet of the NW, whereas the low value C_0 is the

reduced concentration after the growth of each ML of GaAs. Four characteristic time constants are employed to describe the dynamics of the As population inside the Ga droplet: τ_{fill} is the time needed to fill the Ga droplet with As to a concentration of C_0 at the onset of each As₄ pulse; τ_{ML} is the time needed to increase the As concentration from C_0 to C_{cr} inside the Ga droplet after every single nucleation event; τ_{pulse} is the total time duration of each As₄ pulse; τ_{empty} is the time needed to evaporate the excess As from the Ga droplet and restore its equilibrium concentration C_{eq} for the given temperature immediately after each As₄ pulse. For simplicity, we assume that τ_{empty} is shorter than the time-spacing between sequential As₄ pulses. The parameter α ($0 < \alpha < 1$) represents the time as a fraction of τ_{ML} from the completion of the last ML in a pulse until the end of that pulse (α depends on all the aforementioned time constants except τ_{empty}). Even though a growth mode with single nucleation events (mononucleation mode) is generally considered,^{35,36} our analysis is not bound to that because the definition of τ_{ML} also holds for any polynucleation mode.

A distinct feature of DCAPE is the filling of the Ga droplet with As up to a concentration of C_0 at the onset of every single As₄ pulse. The amount of As₄ used for that purpose does not contribute directly to the axial growth of the NWs, but rather re-evaporates from the droplet at the end of each As₄ pulse. Typically, the overall amount of As₄ (N_{rf}) used for the recurring filling of the droplets is considerably large because of the large number (N_{pulse}) of short As₄ pulses used for the growth ($N_{rf} = N_{pulse} \times \tau_{fill}/\tau_{ML}$), given in an equivalent number of GaAs MLs). This is unlike the conventional MBE, where only one filling of the droplets at the beginning of the growth is needed.

For the experimental determination of τ_{fill} and τ_{ML} , we have designed a complex pulse sequence, where the As₄ pulses of a reference sequence are subdivided into multiple equal pulses, keeping the total time of As₄ supply ($N_{pulse} \times \tau_{pulse}$) always constant. By increasing N_{pulse} and decreasing τ_{pulse} accordingly in different DCAPE experiments, we have been able to increase the number of recurring fillings of Ga droplets with As and,

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consequently, increase N_{rf} . An example is shown in Figure 4, where each As_4 pulse of the reference sequence in (a) is subdivided into two pulses in (b) with half the duration of the reference pulse each. The number and duration of Ga pulses were kept unchanged in order to ensure identical growth conditions for all experiments (same V/III ratio and supplied Ga per pulse). The increase of N_{rf} can be measured as a corresponding decrease of GR_{NW} according to the following formula (expressed in MLs per unit of time; see derivation in Supporting Information)

$$\text{GR}_{\text{NW}} = \frac{1}{\tau_{\text{ML}}} - \left(\frac{\tau_{\text{fill}}}{\tau_{\text{ML}}} + \alpha \right) \frac{1}{\tau_{\text{pulse}}} \quad (1)$$

with the precondition that $N_{\text{pulse}} \times \tau_{\text{pulse}} = \text{constant}$. The dependence of GR_{NW} on τ_{pulse} was determined experimentally for $T_{\text{gr}} = 450$ and 500 °C as plotted in Figure 5. In agreement

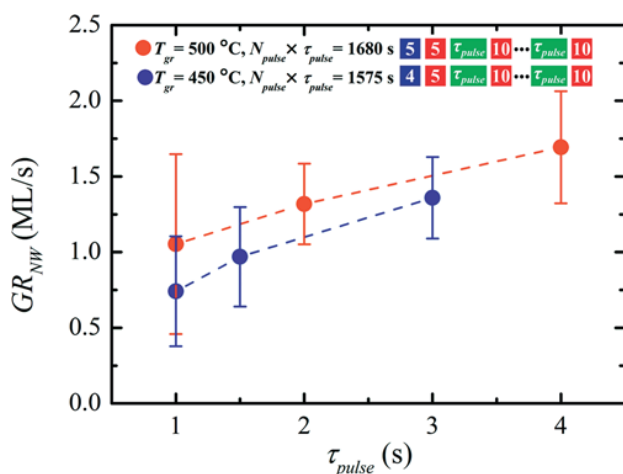


Figure 5. Plot of the axial growth rate of GaAs NWs during the DCAPE stage (GR_{NW}) as a function of the As_4 pulse duration (τ_{pulse}). The total time of As_4 supply ($N_{\text{pulse}} \times \tau_{\text{pulse}}$) is constant at every growth temperature (T_{gr}). The differences between the two data sets are indicated. The pulse sequence is described with the colored boxes (see explanation in Figure 1) and reads from left to right.

with our expectations and eq 1, GR_{NW} was found to decrease monotonically with decreasing τ_{pulse} , confirming the depletion of As in the Ga droplets after every As_4 pulse. We presume that the equilibrium concentration of As inside the Ga droplets (which is in the order of 0.01–0.10% for solid GaAs in equilibrium with Ga-rich liquid at 450–500 °C³⁷) is restored after every As_4 pulse.

Equation 1 contains three unknowns (τ_{fill} , τ_{ML} , and α) and can be solved graphically. This is shown in Figure 6a,b for $T_{\text{gr}} = 450$ and 500 °C, respectively, where τ_{fill} is plotted as a function of τ_{ML} . Each gray-colored area corresponds to a different value of τ_{pulse} , whereas α is allowed to take any value between 0 and 1. Because τ_{fill} and τ_{ML} should be independent of τ_{pulse} , the set of all possible solutions is given by the purple-colored intersection. That is, τ_{ML} is found in the range of 0.5–0.7 s, whereas τ_{fill} is estimated with less accuracy (due to the dependence on the unknown α) to be shorter than 0.5 s. Because As_4 flux was practically the same for all experiments, we believe that the slightly longer τ_{ML} at lower T_{gr} is due to the lower rebound flux of As_4 , as discussed previously. In any case, the relatively long τ_{ML} for the given Ga and As_4 fluxes in combination with the

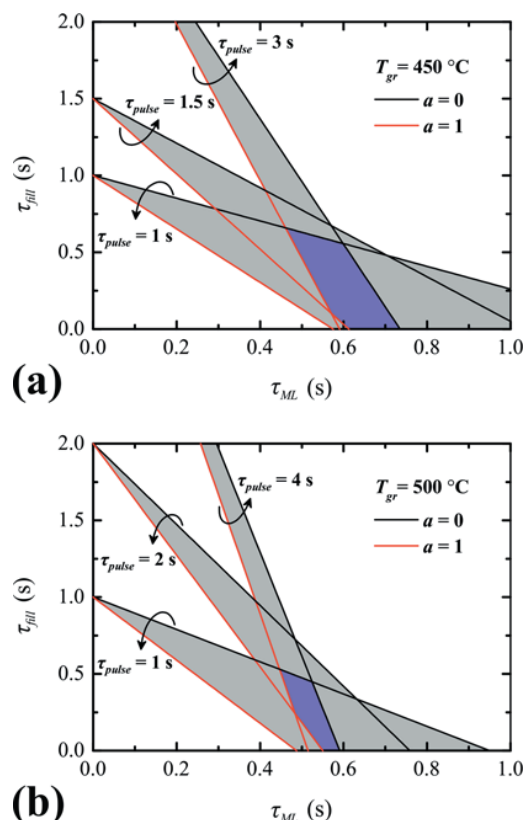


Figure 6. Plots of the characteristic filling time of the Ga droplet with As (τ_{fill}) as a function of the characteristic time-spacing between sequential ML formations (τ_{ML}) for any value of α and for three different As_4 pulse durations (τ_{pulse}). The set of possible solutions is given by the purple-colored intersection of the gray-colored areas. The growth temperature (T_{gr}) was (a) 450 °C and (b) 500 °C.

negligible shut-off transient of As_4 flux in DCAPE makes it possible to control the axial NW growth with high accuracy.

Having good control over the axial growth of NWs is especially important for axial quantum heterostructures, where the length of the various NW segments need to be ultimately controlled in the single-monolayer scale. The resolution of our control is given by the shortest As_4 pulses in Figure 5 ($\tau_{\text{pulse}} = 1$ s), which resulted in growth of 0.74 ± 0.36 MLs at 450 °C and 1.05 ± 0.59 MLs at 500 °C (the error bars represent the standard deviation of the GR_{NW} measurements). Those values are in good agreement with the theoretical minimum and ultimate resolution of axial growth rate, that is, 1 ML per As_4 pulse. Definitely, we cannot rule out possible deviations (e.g., formation of 0, 2, or more MLs in some As_4 pulses) due to the stochastic nature of nucleation. However, the fact that the GR_{NW} standard deviation is clearly lower than 1 ML per As_4 pulse suggests that the effect of stochasticity is limited and the nucleation can be well approximated by the deterministic model in Figure 4. In fact, it was possible to further reduce the GR_{NW} standard deviation (to 0.10 MLs per As_4 pulse) by repeating the experiment with $\tau_{\text{pulse}} = 1$ s at 500 °C using template NWs with an exceptionally narrow length distribution (± 24 nm instead of ± 107 nm for the experiments in Figure 5).

In conclusion, we have investigated the DCAPE of self-catalyzed GaAs NWs on Si(111) and demonstrated the possibility to obtain defect-free zincblende NWs at temperatures as low as 450 °C. Slow axial growth rates and defect-free

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NWs in spite of growth interruptions were proven to be essential advantages of this growth technique. These features have enabled experiments allowing for a better understanding of the growth mechanisms. Therefore, we anticipate that DCAPE will lead to considerably improved epitaxial growth of axial heterostructures with the further advantage of compatibility with fully processed Si-CMOS platforms for monolithic integration. For the latter, dedicated investigations of the nucleation stage in DCAPE will be also needed.

Methods. Growth and Analysis. All samples were grown by solid source MBE using atomic Ga and molecular As₄ beams. The growth temperature T_{gr} refers to the temperature measured on the Si(111) substrate by an optical pyrometer before the initiation of the growth. First, the template NWs were grown in conventional MBE mode (simultaneous supply of Ga and As₄ beams) at 615 °C with a V/III flux ratio of 13. At the end of the template growth, both Ga and As₄ beams were interrupted and the substrate temperature was decreased for the continuation of the growth in DCAPE mode (alternate pulsed supply of Ga and As₄ beams). Mechanical shutters were used for the creation of beam pulses, and a needle valve was employed in addition for the As₄ pulses. The background pressure in the growth chamber was in the 10⁻¹⁰ Torr range during the deposition of Ga. All length and diameter measurements were done by SEM. The diameter was measured at the middle of the NWs, that is, outside the DCAPE segment. Special care was taken to eliminate the image drift during the SEM measurements, allowing for a precise determination of NW lengths. The axial (radial) growth rate of NWs during DCAPE was determined by measuring the final NW length (diameter) after DCAPE, subtracting the measured length (diameter) of template NWs, and dividing by the total time of As₄ supply during DCAPE. For each data point in Figures 2 and 5, a statistical sample consisting of 40–80 NWs was analyzed. The plotted values and the error bars of GR_{NW} correspond to the median value and the standard deviation, respectively.

DCAPE Details. The total amount of supplied Ga and As₄ was kept constant only in the sample series of Figure 2b and Figure 5. For the sample series in Figure 1, the As₄ (Ga) supply per pulse was 1.39 (0.72) ML in (b), 1.39 (0.12) ML in (c), 0.97 (0.30) ML in (d), 0.75 (0.24) ML in (e). For the NWs grown in conventional MBE mode, the As₄ (Ga) flux was equal to 1.61 (0.12) ML/s at 615 °C in (a) and 0.25 (0.28) ML/s at 450 °C in (f). At 450 °C, we chose to use the lowest possible As₄ flux within the practical limits of our experiments in order to maximize the surface diffusivity of Ga adatoms.²⁶ For the sample series in Figure 2a, the Ga supply was varied from 0.12 to 0.72 ML per pulse, with a constant As₄ supply of 1.39 ML per pulse. For the sample series in Figure 2b, the As₄ (Ga) supply per pulse was 0.75 (0.24) ML at 450 °C, 0.97 (0.30) ML at 500 °C, 1.39 (0.12) ML at 550 °C. For the sample series in Figure 5, the As₄ (Ga) flux was 0.25 (0.06) ML/s at 450 °C and 0.24 (0.06) ML/s at 500 °C.

■ ASSOCIATED CONTENT

§ Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.nanolett.6b00527.

Analytical description of the defective segment at the DCAPE/MBE interface and comparison of the twin plane distribution (as measured by TEM) in DCAPE

GaAs NWs grown at 500 °C with two different axial growth rates. Derivation of eq 1 for the axial growth rate during DCAPE. (PDF)

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Notes

The authors declare no competing financial interest.

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Carbon : nickel nanocomposite templates – predefined stable catalysts for diameter-controlled growth of single-walled carbon nanotubes†

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Carbon : nickel (C : Ni) nanocomposite templates (NCTs) were used as catalyst precursors for diameter-controlled growth of single-walled carbon nanotubes (SWCNTs) by chemical vapor deposition (CVD). Two NCT types of 2 nm thickness were prepared by ion beam co-sputtering without (type I) or with assisting Ar⁺ ion irradiation (type II). NCT type I comprised Ni-rich nanoparticles (NPs) with defined diameter in an amorphous carbon matrix, while NCT type II was a homogenous C : Ni film. Based on the Raman spectra of more than 600 individual SWCNTs, the diameter distribution obtained from both types of NCT was determined. SWCNTs with a selective, monomodal diameter distribution are obtained from NCT type I. About 50% of the SWCNTs have a diameter of (1.36 ± 0.10) nm. In contrast to NCT type I, SWCNTs with a non-selective, relatively homogeneous diameter distribution from 0.80 to 1.40 nm covering 88% of all SWCNTs are obtained from NCT type II. From both catalyst templates predominantly separated as-grown SWCNTs are obtained. They are free of solvents or surfactants, exhibit a low degree of bundling and contain negligible amounts of MWCNTs. The study demonstrates the advantage of predefined catalysts for diameter-controlled SWCNT synthesis in comparison to *in situ* formed catalysts.

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Introduction

Carbon nanotubes have been known for more than 20 years¹ and since the very first reports on their synthesis fascinating electronic properties of CNTs were predicted. It was expected that they will be used in many fields of molecular electronics, viz. in transistors,^{2–4} medical devices,^{5,6} chemical and biochemical sensors^{7,8} etc. However, in currently commercialized applications CNTs are mainly used as additives to improve the mechanical, thermal, and electrical properties of bulk materials (resins, plastics, polymers).⁹ Semiconducting SWCNTs are well suited for transistors because of their low electron scattering and small bandgap.¹⁰ For “high-tech” applications, however, precisely-defined electronic properties are needed. This requires eventually one specific diameter and chiral angle of SWCNTs.^{11,12} There are several methods of

CNTs synthesis,^{13–15} and among them CVD is the most versatile and most easily scalable one. The structural and morphological properties of CVD-grown CNTs are affected by many process parameters,¹⁵ such as the carbon precursor nature and concentration, carrier gas pressure and composition, temperature, time, catalyst nature and size, etc. There are many reports that catalyst particles must have a suitable size in order to be active for CNT growth and that the control of the catalyst nanoparticles before and during CVD is very important in order to control CNT yield, diameter and chirality. The most used catalysts for CNT growth are Fe,^{16–31} Co,^{24,32–45} Ni^{17,23,46–53} and their bimetallic alloys.^{38,42,54–56} The CNT synthesis on these catalysts may produce SWCNTs with narrow diameter distribution and few chiralities depending on the catalyst nanoparticle size.^{16,20,21,25,26,32,35,41,51,54}

In almost all studies, the catalysts are prepared *in situ* by an initial annealing step of the precursor prior to CVD or by precursor injection during the CVD process. The control of catalyst size and shape depends on many process parameters in a complex manner, whose optimization often requires numerous iterations.²⁷ The process could be simplified by introducing catalyst templates with predefined NP size and shape, which exhibit sufficient stability against coalescence and grain coarsening. Carbon : nickel nanocomposite (C : Ni NC) thin films of 10 nm to 100 nm thickness have been grown in a large variety of microstructures. This was achieved by variation of

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deposition temperature (RT to 500 °C) and Ni concentration (5 to 30 at% Ni) during thin film growth without and with irradiation by an assisting low-energy Ar⁺ beam. So far, C:Ni NCs were reported with NP diameters from 1.5 nm to 17 nm, spherical and columnar shapes, and in regular or stochastic three-dimensional particle arrangements.^{57–59} In our previous work on SWCNT growth⁴⁸ we reported the successful growth of individual SWCNTs with a Gaussian-like, non-selective diameter distribution of (1.6 ± 0.4) nm from 10 nm thick templates consisting of columnar Ni NPs of (4.0 ± 1.0) nm diameter before the CVD growth process.

In this work, the SWCNT diameter distributions obtained by CVD from two 2 nm thick C:Ni NC template types are compared. NCT type I comprises predefined catalyst NPs, while for NCT type II the catalyst NPs are formed *in situ* during the CVD process. This approach enables a direct comparison of the SWCNT growth from pre-defined and *in situ* formed catalyst particles. Transmission electron microscopy (TEM) and scanning electron microscopy (SEM) were applied to characterize the NCTs before and after the CVD process and to determine the size distribution of the catalyst particles. SWCNT diameter distribution and the presence of multi-walled CNTs were analysed by SEM and Raman spectroscopy. It is demonstrated that SWCNT diameters strongly depend on the NCT type applied for CVD synthesis. Using NCT type I about 50% of the SWCNTs have a diameter of (1.36 ± 0.10) nm, while SWCNTs with a relatively homogeneous diameter distribution from 0.8 nm to 1.4 nm are obtained from NCT type II. Furthermore it is shown that MWCNT formation, SWCNT bundling, and catalyst NP coarsening are successfully suppressed. Finally, the possibilities and limits of SWCNT diameter control by defining the catalyst NP size are discussed.

Experimental

The NCT type I (C:Ni, ~15 at% Ni) was deposited at 300 °C by ion beam co-sputtering of a graphite/Ni zone target onto a Si/SiO₂ (~0.5 μm, thermally oxidized) substrate. Template type II (C:Ni, ~8 at% Ni) was prepared at RT by irradiating the growing C:Ni film with an oblique incidence angle (~60°) assisting Ar⁺ beam of 130 eV ion energy. The sputter ion source (Kaufmann type, IonTech Inc., Fort Collins, USA) was operated with an Ar⁺ ion energy of 1 keV and a beam current of 40 mA, the assisting ion source (Kaufmann type, ISQ40KF, Ion-Tech GmbH, Wüstenbrand, Germany) with Ar⁺ energies of 50 to 130 eV and a total beam current of 9 mA. The base pressure in the deposition chamber was $(1.3 \pm 0.3) \times 10^{-5}$ Pa, the working pressure was about 10^{-2} Pa, and the deposition time was 3 to 4 min. The given film composition was extrapolated from the Rutherford Backscattering analysis of our previous work.^{57–59} Further deposition details are described *ibid.*

The subsequent CVD process consists of two consecutive steps: the NCT pre-treatment and the CNT growth. For this, the C:Ni templates were placed in the CVD oven (Black Magic, Aixtron) and exposed to flowing air (flow rate 200 sccm,

pressure 2 to 3 mbar) for 10 min at 550 °C. After 10 min the reactor was evacuated, the temperature was increased to 735 °C, and the samples were exposed to a mixture of C₂H₂/H₂/Ar (5/300/300 sccm; total pressure 5.1 mbar) for ~20 min.

Cross-sectional transmission electron microscopy images of C:Ni thin films were obtained using an image Cs-corrected Titan 80–300 (FEI) microscope operated at an acceleration voltage of 300 kV. TEM specimens of the C:Ni templates before CNT growth were prepared by sawing, grinding, dimpling, and final Ar ion milling. TEM lamella preparation of the C:Ni templates after pre-treatment and CNT growth was done by *in situ* lift-out using a Zeiss Crossbeam NVision 40 system. To protect the sensitive surface, a carbon cap layer was deposited beginning with electron beam assisted and subsequently followed by Ga focused ion beam (FIB) assisted precursor decomposition. Afterwards, the TEM lamella was prepared using a 30 keV Ga FIB with adapted currents. Its transfer to a 3 post copper lift-out grid (Omniprobe) was done with a Kleindiek micromanipulator. To minimize sidewall damage, Ga ions of only 5 keV energy were used for final thinning of the TEM lamella to electron transparency. SEM of the NCTs before and after CVD growth of the CNTs was performed by detecting secondary electrons using a S-4800 microscope (Hitachi) operated at an accelerating voltage of 10 kV. Raman maps of randomly selected sample areas of $20 \times 20 \mu\text{m}^2$ were recorded with a step width of 1.25 μm using a micro-Raman Labram HR spectrometer, which is equipped with a holographic 1800 lines per mm grating and a liquid nitrogen cooled CCD detector (Horiba-Jobin-Yvon). The laser wavelengths of 532 nm and 632.8 nm, corresponding to photon energies of 2.33 eV and 1.96 eV, respectively, were used for excitation. The obtained Raman spectra were fitted using an in-house software routine based on a Levenberg-Marquardt least-square algorithm. SWCNT diameter distributions were calculated by using the relation $d_{\text{SWCNT}} = 232 \text{ cm}^{-1} \text{ nm} / \nu_{\text{RBM}}$,⁶⁰ wherein ν_{RBM} represents the Raman shift of the radial breathing mode of SWCNTs, and by sorting the obtained diameter values into diameter intervals of 0.05 nm width.

Results

The microstructure of the original NCT type I is heterogeneous and consists of Ni-rich particles embedded in an amorphous carbon (a-C) matrix (Fig. 1). Cross-sectional TEM analysis gives a mean NP diameter of (2.0 ± 0.3) nm, which corresponds to the average NCT thickness. The NPs appear to be well-aligned with a mean NP center to center distance of (4.3 ± 0.3) nm. Lattice fringes were resolved for approximately 40% of 27 NPs analyzed in detail (Fig. 1b and c). Fast Fourier Transform (FFT) analysis gave lattice spacings grouped into (a) 0.22 nm to 0.24 nm and (b) 0.20 to 0.21 nm. In one case crossed lattice fringes with spacings of 0.21 nm and 0.15 nm enclosing an angle of 46° were observed. The latter values are in good agreement with reference data of the (006) and {116} planes of rhombohedral Ni₃C (rh-Ni₃C), which are 0.215 nm, 0.156 nm,



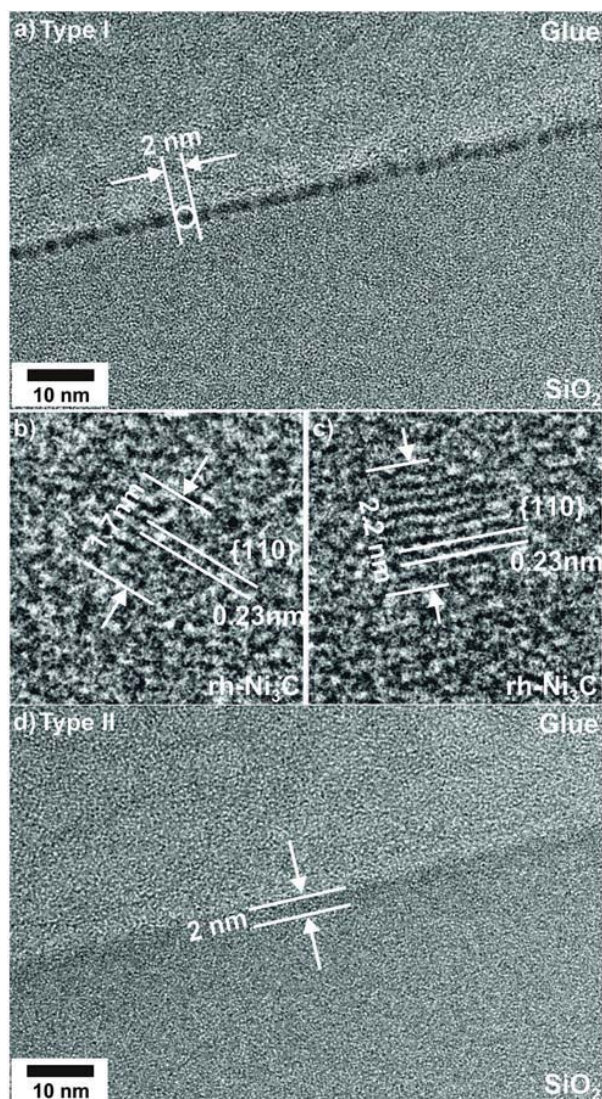


Fig. 1 Cross-sectional TEM images of C : Ni templates before the CVD process of CNTs: (a)–(c) NCT type I; (d) NCT type II. Images (b) and (c) are enlarged views of micrograph (a) showing selected crystalline Ni_3C NPs in NCT type I.

and 43.4° .⁶¹ Moreover, the group (a) lattice fringe spacings are indicative for rh- Ni_3C , namely for its $\{110\}$ planes.⁶¹ Group (b) spacings could be assigned either to rh- Ni_3C or to fcc-Ni. According to these findings, previous own work, and the literature on this material system, the initial Ni-rich phase of NCT type I is assigned to rh- Ni_3C .^{57,62–64}

The cross-sectional TEM image of NCT type II (Fig. 1d) shows the formation of a Ni-enriched layer appearing darker than the SiO_2 substrate and the resin glue on top of the film, which is used for TEM specimen preparation. The layer thickness is 2 nm and hence the same as for the NCT type I. In contrast to NCT type I, no NP formation was detected for NCT type II (Fig. 1d). The homogenous microstructure of C : Ni NCT type II is attributed to the hyperthermal particle and energy flux of

the irradiating Ar^+ ions. It causes ion-enhanced mixing and partial re-sputtering of Ni, which lead to a homogenous distribution of the metal in the a-C matrix.

After CVD growth of CNTs the morphology of NCT I consists of individual NPs as before the CVD process (Fig. 2).

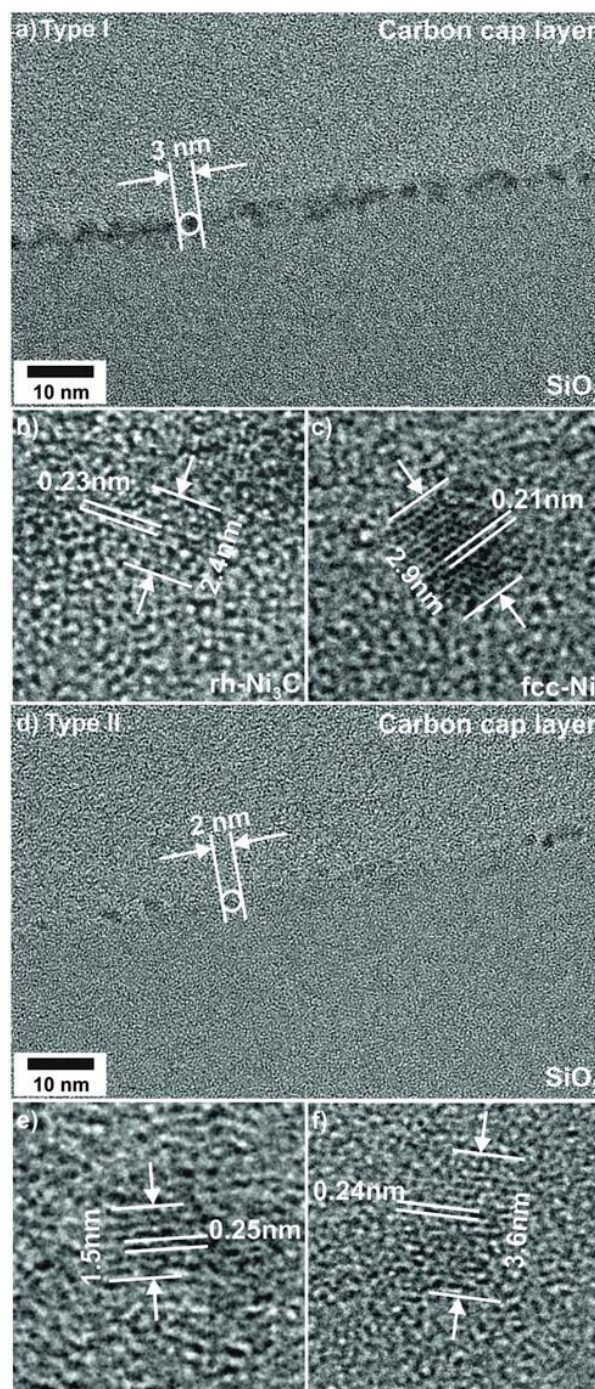


Fig. 2 Cross-sectional TEM images of C : Ni templates after CVD growth of CNTs: (a)–(c) NCT type I; (d)–(f) NCT type II. Images (b) and (c) [(e) and (f)] show selected crystalline NPs in NCT type I [NCT type II].



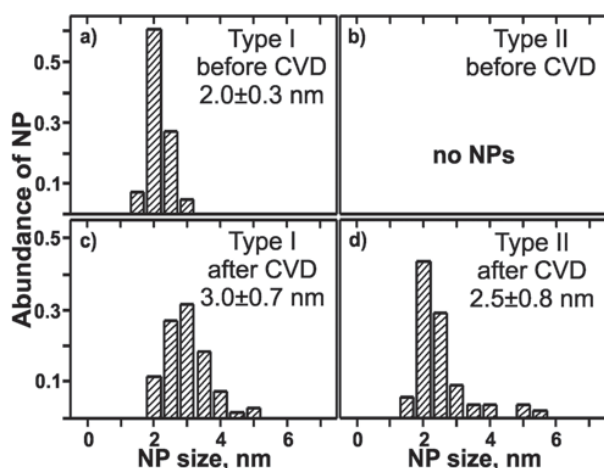


Fig. 3 Normalized size distributions of Ni-rich NPs measured by TEM before and after the CVD process, respectively. NCT type I, (a) and (c), NCT type II, (b) and (d).

Cross-sectional TEM revealed moderate NP coarsening accompanied by broadening of the diameter distribution from initially (2.0 ± 0.3) nm to (3.0 ± 0.7) nm (Fig. 3). The NPs are less aligned and the NCT surface appears rougher than before the CVD process. Lattice fringes were observed for 72% out of 29 NPs studied in detail. FFT analysis revealed the coexistence of Ni-rich NPs with hcp (Fig. 2b) and fcc (Fig. 2c) lattice structure in an approximately equal ratio. The details of this analysis are provided in the ESI.† The FFT of the nanocrystal in Fig. 2b corresponds very closely to a $[1\bar{2}1\bar{3}]$ zone axis image of hcp-Ni (for details see ESI, Fig. 1†).⁶⁵ It has the same Ni sublattice structure as rh-Ni₃C. The nanocrystal displayed in Fig. 2c has fcc crystal structure. Its FFT corresponds to a $[011]$ zone axis image of fcc-Ni (for details see ESI, Fig. 2†).⁶⁵ One out of 29 FFT patterns showed lattice fringes with spacings of 0.22 nm and 0.25 nm enclosing an angle of 56° . One possible assignment would be fcc-NiO, although the observed spacings were enlarged by about 4% compared to the reference.⁶¹ Alternatively, this FFT pattern might be described by Ni₃C $\{110\}$ and $\{014\}$ lattice fringes, whose spacings are 0.22 nm and 0.25 nm with an interplanar angle of 56.7° .

In summary, approximately 50% of NCT type I NPs changed their lattice structure from rh-Ni₃C to fcc-Ni. For another approximately 50% the observed hcp-Ni lattice structure points to the conservation of the initial rh-Ni₃C phase structure. The formation of fcc-NiO could not be verified. Finally it should be noted that TEM analysis is not suitable to provide information about the structure of the a-C matrix.

The morphology of NCT type II, where initially only a contrast enhancement with respect to SiO₂ substrate and resin glue could be detected, was completely changed during the CVD process. The initial homogenous and 2 nm thick Ni-enriched layer was no more detected. Instead, NCT type II is characterized by the coexistence of ranges without (major fraction) and with (minor fraction) separated NPs after the CVD

(Fig. 2d). Their average diameter is (2.5 ± 0.8) nm (Fig. 3). The smallest resolved NP had a diameter of 1.5 nm. Two dominant lattice spacings were identified by FFT analysis, namely 0.21 nm and 0.25 nm. Their assignment to rh-Ni₃C, fcc-Ni, or fcc-NiO is ambiguous as outlined in the previous sections, and supporting interplanar angles could not be determined. The NP formation from NCT type II can be considered as an example for the SWCNT growth using catalysts, which are formed *in situ* during the process of pre-treatment and CVD. The post-CVD NP diameter distribution of NCT type I is narrower than that of NCT type II. This difference can be expressed as relative deviation from the mean NP diameter values. It is 23% for NCT type I and 32% for NCT type II. Moreover, compared to NCT type II, a larger fraction of NCT type I NPs showed lattice fringes. This is a hint for a larger degree of NP crystallinity in NCT type I than in type II after the CVD growth of CNTs.

The TEM results show that the microstructure of the pre-defined NCT type I is largely preserved during the CNT growth process. Its mean NP diameter grew by about 1 nm (*ca.* 50%), and the fraction of crystalline NPs within the template increased. In contrast to that NCT type II underwent a complete re-structuring from a homogenous film into a film of NPs. The NP size distribution for NCT type II after CVD is sufficiently narrow and can be compared with the one obtained from metallic thin films. As an example, pre-annealing of a 0.22 nm-thin Ni film produced NPs with diameter of (4.4 ± 0.6) nm.⁵²

In contrast to the featureless appearance before nanotube growth, SEM images of the NCT samples measured after the CVD process display three types of features: (i) randomly distributed spherical spots of less than 10 nm in diameter, (ii) bent objects of about 80 nm length, and (iii) blurred objects of up to 3 μ m length (Fig. 4). These features are assigned to metallic nanoparticles, multi-walled CNTs (MWCNTs), and single-

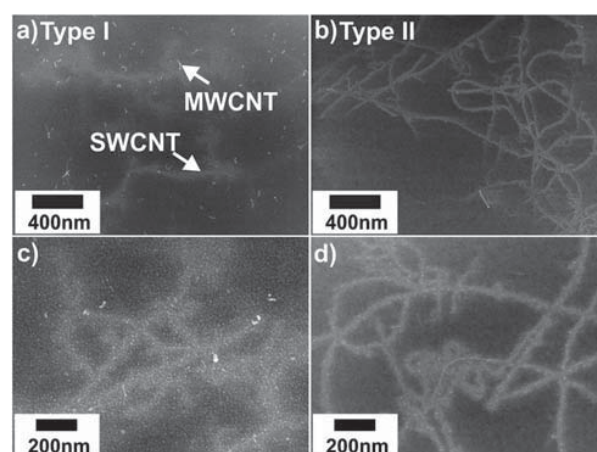


Fig. 4 SEM images of C:Ni templates after CVD growth of CNTs: (a), (c) NCT type I; (b), (d) NCT type II. Compared to the few MWCNTs the individual SWCNTs appear blurred and diffuse due to charging effects and probably also due to thermally-induced vibrations.



walled CNTs, respectively. The NP diameters are apparently larger for NCT type I than for type II. The amount of MWCNTs is generally very low. In NCT type II samples MWCNTs are virtually absent. The statistical analysis of the SEM images points to a similar efficiency of both types in catalyzing SWCNT growth with yields of ~ 5 to 7 tubes per μm^2 .

Raman spectroscopy enables a non-destructive and simple determination of the SWCNT diameter because of the inverse proportionality of diameter and radial breathing mode (RBM) frequency. Hence, the diameter distribution of SWCNTs in the sample can be conveniently elucidated by measuring Raman spectra with two or more excitation laser wavelengths, for example, 532 nm and 632.8 nm. This approach allows detecting SWCNTs in the diameter range from 0.6 nm to 2.3 nm. The reliability of the SWCNT diameter distribution obtained by this method was confirmed by parallel Raman and High-Resolution TEM studies in the literature.^{16,37}

Typical Raman spectra in the range of the RBM obtained during 2D Raman mapping with two excitation wavelengths for NCT types I and II are shown in Fig. 5a and b. The vast majority of the spectra contains only one single RBM line. In total, the Raman spectra of more than 600 individual SWCNTs were analysed in this study. The RBM line widths for SWCNTs grown with both template types cover the range of 4 cm^{-1} to 8 cm^{-1} . They decrease with increasing RBM frequency, similar to the results reported previously in the literature.⁶⁶ The narrow line widths point to long lifetimes of the excited RBM modes, and indicate a high degree of structural order and a negligible contribution of defect-induced phonon decay processes. Such a behavior is consistent with the formation of individual SWCNTs with a low amount of structural defects.⁶⁷ In order to analyze the abundance of SWCNTs, the observed RBM frequencies were assigned to frequency intervals of 10 cm^{-1} width and subsequently summed. The analysis shows

a diameter-selective SWCNT formation for NCT type I: 50% of all observed tubes have RBM frequencies from 160 cm^{-1} to 180 cm^{-1} (Fig. 5c). In contrast to that, a broad distribution of RBM frequencies is found for NCT type II (Fig. 5d). The corresponding diameter distributions of SWCNTs (normalized to 1) are shown in Fig. 6. The SWCNT diameter distribution depends strongly on the template type. The diameter distribution of NCT type I reveals an astonishingly high selectivity for SWCNTs of the diameter interval centered at $(1.360 \pm 0.025)\text{ nm}$, which includes about 33% of the SWCNTs (Fig. 6a). Moreover, 51% of all synthesized SWCNTs fall into a narrow diameter range of 1.26 nm to 1.46 nm.

Applying the formula for the nanotube diameter d ,

$$d = (a_0/\pi)\sqrt{n^2 + nm + m^2},$$

with the lattice constant $a_0 = 0.246\text{ nm}$ and the chiral indices n and m , in total 9 SWCNT chiralities can be expected in the diameter range of $(1.360 \pm 0.025)\text{ nm}$. Two of them are metallic, 7 are semiconducting. The analysis of the electronic transition energies of these 9 chiralities shows that only the E_{33} electronic transition of semiconducting SWCNTs is in resonance with the laser energy of 2.33 eV.⁶⁸ This criterion requires that the difference between laser and electronic transition energy is $\Delta E \leq \pm 0.10\text{ eV}$.⁶⁹ The combination of these two informations reduces the chiralities preferentially observed in this study to three: (12,8), (17,1), and (16,3).

The proposed diameter selectivity is further supported by the following findings of the Raman analysis. The next thinner SWCNTs, which are expected to be in resonance with laser radiation of 532 nm, would be (13,6), (14,4) and (15,2) with diameters of 1.32 nm, 1.28 nm and 1.26 nm and RBM frequencies of 175 to 185 cm^{-1} , respectively. Their abundance is only of the order of 5% (Fig. 6). At slightly larger diameters than $(1.360 \pm 0.025)\text{ nm}$ the SWCNTs (15,5), (11,10) and (14,7) with 1.41 nm, 1.43 nm and 1.45 nm should resonate with 532 nm laser excitation. The abundance of this fraction is about 7%. That means that the low and high limit of the SWCNT diameter distribution maximum is precisely defined for NCT type I.

It can be further concluded that semiconducting tubes with diameters of $\sim 1.6\text{ nm}$ and RBM frequencies of around

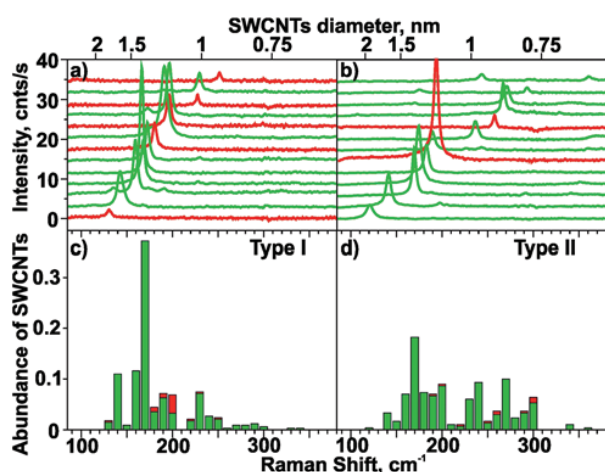


Fig. 5 Selected RBM range Raman spectra (a, b) and RBM distributions (c, d) of SWCNTs formed from C : Ni templates: (a), (c) NCT type I; (b), (d) NCT type II. The color code represents spectra measured with 532 nm (green) and 632.8 nm (red) laser excitation, respectively.

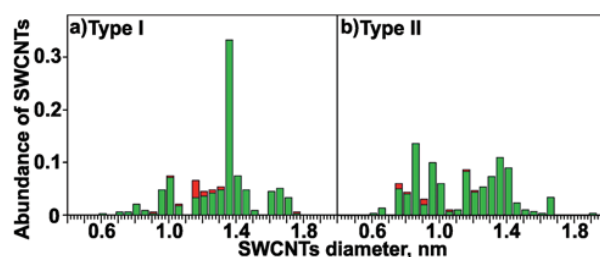


Fig. 6 Normalized diameter distributions of SWCNTs synthesized from C : Ni templates as derived from the abundance of observed RBM lines in the corresponding diameter intervals: (a) NCT type I; (b) NCT type II. The color code represents data obtained with 532 nm (green) and 632.8 nm (red) laser excitation, respectively.



140 cm^{-1} contribute only to a minor extent to the observed diameter distribution (Fig. 6). Finally, for 632.8 nm laser excitation, the most abundant SWCNTs have RBM frequencies of around $\sim 200 \text{ cm}^{-1}$ for NCT type I. They belong to metallic tubes with a diameter of 1.2 nm, whose M_{11} transition is in resonance with the laser energy of 1.96 eV. These metallic tubes cause the local maximum in the diameter distribution for 632.8 nm laser excitation, but their contribution to the overall SWCNT diameter distribution is of the order of less than 5%.

In contrast to type I, NCT type II yields a relatively uniform distribution of SWCNT diameters from 0.8 to 1.4 nm (88% of all SWCNTs). The gap of the diameter distribution at about 1.15 nm, which is also observed for NCT type I, is explained by the missing resonance of such SWCNTs with 532 nm laser radiation. For this reason the diameter distribution is not described as bimodal but as uniform. The upper limit of the SWCNT diameter distribution obtained with NCT type II is defined by the very low amount of SWCNTs with an RBM frequency of about 140 cm^{-1} (<5%) corresponding to diameters of about 1.7 nm. The high overall yield of SWCNT with small diameters (40% in the 0.6–1.0 nm range) is however remarkable.

Since NCT type I is apparently a very promising pre-defined and stable catalyst for diameter-selective SWCNT growth, its morphology after pre-treatment was investigated. For this purpose the growth process was terminated after the pre-treatment step (exposing the NCTs to flowing air at 550 °C for 10 min) and the pre-treated template was analyzed using cross-sectional TEM and Raman spectroscopy (Fig. 7). The Raman

spectra of NCT type I show the typical sp^2 -carbon (sp^2 -C) Raman lines, *i.e.* the D line at around 1350 cm^{-1} and the G line at around 1550 cm^{-1} , before and after pre-treatment (Fig. 7a). The intensity of the sp^2 -C Raman lines decreased to 30% after the pre-treatment, and simultaneously the intensity of the Si-2TO lines at around 950 cm^{-1} increased by approximately 40% (not shown). These observations point to a partial loss of a-C, presumably by oxidation to CO_2 . Moreover, narrower line widths and G line up-shift indicate a higher degree of graphitization of the a-C matrix. Cross-sectional TEM analysis reveals a mean NP diameter of $(2.2 \pm 0.4) \text{ nm}$, which is the same as in the virgin NCT within the experimental accuracy. In conclusion, the pre-treatment step causes slight roughening of NCT type I due to partial removal of the embedding a-C matrix. These changes have apparently no effect on the NP morphology.

Discussion

In the following section the conclusions from the presented results for CVD growth of SWCNTs using C : Ni NCTs with and without predefined NPs are discussed.

(a) NP stability of NCT type I.

Without stabilization, Ni thin films on SiO_2 surfaces undergo clustering *via* surface diffusion for temperatures larger than -173 °C .^{70,71} Ni cluster diameters of 8 nm to 20 nm were reported for nominally 1 nm thick Ni films.^{71,72} Bulk diffusion of Ni into a SiO_2 layer on Si and formation of NiSi_2 starts at elevated temperatures ($\geq 425 \text{ °C}$), depending on SiO_2 thickness. The bulk diffusion onset temperature for a 2.5 nm thick SiO_2 layer was reported to be 800 °C .⁷⁰ These data demonstrate that without stabilization much larger Ni particles are formed at temperatures similar to those during catalyst pre-treatment and CVD growth in the present study.

In fact, our investigations have shown that a defined microstructure of Ni-rich NPs with narrow diameter distribution of $(2.0 \pm 0.3) \text{ nm}$ can be stabilized up to temperatures of 735 °C and used to grow SWCNTs. This has been achieved by embedding the NPs in a protective matrix of a-C, forming a NCT of type I. The protective a-C is fully stable in ambient conditions and allows easy handling of the NCTs in air. Thus, the a-C matrix ensures the safe transport of the templates and their transfer into the CVD oven. During the pre-treatment step at 550 °C in air, the a-C matrix is partially conserved. This ensures that the Ni-rich NPs keep their initial positions, diameter distribution and spacing. In this way the NPs are prevented from coagulation and grain coarsening. This observation is attributed to the strong interaction between Ni and C, which is *e.g.* larger than that between Ni and SiO_2 .⁷³ The rest of the a-C matrix presumably undergoes oxidation to CO_2 . After pre-treatment, the NCT type I enters the CVD synthesis with pre-defined, almost unchanged diameter in activated form.

(b) Relation between SWCNT and catalyst properties.

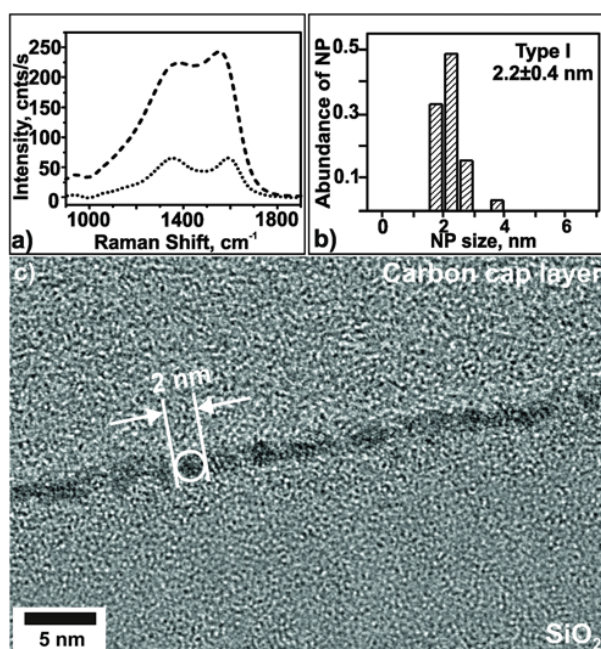


Fig. 7 Characterization of NCT type I after the pre-treatment step: (a) Raman spectrum (dotted line) in comparison to that of the pristine template (dashed line), (b) normalized size distribution of Ni-rich NPs measured by TEM, (c) cross-sectional TEM image.



It is obvious to explain the narrower and apparently selective SWCNT diameter distribution obtained by CVD growth using NCT type I by the pre-defined NP diameters and their demonstrated stability. As a consequence, the carbon precursors are exposed to almost equally-sized catalytically active NPs during the CVD process. In contrast to that situation, the initially homogeneously distributed Ni in NCT type II undergoes a coarsening, the carbon precursor is exposed to catalytic particles of different size and as a consequence, the SWCNT distribution is broader and not selective. On the other hand the smaller SWCNT diameters obtained with NCT type II can be attributed to the fact that NPs grow from an initially homogeneous Ni distribution to a size of (2.5 ± 0.8) nm. The behavior of NCT type II resembles catalysts whose catalytically active particles are formed *in situ* from metal thin films during the CVD process. The results presented here demonstrate that narrower SWCNT diameter distributions are accessible by the use of pre-defined, stable catalyst templates.

Apparently the best strategy to obtain SWCNTs with narrow diameter distribution is to use a catalyst with stable NPs of pre-defined size and to prevent them from coarsening during the CVD process. Several approaches to realize these preconditions were described in the literature. First, the catalyst NPs can be sparsely distributed over the inert substrate so that the large distance between the particles would prevent their aggregation.^{21,23,44,53} For instance, Paillet *et al.*⁵³ studied the CVD synthesis of SWCNTs on Ni NPs with the mean size of (4.7 ± 1.4) nm using C_2H_4 as a carbon source. The narrow size distribution of SWCNTs was achieved at 850 °C when the density of NPs and the synthesized SWCNTs was small (40 and 100 NPs per μm^2 led to 0.1–0.2 and ~ 3 SWCNTs per μm^2 , respectively). Importantly, for these conditions the Ni NPs had the same size before and after the CVD. An increase of the surface density to several hundred NPs per μm^2 led to aggregation of NPs and formation of amorphous carbon and big filamentous structures. An increase of the reaction temperature to 950 °C also resulted in larger Ni NPs with broader size distribution.⁵³ In our work, a narrow SWCNT diameter distribution was successfully obtained with a much higher density of catalyst NPs (*ca.* 20 000 NPs per μm^2) and synthesized SWCNTs (5–7 SWCNTs per μm^2).

So far, the most successful approach to obtain SWCNTs with a narrow diameter distribution is the use of an inert matrix, which protects NPs from coarsening or makes this process much slower than SWCNT nucleation. Well-defined Fe NPs were obtained *in situ* by a preliminary process step and stabilized during CVD by embedding them in an aluminum oxide matrix.^{16,27,29,31} This method offers catalyst NPs of different size with narrow diameter distribution on the substrate surface, but the size of NPs is not constant over the reaction time. The other widely used supports for nano-scaled metal catalysts are porous materials like mesostructured silica,^{32,33,36,39,40,45,74,75} porous magnesium oxide^{24,34,47} and zeolites,^{42,56,76} which offer SWCNTs with a selective chirality^{32–34,36,39,45,74} and sufficient yield. Catalyst particles immobilized in the substrate pores retain their size over reac-

tion time in a broad temperature range, however SWCNTs are formed not only on the surface but also in the bulk of the catalyst/matrix composite. As a result, they are bundled and inevitably mixed with the catalyst, which requires the use of laborious purification procedures.

In this work a new type of catalyst for diameter-selective SWCNT growth is presented. C:Ni nanocomposite template type I combines the following advantages: narrow catalyst diameter distribution, stability during the CVD process, potential for growing individual SWCNT with a significant length and yield. The study confirms the hypothesis that the diameter distribution of CVD-grown SWCNTs is determined by the catalyst morphology and the size of the catalyst particles at the initial point of the SWCNT nucleation.^{44,77} In NCT type II, formation and growth of the catalyst NPs competes with the simultaneous SWCNT growth. Thus, the size distribution of the catalyst NPs is changing with time, and so does the diameter distribution of the nucleating SWCNTs. On the contrary, in NCT type I the catalyst NPs with narrow and weakly varying size distribution are available from the very beginning and during the whole time of the SWCNT growth. Hence, the diameter distribution of the synthesized SWCNTs is much narrower. Certainly, the study also shows that control of catalyst diameter is a necessary but not a sufficient condition for diameter-selective or even chirality-selective growth of SWCNTs. Further well-established parameters controlling the SWCNT diameter distribution are growth temperature, nature and concentration of precursors, and catalyst pre-treatment.

A final question that shall be briefly addressed is the chemical state of the catalytically active Ni-rich NPs. After the CVD process, fcc-Ni and Ni_3C were identified by the TEM analysis (Fig. 3 and ESI†). It is reasonable to assume that these two Ni phases are catalytically active. fcc-Ni is commonly considered as most suitable Ni based catalyst for CNT growth.^{15,17,49,78} Recent papers also reported Ni_3C as an active catalyst, at least for carbon nanofibers⁶⁴ and initial graphene layers of CNTs.⁵⁰ Moreover, indications for a minor fraction of NiO after the CVD process were obtained. A conclusion about its catalytic activity can however not be drawn on the basis of the presented results. *In situ* XPS and TEM studies could help to clarify the phase and chemical state of the catalytically active Ni species.

Conclusions

A new type of pre-defined stable catalyst for diameter-controlled growth of SWCNTs was prepared and comprehensively characterized. It comprises dispersed catalytic particles, whose size is largely preserved during the CVD synthesis. This way, the problem of the Ostwald ripening is circumvented, and a new route for much better control over the diameter of CVD-grown SWCNTs is opened. Diameter-selectivity of NCT based SWCNT growth was demonstrated for the first time. A fraction of 50% of SWCNT diameters was confined to a range of (1.36 ± 0.10) nm. In comparison to the growth using the pre-defined



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NPs, a non-selective SWCNT distribution, with 88% of all SWCNT in the diameter range of 0.80 nm to 1.40 nm was obtained using a NCT with a homogenous microstructure under otherwise identical conditions (NCT type II).

The as-grown SWCNTs themselves are ideal candidates for further comprehensive characterization, *e.g.* by coupling of Raman spectroscopy with atomic force or scanning tunneling microscopy. They consist predominantly of separated SWCNTs, are free of solvents or surfactants, exhibit a low degree of bundling, and contain negligible amounts of MWCNTs. Their length to diameter ratio was of the order of 200. The narrow RBM line widths of 4 cm⁻¹ to 8 cm⁻¹ show a high structural order of the as-grown SWCNTs and point to a low amount of intrinsic structural defects and negligible nanotube bundle formation.

The presented approach opens a new way to perform CVD synthesis of SWCNTs using catalyst particles with well-defined size distribution. Usually in CVD growth of SWCNTs, the metallic NPs are synthesized *in situ*, which severely limits the possibility to reliably ascertain their homogeneity and quality. The NCTs used in this study can be prepared prior to CNT synthesis, are not destroyed when exposed to air, and their properties can be characterized with a high degree of certainty as described above.

Further optimization of the size distribution, stability, and catalytic activity of metallic NPs in NCTs promises even better control over the size and diameter distribution of CVD-synthesized SWCNTs. The a-C matrix could be replaced by thermally stable oxides, which would improve the stability of the catalytic NPs during CVD growth of SWCNTs. Moreover, the catalytic activity and diameter selectivity can be improved by using nanocomposite templates with embedded NP alloys like FeNi or FeCo instead of pure metals.

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Slow Noncollinear Coulomb Scattering in the Vicinity of the Dirac Point in GrapheneJ. C. König-Otto,^{1,2,*} M. Mittendorff,³ T. Winzer,⁴ F. Kadi,⁴ E. Malic,⁵ A. Knorr,⁴ C. Berger,^{6,7} W. A. de Heer,⁶A. Pashkin,¹ H. Schneider,¹ M. Helm,^{1,2} and S. Winnerl¹¹*Helmholtz-Zentrum Dresden-Rossendorf, P.O. Box 510119, 01314 Dresden, Germany*²*Technische Universität Dresden, 01062 Dresden, Germany*³*University of Maryland, College Park, Maryland 20742, USA*⁴*Technische Universität Berlin, Hardenbergstraße 36, 10623 Berlin, Germany*⁵*Chalmers University of Technology, SE-41296 Göteborg, Sweden*⁶*Georgia Institute of Technology, Atlanta, Georgia 30332, USA*⁷*Institut Néel, CNRS-Université Alpes, 38042 Grenoble, France*

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The Coulomb scattering dynamics in graphene in energetic proximity to the Dirac point is investigated by polarization resolved pump-probe spectroscopy and microscopic theory. Collinear Coulomb scattering rapidly thermalizes the carrier distribution in \mathbf{k} directions pointing radially away from the Dirac point. Our study reveals, however, that, in almost intrinsic graphene, full thermalization in all directions relying on noncollinear scattering is much slower. For low photon energies, carrier-optical-phonon processes are strongly suppressed and Coulomb mediated noncollinear scattering is remarkably slow, namely on a ps time scale. This effect is very promising for infrared and THz devices based on hot carrier effects.

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Coulomb scattering is a nonlinear many-body effect that transforms a nonequilibrium carrier distribution in a semiconductor into a hot Fermi-Dirac distribution [1]. In the gapless semiconductor graphene with linear energy dispersion, Coulomb scattering is known to be particularly strong [2–6]. A large number of both degenerate and multicolor pump-probe experiments, nonlinear THz spectroscopy, time-resolved photocurrent measurements, and time-resolved angle-resolved photoelectron emission spectroscopy (tr-ARPES) have provided detailed insights into the carrier dynamics of graphene [7–15]. Together with theoretical studies, they revealed that carriers typically thermalize on a sub-100 fs time scale [8,11,16]. This rapid transition from a nonequilibrium distribution to a hot thermalized distribution is mediated by both Coulomb scattering and scattering via optical phonons. These two processes have, so far, not been disentangled as they typically occur on the same time and energy scales [8,12,17]. However, when not just the electron energy, but also the angular distribution in \mathbf{k} space is considered, it has been found that the distribution, in fact, thermalizes rapidly along all \mathbf{k} directions radially pointing away from the Dirac point. In contrast, the thermalization between different \mathbf{k} directions is considerably slower. This angular thermalization that is mediated mainly by efficient optical-phonon scattering is completed only after 150 fs [18–22]. In these studies, an initial anisotropic electron distribution is generated by pumping with linearly polarized radiation [2,21,23]. This anisotropy is linked to pseudospin flipping in interband transitions [24,25].

In this Letter, we utilize this anisotropy to trace the temporal and directional characteristics of pure Coulomb

scattering in the vicinity of the Dirac point. To this end, we apply a photon energy of 88 meV that is well below the optical phonon energy (~ 200 meV) so that scattering via optical phonons is strongly suppressed. In order to ensure that scattering of the thermalized carrier distribution with optical phonons is also negligible, the study is carried out at low temperature and low enough fluences. Acoustic phonon scattering takes place on a longer, 100 ps time scale. Both the experiments and the theory reveal that, in this regime, angular thermalization characterized by an isotropic distribution is approached remarkably slowly, namely on a ps time scale.

The pump-probe experiments presented here are carried out in transmission geometry on epitaxial multilayer graphene (~ 50 layers), which was grown on the C face of 4H-SiC. Explicitly, the layers are rotationally stacked, so that each layer has the electronic structure of graphene [26,27]. The sample was kept at 20 K, and differential transmission signals (DTSSs) were recorded. In our degenerate pump-probe experiments, the free-electron laser FELBE provides radiation at a photon energy of 88 meV (pulse duration 4 ps, repetition rate 13 MHz). At this photon energy, the measured signals stem predominantly from interband transitions in the almost intrinsic graphene layers ($|E_f| = 10\text{--}20$ meV), and only negligible contributions from intraband absorption are expected [28–30]. The probe-beam polarization is set to 45° with respect to the polarization of the pump beam by a grating polarizer on a polymer foil. Polarizers mounted in front of a mercury cadmium telluride detector, with orientation parallel or perpendicular to the pump beam polarization, allow one to measure the pump-induced transmission for

$E_{\text{probe}} \parallel E_{\text{pump}}$ and $E_{\text{probe}} \perp E_{\text{pump}}$, respectively. Normal incidence on the sample is used for the probe beam, while the pump beam is offset with respect to the probe beam by a small angle. We also performed a two-color experiment, where a Ti:sapphire laser synchronized to FELBE [31] with a jitter of 5 ps is employed for pumping at a photon energy of 1.5 eV (pulse duration 3 ps, repetition rate 78 MHz) while probing at 88 meV. Also, in this experiment, the response is dominated by interband transitions in the almost intrinsic graphene layers.

In our theoretical investigation, we solve the graphene Bloch equations based on the Heisenberg equation of motion in Born-Markov approximation [32–34]. By accounting for the semiclassical light-matter, carrier-carrier, and carrier-phonon interactions on a consistent microscopic footing, we are able to realistically model the carrier dynamics resolved in time, energy, and momentum angle without any adjustable parameter [2]. The screening of the Coulomb interaction is treated within the static limit of the Lindhard equation [1]. Dynamical screening [5,35] yields very similar results due to the induced broadening of the phase space for collinear scattering and the resulting softening of the strict energy conservation [36]. The latter has been included in a self-consistent procedure taking into account the finite lifetime of two-particle correlations for electrons or phonons [36,37].

In Fig. 1(a), we show schematically, the expected results for the angle-dependence of the optically induced population ρ without scattering or saturation effects. It shows a clear anisotropy and follows a $\rho \sim |\sin(\Phi_k)|^2$ dependence featuring nodes at $\Phi_k = 0$ (and π), where Φ_k is the angle between the amplitude of the pump beam E_{pump} and the k vector of electrons (defined as the relative momentum with respect to the Dirac point) [2]. In the following, the states around $\Phi_k = \pi/2$ (and $3\pi/2$) with respect to the pump beam will be referred to as antinode states and the states around $\Phi_k = 0$ (and π) as node states. Because of Pauli blocking, the $E_{\text{probe}} \parallel E_{\text{pump}}$ ($E_{\text{probe}} \perp E_{\text{pump}}$) configuration preferentially probes the antinode (node) states. Before discussing the experimental findings of the degenerate pump-probe experiments in detail, the main aspects of the involved physics are briefly highlighted. For energies below the optical phonon energy, Coulomb interaction is expected to be the main scattering mechanism. This involves both intraband and interband (Auger-type) processes [cf. Fig. 1(b)]. Coulomb scattering in graphene is predominantly collinear, as $|V|^2 \propto (1 + \cos \phi_1)(1 + \cos \phi_2)$, where V is the intraband Coulomb matrix element and ϕ_i is the angle between the momenta of initial and final state of two scattering electrons $i = 1, 2$ [21,38]. That means the scattering cross section is maximal for collinear processes ($\phi_i = 0$), decreases with increasing scattering angle, and vanishes for back-scattering ($\phi_i = \pi$). In a degenerate pump-probe experiment at 88 meV, i.e. well below the optical phonon threshold,

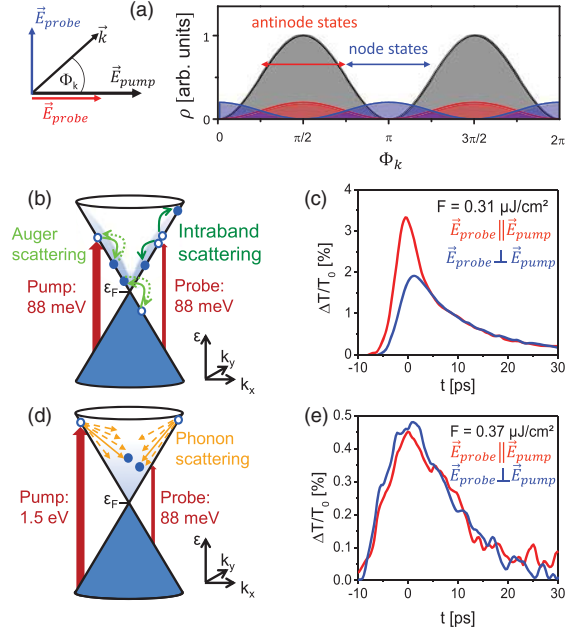


FIG. 1. (a) Schematic of the angular dependence of the occupation of optically excited carriers in the conduction band at the energy $\hbar\omega/2 = 44$ meV excited by the pump beam (black), a probe beam copolarized to the pump beam (red), and cross-polarized to the pump beam (blue). Schematic representation of the single-color (b) and two-color (d) experiments with important scattering mechanisms. Pump-induced transmission change for probing the antinode (red curves) and node states (blue curves), respectively, for the single-color (c) and two-color (e) experiments.

the different DTS for the two orthogonal probe pulse polarizations confirm that an anisotropic distribution persists for several ps [cf. Fig. 1(c)]. This is direct evidence that noncollinear scattering has not led to an angular thermalization. In contrast, pumping at 1.5 eV but with almost similar pulse duration and fluence enables scattering via optical phonons [cf. Fig. 1(d)]. As a result, the distribution probed at a photon energy of 88 meV is isotropic for all delay times [cf. Fig. 1(e)]. This comparison of single-color and two-color experimental results unambiguously shows that the dynamics in the vicinity of the Dirac point contains important aspects that are beyond the widely applied thermodynamic understanding of the carrier dynamics in graphene. The thermodynamic model implies thermalization on a sub-100 fs time scale and subsequent cooling (see, also, Supplemental Material [39]). Consequently, the induced transmission on time scales longer than 100 fs depends only on pump fluence, but not on pump photon energy. Many recent tr-ARPES and pump-probe experiments can be described very well by the thermodynamic model [11,15,17,49]. In these experiments, ultrafast thermalization is ensured by the excitation conditions, leading to both strong Coulomb and electron-phonon scattering. Our experiments show, however, that this is

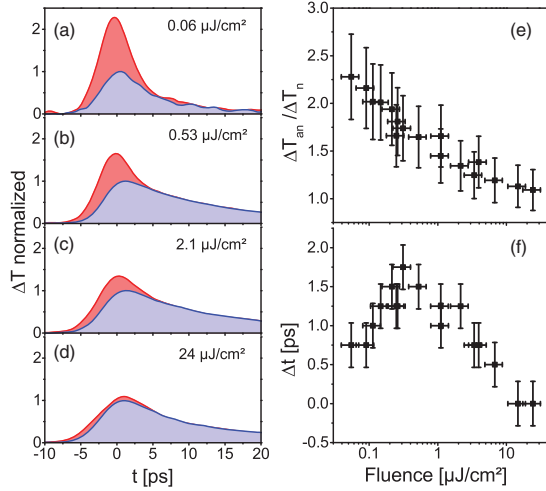


FIG. 2. (a)–(d) Single-color differential transmission for probing the antinode (red curves) and node (blue curves) states, respectively, normalized to the peak value obtained for probing the node states for different pump fluences. (e) Fluence dependence of the maximal induced transmission change for probing the antinode states divided by the maximal induced transmission change for probing the node states. (f) Fluence dependence of the temporal shift of the maximum induced transmission for probing the node states with respect to the maximum obtained for probing the antinode states.

not a general case. Comparably long time scales (few ps) for Coulomb scattering and a strong angular dependence of Coulomb scattering have also been predicted in a study on the impact-excitation of extrinsic electrons in graphene [38].

Next, for low-energy excitations, the carrier dynamics is investigated in a wide range of fluences. At low fluences, the induced maximum transmission change for probing the antinode states is about 2.3 times larger than for probing the node states, as shown in Figs. 2(a) and 2(e). In the absence of scattering, or for purely collinear scattering, one would expect a factor of 3 [18]. With increasing fluence, the ratio between the maximum transmission obtained for probing the antinode and node states $\Delta T_{an}/\Delta T_n$ asymptotically approaches the value 1, corresponding to a completely angularly thermalized distribution. Apart from the signal amplitudes, the temporal positions of the signal maxima for the two probing conditions also feature a pronounced fluence dependence [see Figs. 2(a)–2(d) and 2(f)]. The peaks for the two probe polarizations occur almost at the same time when low and high fluences are applied. At intermediate fluences ($\sim 0.3 \mu\text{J}/\text{cm}^2$), the signal probing the node states is delayed by about 1.6 ps with respect to the signal probing the antinode states. To understand this phenomenon, it is instructive to discuss three different fluence regimes: low, intermediate, and high. The Coulomb scattering rate scales with the number of available scattering partners, which, in turn, scales with the applied pump fluence. For low fluences, noncollinear Coulomb scattering

is most inefficient resulting in $\Delta T_{an}/\Delta T_n \approx 3$. In the absence of noncollinear scattering, the signal for probing the node states stems from optically excited carriers around $\Phi_k = \pi/4$ [and $3\pi/4, 5\pi/4, \dots$ cf. Fig. 1(a)]; consequently, there is no time delay between the signals for probing the node and antinode states. At intermediate fluences, noncollinear scattering becomes relevant, resulting in reduced values for $\Delta T_{an}/\Delta T_n$ and in a delayed maximum for probing the node states. This delay indicates the transfer of carriers from antinode states to node states. Finally, in the high-fluences regime, full thermalization occurs on time scales beyond the resolution of the experiment. This results in basically identical DTSs for probing antinode and node states, respectively. Naturally, these curves feature similar amplitudes and similar temporal positions of the maxima.

Note that saturation of pump-probe signals due to phase space filling can also affect the amplitude and temporal position of the induced transmission maxima. However, while the latter effect can qualitatively explain the reduction of the anisotropy with increasing fluence, it cannot explain the peculiar shift of the maximum for probing the node states. We have also performed polarization-resolved experiments at elevated temperatures up to room temperature. For temperatures above 100 K, the decay of the signal becomes fast and the amplitude drops rapidly (see, also, [29]) resulting in a decreased signal-to-noise ratio that does not allow one to trace the noncollinear scattering contribution over a large range of fluences. Nevertheless, we could not find any indication that the dynamics of the noncollinear Coulomb scattering are influenced by the substrate temperature up to 300 K (for more details, see the Supplemental Material [39]).

For a detailed insight into the relaxation dynamics and, also, for a clear separation of the saturation effect from noncollinear scattering, microscopic modeling is applied. The modeling is performed for an intrinsic graphene monolayer, nevertheless, the results are still valid for the more complex sample structure used in the experiment. For more information, see the Supplemental Material [39]. For directly visualizing the carrier dynamics, the carrier occupations along the direction of the center of the antinodes (center of the nodes) corresponding to $\mathbf{k} \perp \mathbf{E}_{\text{pump}}$ ($\mathbf{k} \parallel \mathbf{E}_{\text{pump}}$) are depicted in Fig. 3 for two fluences in a range where the largest qualitative differences are found. At low fluences and early times, the optically generated carriers at an energy of $\hbar\omega/2 = 44$ meV are clearly visible in the occupation along $\Phi_k = \pi/2$. However, a hot distribution is reached rapidly along the respective Φ_k direction due to efficient collinear scattering. As noncollinear scattering is less efficient, the distribution remains anisotropic on a time scale of several ps. For higher pump fluences, cf. Figs. 3(d)–3(f), both collinear and noncollinear Coulomb scattering becomes more efficient. As a result, thermalization along a particular Φ_k direction is considerably faster, and an almost isotropic

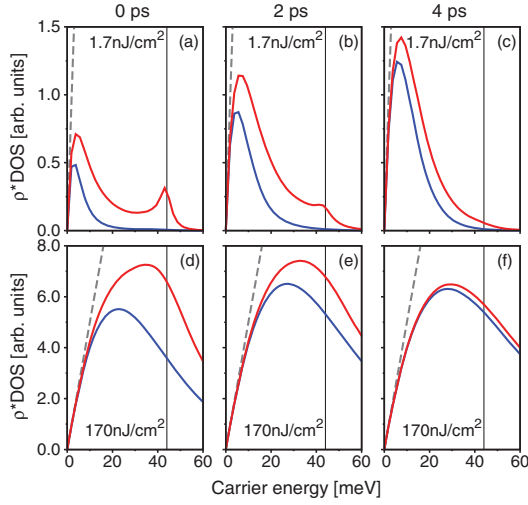


FIG. 3. (a)–(f) Carrier occupation multiplied by the density of states for three different times (time zero corresponds to maximum intensity of the pump pulse) and two different pump fluences. Red curves correspond to the $k \perp E_{\text{pump}}$ ($\Phi_k = \pi/2$), blue curves to $k \parallel E_{\text{pump}}$ ($\Phi_k = 0$). The solid line denotes the energy $\hbar\omega/2$, the dashed line half-filling of the available states.

distribution is reached within ~ 4 ps. Note that, even for high fluences, the occupation numbers at the probe energy of 44 meV are well below half-filling of available states. Therefore, saturation effects can be ruled out as the main mechanism of the observed dynamics.

With increasing pump fluence, the temperature of the thermalized electrons increases. Consequently, electronic states above 200 meV also eventually become populated and, thus, emission of optical phonons may contribute to the relaxation dynamics [29]. To investigate the strength of this effect, the DTS curves are calculated for the full dynamics and for the dynamics without phonons. The differences are negligible, a detailed comparison is shown in the Supplemental Material [39]. In summary, those calculations, together with the extraordinary lifetime of the anisotropy compared to experiments in the near infrared [18,20] and the comparison of the two color and the degenerate pump probe experiment [cf. Figs. 2(c) and 2(e)], prove that noncollinear Coulomb scattering is the only remaining relaxation channel to reduce the anisotropy. This channel can be effectively controlled and studied by the variation of the pump fluence.

Finally, we quantitatively investigate the decay of the anisotropic carrier distribution into a thermalized distribution and compare experiment and theory. To this end, the temporal evolution of the ratio between the induced transmissions for probing the antinode and node states, respectively, $\Delta T_{an}/\Delta T_n$ is evaluated for both experimental and theoretical data (see Fig. 4). Starting from a value of 3, corresponding to the optically excited nonequilibrium distribution, the ratio $\Delta T_{an}/\Delta T_n$ drops to the value of 1,

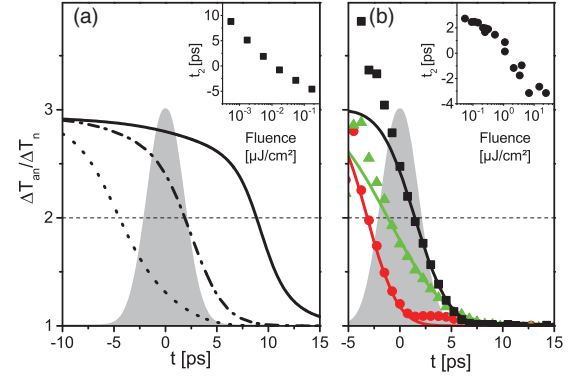


FIG. 4. The decay of the ratio $\Delta T_{an}/\Delta T_n$ is depicted for different fluences. (a) theoretical data: 171 nJ/cm² dotted line; 5.4 nJ/cm² dotted-dashed line; 0.54 nJ/cm² solid line. (b) experimental data: 24 μ J/cm² circle; 2.1 μ J/cm² triangle; 0.5 μ J/cm² square. The gray shaded areas indicate the pump pulse. For the experimental data in (b), fit curves based on an error function that were used to extract the value of t_2 are shown. In the insets, the time t_2 ascribed to the half-way decay of the optically induced anisotropy is shown over the fluence.

characterizing the isotropic thermalized distribution. While the overall shape of this ratio as a function of time depends only weakly on fluence, the point in time at which the distribution loses its anisotropic character depends strongly on fluence. For low fluences, the anisotropic distribution persists until the pump pulse has vanished, while for high fluences, a significant loss in anisotropy occurs already during the rising edge of the pulse [cf. Fig. 4(a)]. The experimental dynamics [cf. Fig. 4(b)] is qualitatively very similar to the theoretically predicted one. Since, for early times, the signal is small compared to the noise, the ratio $\Delta T_{an}/\Delta T_n$ at those times is instable as one essentially divides by zero. Note that a significant part of the dynamics occurs during the pump pulse; hence, there is a balance of refreshing the anisotropic distribution by optical pumping and a decay of the anisotropy by noncollinear scattering. For a quantitative comparison, the time t_2 at which the distribution is half-way angularly thermalized (i.e., $\Delta T_{an}/\Delta T_n = 2$), is extracted from both the theoretical and experimental data [cf. insets of Figs. 4(a) and 4(b)]. In both experiment and theory, t_2 is positive for small fluences, reflecting a pronounced anisotropic distribution after the excitation pulse maximum. Increasing noncollinear Coulomb scattering by applying higher fluences results in a reduction of t_2 down to values corresponding to the very front of the excitation pulse (~ -4 ps). However, there is a notable offset between the theoretical and the experimental data; i.e., higher fluences are required in the experiment to achieve similar effects. This might be ascribed to an overestimation of the fluence in the experiment since the fluence is reduced for the layers underneath the top layer. Furthermore, there is an uncertainty in the determination of the temporal position of the pump-pulse

maximum. Last, but not least, it has to be considered that no adjustable parameters are used in the comparison.

The comparably long time for angular thermalization is expected to be advantageous for infrared and THz device applications based on hot carriers such as fast detectors and modulators [50–52]. To improve the efficiency of such devices, the higher electron temperature in the direction perpendicular to the direction of the polarization can be exploited by applying appropriate electrode geometries. The relaxation time on the ps time scale is attractive as it is long enough to result in a significant contribution to the extractable signal of such devices. On the other hand, the ps time scale is fast enough to prevent a deterioration of the operation speed of the device, resulting in a high temporal resolution. Anisotropic photoconductive effects have been observed for excitation with visible radiation [53]. Our study suggests that this effect should be much larger for photon energies below the optical phonon energy. Besides its fundamental importance, the present study is of particular relevance for gated devices, where the Fermi level is shifted to the charge neutrality point [54,55]. Furthermore, our calculations predict that an anisotropic carrier distribution on a ps time scale can also be observed in doped graphene; however, in this case, the pump-induced change in transmission is much smaller (see Supplemental Material [39]). These preliminary results suggest that our main findings may also be relevant for devices employing doped graphene. For all the mentioned device applications, it is highly attractive that anisotropic carrier distribution also persists at room temperature.

In conclusion, our study has revealed that low energy Coulomb scattering in almost intrinsic graphene exhibits an unusual twofold nature: It rapidly thermalizes the distribution along all k directions pointing radially away from the Dirac point, while preserving the optically induced anisotropy on a ps time scale. This effect is attractive for optoelectronic devices based on hot carriers.

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Statistics

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Optical Materials Express **6**, 711 (2016)
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The formation of silver metal nanoparticles by ion implantation in silicate glasses
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Electron-beam pulse annealed Ti-implanted GaP
Journal of Applied Physics **120**, 085103 (2016)
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Tuning the structure and crystal habit of mesocrystals using magnetic fields
Nanoscale **8**, 15571 (2016)
179. Xie, Y.; Chen, X.; Zhang, Z.; Song, W.; Zhou, S.; Yang, Z.
Magnetocapacitance in $\text{CdCr}_{1.8}\text{In}_{0.2}\text{S}_4$ Single Crystal Annealed in Cadmium Vapor
IEEE Transactions on Magnetics **52**, 2501404 (2016)
180. You, T.; Selvaraj, L. P.; Zeng, H.; Luo, W.; Du, N.; Buerger, D.; Skorupa, I.; Prucnal, S.; Lawrenz, A.; Mikolajick, T.; Schmidt, O. G.; Schmidt, H.
An Energy-Efficient, BiFeO_3 -Coated Capacitive Switch with Integrated Memory and Demodulation Functions
Advanced Electronic Materials **2**, 1500352 (2016)
181. Yu, H.; Kopach, A.; Misko, V. R.; Vasylenko, A. A.; Makarov, D.; Marchesoni, F.; Nori, F.; Baraban, L.; Cuniberti, G.
Confined catalytic Janus swimmers in a crowded channel: geometry-driven rectification transients and directional locking
Small **12**, 5882 (2016)
182. Zipf, L.; Merchel, S.; Bohleber, P.; Rugel, G.; Scharf, A.
Exploring ice core drilling chips from a cold Alpine glacier for cosmogenic radionuclide (^{10}Be) analysis
Results in Physics **6**, 78 (2016)

Popular-science publications

183. Helm, M.; Michel, P.; Gensch, M.; Wagner, A.
Alles im Fluss
Physik Journal **15**(1), 29-34 (2016)
184. Merchel, S.; DREAMS-Team; DREAMS-Users
Bestimmung langlebiger Radionuklide mittels Beschleunigermassenspektrometrie (AMS) für archäometrische Fragestellungen
Metalla **8** (Special Issue), 30-33 (2016)
185. Schultheiß, H.
Optik einmal anders
Physik Journal **15**(10), 16-17 (2016)

Concluded scientific degrees

PhD theses

1. Eßer, F.
Cyclotron resonance and photoluminescence studies of dilute GaAsN in magnetic fields up to 62 Tesla
TU Dresden, 18.08.2016
2. Fehrenbacher, M.
Terahertz near-field investigation of a plasmonic GaAs superlens
TU Dresden, 03.03.2016
3. Heidarian, A.
Study of the static and dynamic magnetization across the first order phase transition in FeRh thin films
TU Dresden, 22.01.2016
4. Kosub, T.
Ferromagnet-free magnetoelectric thin film elements
TU Chemnitz, 25.11.2016
5. Neubert, M.
Die Rolle des Sauerstoffanteils in Titandioxid bei Tantal-Dotierung zur Verwendung als transparentes leitfähiges Oxid
TU Dresden, 01.02.2016
6. Steinbach, G.
Ferromagnetic colloidal particles with anisotropic magnetization distribution: self-assembly and response to magnetic fields
TU Chemnitz, 10.05.2016
7. Stephan, D.
Inter-sublevel dynamics in single InAs/GaAs quantum dots probed by strong terahertz excitation
TU Dresden, 12.09.2016
8. Teschome, B.
Functional DNA origami nanostructures for nanoelectronics
TU Dresden, 13.12.2016
9. Yildirim, O.
Effect of microstructure on the magnetic properties of transition metal implanted TiO₂
TU Dresden, 24.02.2016

Master/Diploma theses

1. Abdou, J.
Thermodynamics and kinetics of Y-Ti-O nanoclusters in bcc-Fe
RWTH Aachen, 30.06.2016
2. Agha, C.
Investigations on the dependence of the Sticking Coefficient of Silver (Ag) on the topography of Silicon (Si) surfaces using in-situ Rutherford Backscattering Spectrometry
TU Dresden, 18.08.2016
3. Braun, M.
Elektrische und optische Eigenschaften von dotierten ZnO Dünnschichten nach Temperung mit spektral modifizierten Blitzlampen
Westfälische Hochschule Zwickau, 15.11.2016
4. Iseke, H.
Charakterisierung von graphenbasierten Detektoren im NIR–UV Bereich
TU Dresden, 19.09.2016
5. Khan, M. B.
Fabrication of sub-20 nm Silicon Nanowires
TU Dresden, 07.07.2016
6. Körber, L.
Phasenverschiebung und Transmission von Spinwellen durch magnetische Domänenwände
TU Dresden, 12.12.2016
7. Strobel, A. T.
Verwendung einer stromdurchflossenen Spule zur Simulation der Wechselwirkung von magnetischen Januskugeln
TU Chemnitz, 12.04.2016

Appointments and honors

Appointments

1. **Schultheiß, Helmut**

Head of the Emmy Noether Junior Research Group “Magnonics: Spin waves bridging Spintronics and Photonics” was appointed as **Associate Editor** of the journal Magnetic Letters of the IEEE Magnetics Society.

Awards and honors

1. **Schultheiß, Helmut**

Head of the Emmy Noether Junior Research Group “Magnonics: Spin waves bridging Spintronics and Photonics” was announced as the **Walter Schottky Prize 2017** laureate for „his fundamental research about understanding spin wave propagation in nanostructures and their application in new functional devices for transport and logic processing of information“.

2. **Arora, Himani**

PhD student in the division “Scaling Phenomena” and fellow of the International Research School NanoNet won a **Cfaed Inspire Grant** by the Center for Advancing Electronics Dresden (cfaed) to collaborate with Prof. James Hone at Columbia University, N.Y., USA for a 3-months stay in the field of heterostructures of 2-dimensional materials. In addition, she received the **Best Student Paper Award** for her contribution "Building electronics based on two-dimensional materials" at the IEEE RADIO 2016 conference.

3. **Ehrler, Jonathan**

PhD student in the division “Magnetism” obtained an **IEEE Travel Grant** to attend the IEEE Summer School in Sendai, Japan, July 10 – 16, 2016.

4. **Günther, Florian**

PhD student in the division “Scaling Phenomena”, fellow of the Center for Advancing Electronics Dresden (cfaed), and member of the International Research School NANONET acquired a **DAAD PhD Student Grant** to collaborate with the group of Prof. Ubirajara Rodrigues Filho at the University Sao Paolo, Campus Sao Carlos, Brazil on semiconducting and dielectric materials for polymer electronics for one year starting October 2016.

5. **Kilibarda, Filip**

PhD student in the division “Scaling Phenomena” and fellow of the International Research IHRS NANONET won the **2nd Poster Prize** for his contribution "Single molecule level measurements" at the NANONET International Workshop 2016, Prague, Czech Republic, August 30 – September 02, 2016.

6. **Trinh, Thu Trang**

PhD student in the division “Magnetism” received a **DAAD Congress Grant** to participate at the International Hydrogen Conference in Grand Teton, USA, September 11 – 14, 2016.

7. **Wang, Mao**

PhD candidate in the Helmholtz Young Investigator Group "Functional Materials" of the division "Semiconductor Materials" received the **Best Student Poster Award** with her presentation "Electrical conduction and negative magnetoresistance in tellurium-hyperdoped silicon" at the 33rd International Conference on the Physics of Semiconductors, Beijing, China, July 31 – August 08, 2016.

8. **Yuan, Ye**

PhD candidate in the Helmholtz Young Investigator Group "Functional Materials" of the division "Semiconductor Materials" won the **1st Prize** at the **Young Scientist Contest** with his oral presentation "Ferromagnetic Mn-implanted GaP: Microstructures vs. magnetic properties" at the 11th International Conference: Ion Implantation and Other Applications of Ions and Electrons, Kazimierz Dolny, Poland, June 13 – 16, 2016.

Invited conference contributions, colloquia, lectures and talks

Invited conference talks

1. Bali, R.; Hlawacek, G.; Liersch, V.; Röder, F.; Wintz, S.; Semisalova, A.; Fassbender, J.
Positive magnetic patterning using ion beams – principles and device applications
603. WE-Heraeus-Seminar: Magnonics – Spin Waves Connecting Charges, Spins and Photons, 06. – 08.01.2016, Bad Honnef, Germany
2. Berencén, Y.; Prucnal, S.; Liu, F.; Wang, M.; Zhou, S.; Helm, M.; Rebohle, L.; Skorupa, W.
Non-equilibrium thermal processing for hyperdoping Si
11th International Conference of ion implantation and other applications of ions and electrons – ION 2016, 13. – 16.06.2016, Kazimierz Dolny, Poland
3. Bernert, K.; Sluka, V.; Fowley, C.; Lindner, J.; Deac, A. M.; Fassbender, J.
General switching voltages for magnetic tunnel junctions with in-plane and/or perpendicular-to-plane anisotropy free layers
New Horizons for Memory Storage: Advancing Non-volatile Memory with Atomistic Simulations, 29.06. – 01.07.2016, Dublin, Ireland
4. Bishop, S.; Ludwig, P.; Egli, R.; Chernenko, V.; Deveva, B.; Faestermann, T.; Famulok, N.; Fimiani, L.; Gomez, J.; Hain, K.; Korschinek, G.; Hanzlik, M.; Merchel, S.; Rugel, G.
Time-Resolved Two Million Year Old Supernova Activity Discovered in the Earth's Microfossil Record
Carpathian Summer School of Physics 2016, 26.06. – 09.07.2016, Sinaia, Romania
5. Bishop, S.; Ludwig, P.; Egli, R.; Chernenko, V.; Deveva, B.; Faestermann, T.; Famulok, N.; Fimiani, L.; Gomez, J.; Hain, K.; Korschinek, G.; Hanzlik, M.; Merchel, S.; Rugel, G.
Time-Resolved Two Million Year Old Supernova Activity Discovered in the Earth's Microfossil Record
Max-Planck-Institute for Physics (MPP) Colloquium, 17.05.2016, Munich, Germany
6. Böttger, R.; Heinig, K.-H.; Bischoff, L.; Huebner, R.
Self-organized Ge & Si nanostructures by heavy-ion irradiation
24th International Conference on the Application of Accelerators in Research and Industry CAARI, 30.10. – 04.11.2016, Fort Worth, TX, USA
7. Buljan, M.; Nekic, N.; Sancho Paramon, J.; Jercinovic, M.; Bogdanovic-Radovic, I.; Grenzer, J.; Hübner, R.; Bernstorff, S.
Self-ordered Ge-based core/shell quantum dots in glass matrix
20th International Vacuum Congress (IVC-20), 21. – 26.08.2016, Busan, Korea
8. Dimakis, E.
The role of Ga droplets in the epitaxy of GaAs nanowires on Si substrates
FemtoTera Workshop, 29.09.2016, Óbuda University, Budapest, Hungary
9. Engler, M.; Ou, X.; Facsko, S.
“Reverse Epitaxy” on semiconductor surfaces by low energy ion irradiation
11th International Conference on Ion Implantation and other Applications of Ions and Electrons – ION 2016, 13. – 16.06.2016, Kazimierz Dolny, Polska
10. Erb, D.; Engler, M.; Ou, X.; Facsko, S.
Reverse epitaxy: Nanopattern formation by vacancy self-assembly upon low energy ion irradiation of crystalline semiconductor surfaces
24th International Conference on the Application of Accelerators in Research and Industry CAARI, 30.10. – 04.11.2016, Fort Worth, TX, USA

11. Facsko, S.; Ou, X.; Engler, M.; Erb, D.; Lenz, K.
Properties of Metallic Thin Films Grown On Periodically Corrugated Surfaces
18th International Conference on Solid Films and Surfaces (ICSFS 18), 28.08. – 02.09.2016, Chemnitz, Germany
12. Fassbender, J.
Ion beam modification of magnetic materials – revisited
DPG Spring Meeting, 06. – 11.03.2016, Regensburg, Germany
13. Georgiev, Y.
Novel Nanoelectronic Devices and Technologies for Their Fabrication
IEEE 25th International Scientific Conference Electronics – ET 2016, 12. – 14.09.2016, Sozopol, Bulgaria
14. Heinig, K. H.; Facsko, S.; Stegemann, K. H.; Pruefer, T.; Xu, X.; Hlawacek, G.; Huebner, R.; Wolf, D.; Bischoff, L.; Moeller, W.; Borany, J.
Ion Irradiation Assisted Fabrication of Si Quantum Dots for Ultra-Low Power Electronics
20th International Conference on Ion Beam Modification of Materials (IBMM 2016), 30.10. – 4.11.2016, Wellington, New Zealand
15. Helm, M.
Solid state spectroscopy with THz free electron lasers
Laser Optics 2016, 27.06. – 01.07.2016, St. Petersburg, Russland
16. Helm, M.
MIR and THz spectroscopy of condensed matter
Summer School NanoTech Tera-Mir, 29.08. – 01.09.2016, Porquerolles, Hyeres, France
17. Helm, M.; König-Otto, J. C.; Mittendorff, M.; Pashkin, A.; Schneider, H.; Winnerl, S.; Wendler, F.; Winzer, T.; Malic, E.; Knorr, A.
Surprising effects of electron-electron scattering in graphene revealed by THz pump-probe spectroscopy
Teranano VII, 02. – 07.10.2016, Porquerolles, France
18. Helm, M.; König-Otto, J. C.; Mittendorff, M.; Pashkin, A.; Schneider, H.; Winnerl, S.; Wendler, F.; Winzer, T.; Malic, E.; Knorr, A.
Surprising effects of electron-electron scattering in graphene revealed by THz pump-probe spectroscopy
International Workshop on Terahertz Science, Nanotechnologies and Applications, 16. – 22.07.2016, Erice, Italy
19. Helm, M.; König-Otto, J. C.; Mittendorff, M.; Pashkin, A.; Schneider, H.; Winnerl, S.; Wendler, F.; Winzer, T.; Malic, E.; Knorr, A.
The strange implications of electron-electron scattering in graphene
International Conference on Terahertz Emission, Metamaterials and Nanophotonics (TERAMETANANO 2016), 03. – 10.04.2016, Cartagena, Colombia
20. Helm, M.; König-Otto, J.; Mittendorff, M.; Pashkin, A.; Schneider, H.; Wendler, F.; Winzer, T.; Malic, E.; Knorr, A.; Winnerl, S.
Surprising effects of electron-electron scattering in graphene
5th Russia-Japan-USA-Europe Symposium on Fundamental & Applied Problems of Terahertz Devices & Technologies (RJUSE TeraTech-2016), 31.10. – 04.11.2016, Sendai, Japan
21. Hlawacek, G.; Veligura, V.; Bali, R.
Application of Helium Ion Microscopy to study radiation damage
24th International Conference on the Application of Accelerators in Research and Industry CAARI, 30.10. – 04.11.2016, Fort Worth, TX, USA
22. Kaltenecker, K. J.; König-Otto, J. C.; Mittendorff, M.; Winnerl, S.; Schneider, H.; Helm, M.; Helm, H.; Walther, M.; Fischer, B. M.
Tracing the Gouy phase shift of focused, radially polarized THz pulses
41st International Conference on Infrared, Millimeter and Terahertz Waves, 25. – 30.09.2016, Kopenhagen, Denmark
23. Kelling, J.; Ódor, G.; Heinig, K. H.; Weigel, M.; Gemming, S.
Pushing the Limits of Lattice Monte-Carlo Simulations using GPUs
Perspectives of GPU computing in Science, 26. – 28.09.2016, Roma, Italia

24. Krause, M.
Introduction into Raman spectroscopy
Friends²-Workshop: Advanced coating and characterization techniques, 19. – 20.09.2016, Dresden-Rossendorf, Germany
25. Krause-Rehberg, R.; John, M.; Akhmadaliev, S.; Böttger, R.; Anwand, W.; Wagner, A.
Improvement of Depth Resolution of VEPAS by Sputtering Techniques
14th International Workshop on Slow Positron Beam Techniques & Applications, 22. – 27.05.2016, Matsue, Japan
26. Lenz, K.; Narkowicz, R.; Reiche, C. F.; Kákay, A.; Mühl, T.; Büchner, B.; Suter, D.; Lindner, J.; Fassbender, J.
Magnetization dynamics of a single Fe-filled carbon nanotube detected by ferromagnetic resonance
5th International Conference on Microwave Magnetics, 05. – 08.06.2016, Tuscaloosa, USA
27. Makarov, D.
Magnetic functionalities for flexible interactive electronics
61st Annual Conference on Magnetism and Magnetic Materials (MMM 2016), 31.10. – 04.11.2016, New Orleans, USA
28. Makarov, D.
Shapeable magnetic sensorics
Brazilian-German Frontiers of Science & Technology Symposium, 20. – 23.09.2016, Campinas, Brazil
29. Makarov, D.
Curvilinear magnetism
7th International Conference on Metamaterials, Photonic Crystals and Plasmonics (META 2016), 25. – 28.07.2016, Malaga, Spain
30. Makarov, D.
Active and passive electronics for smart implants
MRS Fall Meeting 2016, 27.11. – 02.12.2016, Boston, USA
31. Makarov, D.
Magnetism in curved geometries
8th Joint European Magnetic Symposia (JEMS 2016), 21. – 26.08.2016, Glasgow, United Kingdom
32. Makarov, D.; Streubel, R.; Perez Rodriguez, N.; Pierce, D. T.; Unguris, J.; Pofahl, S.; Schäfer, R.; Schmidt, M.; Baenitz, M.; Kronast, F.; Wilhelm, H.; Rößler, U. K.
Observations on surface magnetic order in FeGe and FeSi
MRS Fall Meeting 2016, 27.11. – 02.12.2016, Boston, USA
33. Markwitz, A.; Williams, G.; Osipowicz, T.; Hübner, R.; Saumitra, V.; Gupta, P.
Self Assembly of Magnetic Nanoclusters in Diamond-Like Carbon: A Diffusion Enhanced Process Activated by Collision Cascades
International Union of Materials Research Societies – International Conference on Electronic Materials (IUMRS-ICEM) 2016, 04. – 08.07.2016, Singapore, Singapore
34. Merchel, S.; Khojasteh Mohammadi, Nasrin B.; Pavetich, S.; Rugel, G.; Scharf, A.; Dreams-Users; Dreams-Friends
Determination of long-lived cosmogenic radionuclides by accelerator mass spectrometry
International Conference on Radioanalytical and Nuclear Chemistry (RANC-2016), 10. – 15.04.2016, Budapest, Hungary
35. Osten, J.; Greene, P.; Lenz, K.; Fassbender, J.; Jenkins, C.; Arenholz, E.; Endo, T.; Iwata, N.; Liu, K.
Use of ion irradiation to tune magnetic anisotropy
Ion Beams in Materials Engineering and Characterization (IBMEC), 28.09. – 01.10.2016, New Delhi, Indien
36. Otolara, J. A.; Yan, M.; Lindner, J.; Fassbender, J.; Hertel, R.; Kakay, A.
Spin waves going 3D – chiral effects in curved magnetic nanowires
603. WE-Heraeus-Seminar: Magnonics – Spin Waves Connecting Charges, Spins and Photons, 06. – 08.01.2016, Bad Honnef, Deutschland

37. Pashkin, A.
Nonlinear Multi-Terahertz Excitation of Strongly Correlated Materials
Gordon Research Conference "Ultrafast Phenomena in Cooperative Systems", 14. – 19.02.2016, Lucca (Barga), Italy
38. Pashkin, A.
FEL-based nonlinear THz spectroscopy at the ELBE accelerator
Workshop on an accelerator based source for nonlinear THz science, 03. – 04.10.2016, SwissFEL, Windisch, Switzerland
39. Pelizzo, M. G.; Corso, A. J.; Zuppella, P.; Böttger, R.; Tessarolo, E.
Optical components in harsh space environment
SPIE Meeting – Planetary Defense and Space Environment Applications (Conference OP410), 28.08. – 01.09.2016, San Diego, United States
40. Prucnal, S.
Ge comeback – new properties of an old material
11th International Conference on Ion Implantation and other Applications of Ions and Electrons – ION 2016, 13. – 16.06.2016, Kazimierz Dolny, Polska
41. Rebohle, L.; Schumann, T.; Prucnal, S.; Skorupa, W.
Ultra-fast Thermal Processing
The International Conference on Coatings on Glass and Plastics, 12. – 16.06.2016, Braunschweig, Germany
42. Rebohle, L.; Schumann, T.; Prucnal, S.; Skorupa, W.
Ultra-fast thermal processing of TCO and other thin films
6. Workshop Transparente leitfähige Materialien, 22. – 23.11.2016, Erfurt, Germany
43. Schneider, H.
Terahertz free-electron laser spectroscopy of semiconductor nanostructures
8th International Symposium on Ultrafast Phenomena and Terahertz Waves (ISUPTW 2016), 10. – 12.10.2016, Chongqing, China
44. Schneider, H.
Semiconductor spectroscopy with infrared and THz free-electron lasers
International Conference on "Synchrotron and Free electron laser Radiation: generation and application" (SFR-2016), 04. – 07.07.2016, Novosibirsk, Russia
45. Schneider, H.; Stephan, D.; Zybelle, S.; Winnerl, S.; Bhattacharyya, J.; Eßer, F.; Helm, M.
Exciton dynamics in semiconductor quantum wells and single quantum dots studied with a THz free-electron laser
International Workshop on "Terahertz Science, Nanotechnologies and Applications", 16. – 22.07.2016, Erice, Italien
46. Schneider, H.; Stephan, D.; Zybelle, S.; Winnerl, S.; Bhattacharyya, J.; Eßer, F.; Helm, M.
Terahertz free-electron laser spectroscopy of excitons in III-V semiconductor quantum wells and single quantum dots
SPIE Photonics Asia, Symposium on "Infrared, Millimeter Wave, and Terahertz Technologies", 12. – 14.10.2016, Beijing, China
47. Schumann, E.; Lungwitz, F.
UV-vis-NIR spectroscopic Ellipsometry and Photospectrometry of thin films
Friends²-Workshop: Advanced coating and characterization techniques, 19. – 20.09.2016, Dresden-Rossendorf, Germany
48. Skorupa, W.
Subsecond thermal processing for the advancement of thin layers and functional coatings
SVC (Society of Vacuum Coaters), 59th Annual Technical Conference (TechCon), 09. – 13.05.2016, Indianapolis, IN, USA
49. Skorupa, W.
Engineering for NEMS with ion beams & flash annealing
Workshop on "Ion Implantation – improving the MEMS processes?", 03.03.2016, Erfurt, Germany

50. Sluka, V.; Weigand, M.; Kakay, A.; Schultheiß, K.; Warnatz, T.; Erbe, A.; Tiberkevych, V.; Slavin, A.; Deac, A.; Lindner, J.; Fassbender, J.; Raabe, J.; Wintz, S.
Directional Spin Wave Emission From Topological Spin Textures
1st Baltic Spin conference, 09. – 13.08.2016, Jurmala, Lettland
51. Smekhova, A.; Eggert, B.; Cöster, B.; La Torre, E.; Szyjka, T.; Ollefs, K.; Bali, R.; Potzger, K.; Cornelius, S.; Liedke, M. O.; Lindner, J.; Salamon, S.; Rogalev, A.; Wende, H.
Ferromagnetism of Fe₆₀Al₄₀ thin films under a magnifying glass of hard X-rays
50th PNPI School on condensed matter physics, 14. – 19.03.2016, St. Petersburg, Russian Federation
52. Wagner, A.; Anwand, W.; Ehrler, J.; Krause-Rehberg, R.; Liedke, M. O.; Potzger, K.; Trinh, T. T.
Positron Annihilation Lifetime Spectroscopy at a Superconducting Electron Accelerator
14th International Workshop on Slow Positron Beam Techniques & Applications, 22. – 27.05.2016, Matsue, Japan
53. Wagner, K.; Kakay, A.; Schultheiß, K.; Henschke, A.; Sebastian, T.; Schultheiß, H.
Magnetic domain walls as reconfigurable spin-wave nanochannels
61st Annual Conference on Magnetism and Magnetic Materials (MMM 2016), 31.10. – 04.11.2016, New Orleans, USA
54. Wagner, K.; Kakay, A.; Schultheiß, K.; Henschke, A.; Sebastian, T.; Schultheiß, H.
Magnetic domain walls as reconfigurable spin-wave nanochannels
SPIE Optics and Photonics Conference, Spintronics IX Symposium, 28.08. – 01.09.2016, San Diego, USA
55. Wilhelm, R. A.; Gruber, E.; Smejkal, V.; Schwestka, J.; Kozubek, R.; Hierzenberger, A.; Schleberger, M.; Facsko, S.; Aumayr, F.
Highly charged ion interaction with graphene
12th European Conference on Atoms, Molecules and Photons (ECAMP), 05. – 09.09.2016, Frankfurt., Germany
56. Wilhelm, R. A.; Gruber, E.; Smejkal, V.; Schwestka, J.; Kozubek, R.; Hierzenberger, A.; Schleberger, M.; Facsko, S.; Aumayr, F.
Charge equilibration and energy loss of slow highly charged ions in single layer graphene
27th International Conference on Atomic Collisions in Solid (ICACS), 24. – 29.07.2016, Lanzhou, China
57. Winnerl, S.; Fehrenbacher, M.; Kuschewski, F.; von Ribbeck, H.-G.; Döring, J.; Kehr, S.; Eng, L. M.; Schneider, H.; Helm, M.
Plasmonic superlens based on doped GaAs
Smaller and Faster: Infrared and Terahertz Spectral-Imaging at the Nanoscale with Synchrotron and Free Electron Laser Sources (SAFE), 01. – 02.12.2016, Trieste, Italy
58. Winnerl, S.; König-Otto, J. C.; Mittendorff, M.; Pashkin, A.; Schneider, H.; Helm, M.; Winzer, T.; Wendler, F.; Malic, E.; Knorr, A.
Surprising Coulomb-scattering effects in graphene revealed by polarization-resolved THz spectroscopy
5th EOS Topical Meeting on Terahertz Science & Technology, 08. – 11.05.2016, Pecs, Hungary
59. Winnerl, S.; Mittendorff, M.; König-Otto, J. C.; Wendler, F.; Malic, E.; Knorr, A.; Pashkin, A.; Schneider, H.; Helm, M.
Population and polarization dynamics in Landau-quantized graphene – evidence for strong Auger scattering
22nd International Conference on High Magnetic Fields in Semiconductor Physics (HMF-22), 24. – 29.07.2016, Sapporo, Japan
60. Wintz, S.; Sluka, V.; Weigand, M.; Kakay, A.; Schultheiß, K.; Erbe, A.; Tiberkevych, V.; Slavin, A.; Deac, A.; Lindner, J.; Raabe, J.; Fassbender, J.
Spin Wave Emission From Topological Spin Textures
SPIE Optics and Photonics Conference, Spintronics IX Symposium, 28.08. – 01.09.2016, San Diego, USA

61. Zhou, S.
Application of Ion Beams to Fabricate and Tune Properties of Dilute Ferromagnetic Semiconductors
24th International Conference on the Application of Accelerators in Research and Industry CAARI, 30.10. – 04.11.2016, Fort Worth, TX, USA
62. Zhou, S.
Ion implantation + sub-second annealing: a route towards hyperdoped semiconductors
26th annual meeting of MRS-J, 19. – 22.12.2016, Yokohama, Japan

Colloquia, lectures and talks (without conference talks)

63. Dimakis, E.
Growth and applications of III–V nanowires on Si substrates
Seminar, Institute of Electronic Structure and Laser, Foundation for Research and Technology-Hellas (FORTH), 05.08.2016, Heraklion, Greece
64. Dimakis, E.
Growth and applications of III-V nanowires on Si substrates
Seminar, ANKA - Karlsruhe Institute of Technology (KIT), 15.09.2016, Eggenstein-Leopoldshafen, Germany
65. Fassbender, J.
Nanomagnets – created and tailored by ions
Colloquium, 07.07.2016, Leipzig, Germany
66. Fassbender, J.
Nanomagnets – created and tailored by ions
Colloquium, 17.05.2016, Krakow, Polen
67. Fassbender, J.
Ion Beam Modification of Magnetic Materials
Colloquium CIC Nanogune San Sebastian, 16.02.2016, San Sebastian, Spanien
68. Fassbender, J.
Magnetic storage technology and spintronic applications
Invited talk at ICT/Applied Materials, 09.03.2016, Munich, Germany
69. Georgiev, Y.
Top-down fabrication of silicon nanowires
Seminar SENTECH "Plasma Process Technology", 07.04.2016, Berlin, Germany
70. Hlawacek, G.
Towards an analytic ion microscope
HZB Seminar, 09.02.2016, Berlin, Germany
71. Liu, Y.
Defect-induced magnetism in SiC: The new opportunity in spintronics
Seminar in Shanghai Institute of Ceramics, Chinese Academy of Sciences, 25.01.2016, Shanghai, China
72. Makarov, D.
Curved magnetic nanomembranes
Seminar at Argonne National Laboratory, 28.11.2016, Lemont, USA
73. Makarov, D.
Magnetic functionalities for flexible interactive electronics
Special seminar, Institute of Physics, Johannes Gutenberg-Universität Mainz, 29.08.2016, Mainz, Germany
74. Rebohle, L.; Schumann, T.; Prucnal, S.; Skorupa, W.
Ultra-fast thermal processing for thin metallic films
FLA Seminar, CERN, 19.10.2016, Geneva, Switzerland

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75. Schneider, H.
Terahertz spectroscopy of semiconductor nanostructures with a free-electron laser
Seminar, Chinese Academy of Engineering Physics, 18.10.2016, Chengdu, China
76. Schneider, H.
Terahertz spectroscopy of semiconductor nanostructures with a free-electron laser
Seminar, Ecole Normale Supérieure, Laboratoire Pierre Aigrain, 23.09.2016, Paris, France
77. Winnerl, S.
Unusual Coulomb Effects in Graphene
Seminar lecture in graduate college Electronic Properties of Carbon Based Nanostructures, 22.01.2016, Regensburg, Germany
78. Zhou, S.
Ion implantation + sub-second annealing: a route towards hyperdoped semiconductors
Invited lecture at Shanghai Institute of Microsystem and Information Technology, 05.07.2016, Shanghai, China
79. Zhou, S.
Application of Ion Beams to Fabricate and Tune Properties of Dilute Ferromagnetic Semiconductors
Invited lecture at Shanghai University, 07.07.2016, Shanghai, China
80. Zhou, S.
Ion implantation + sub-second annealing: a route towards hyperdoped semiconductors
Invited lecture at University of Electronic Science and Technology of China, 18.07.2016, Chengdu, China
81. Zhou, S.
Precisely doping semiconductors by ion implantation
Invited seminar at University of Warsaw, 22.09.2016, Warsaw, Poland

Conferences, workshops, colloquia and seminars

Organization of conferences and workshops

1. Arie, A.; Dunin-Borkowski, R.; Gemming, S.
620. Wilhelm and Else Heraeus Seminar: Interaction of shaped electron wavefunctions with light and matter
19. – 23.06.2016, Bad Honnef, Germany
2. Erbe, A.; Honolka, J.; Zahn, P.
NANO.NET International Workshop 2016
30.08. – 02.09.2016, Prague, Czech Republic
3. Faßbender, J.; Helm, M.
Institute Retreat (Off-site Seminar)
01./02.06.2016, Altenberg, Germany
4. Gemming, S.; Zahn, P.
MEMRIOX International Workshop 2016
25. – 27.09.2016, Lohmen, Germany
5. Heintze, C.; Hilger, I.; Bergner, F.; Posselt, M.
3rd International Workshop on ODS Materials
21./22.04.2016, Dresden, Germany
6. Hilger, I.; Heintze, C.
3rd ODISSEUS Workshop of the Oxide Dispersion Strengthened Steels group of young European Scientists
19./20.04.2016, Dresden, Germany
7. Krasheninnikov, A.V.; Skakalova, V.; Tapasztó, L.
EMRS Spring Meeting, Symposium Y: Graphene and related materials
02. – 06.05.2016, Lille, France
8. Krasheninnikov, A.V.; Foster, A.S.
International Physics Boat Workshop: Atomic structure of nanosystems from first-principles simulations and microscopy experiments
31.05 – 02.06.2016, Helsinki, Finland – Stockholm, Sweden
9. Krause, M.; Gemming, S.
FRIENDS²-Workshop: Advanced coating and characterization techniques
19./20.09.2016, Dresden, Germany
10. Makarov, D.; Gentile, P.; Ortix, C.; Ivan Vera-Marun, I.
TOP-SPIN 2: Spin and Topological phenomena in nanostructures
26./27.05.2016, Groningen, The Netherlands
11. Posselt, M.; Smith, R.; Uberuaga, B.
International Focus Workshop: Bridging-Time Scale Techniques and their Applications in Atomistic Computational Science
12. – 15.09.2016, Dresden, Germany
12. Schultheiß, H.; Chumak, A.
603. Wilhelm and Else Heraeus Seminar: Magnonics – Spin waves connecting charges, spins and photons
06. – 08.01.2016, Bad Honnef, Germany
13. Zhou, S.; von Bardeleben, J.; Gali, A.
International Workshop on Defect engineering in SiC for quantum technology
8./9.12.2016, Dresden, Germany

Colloquia

1. Back, Christian H.
University of Regensburg/Institute of Experimental and Applied Physics, Germany
Spin orbit fields at the Fe/GaAs(001) interface
24.11.2016
2. Carman, Greg P.
University of California, Los Angeles, USA
Magnetics + Mechanics + Nanoscale = Electromagnetics Future
05.12.2016
3. Ensslin, Klaus
ETH Zurich, Switzerland
Fermionic cavities in high-mobility electron gases
06.06.2016
4. Hono, Kazuhiro
University of Tsukuba/National Institute for Materials Science, Japan
Materials challenges for next-generation, high-density magnetic recording – media and read heads
02.09.2016
5. Lerch, Wilfried
centrotherm AG, Blaubeuren, Germany
Advanced thermal processing: Milli, nano, atto... and then?
09.09.2016
6. Rauschenbach, Stephan
Max-Planck-Institute for Solid State Research, Stuttgart, Germany
Electrospray ion beam deposition – Preparative mass spectrometry for the surface science of individual nonvolatile molecules and functional thin films
21.04.2016
7. Repp, Jascha
University of Regensburg/Institute of Experimental and Applied Physics, Germany
Scanning probe microscopy of individual molecules
30.05.2016
8. Sadowski, Janusz
Lund University, Sweden and Polish Academy of Sciences/Institute of Physics, Poland
Segregation of magnetic MnAs nanocrystals in GaMnAs layers, superlattices and nanowires
17.11.2016
9. Svensson, Bengt
University of Oslo, Norway
Ion implantation and defects: More fun... or graveyard?
09.09.2016
10. Vandervorst, Wilfried
IMEC and KU Leuven, Belgium
Advances in metrology for complex systems embedded in small volumes
21.01.2016
11. Wallner, Anton
Australian National University/Department of Nuclear Physics, Canberra, Australia
Long-lived radionuclides – unique signatures for astrophysical and environmental applications
19.10.2016
12. Zichner, Ralf
Fraunhofer Institute for Electronic Nano Systems (ENAS), Chemnitz, Germany
From basics via application to innovation: Facts, fiction... or fancy?
09.09.2016
13. Züttel, Andreas
École polytechnique fédérale de Lausanne (EPFL), Switzerland
From renewable energy to hydrogen and synfuels
17.03.2016

Seminars

1. Asano, Kohta
National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba, Japan
Nanometer-sized metal clusters embedded in immiscible matrix induced by hydrogenation
26.01.2016
2. Avasthi, Devesh K.
Amity University Noida, India
Synthesis and engineering of nanostructures by ion beams
02.05.2016
3. Barsukov, Igor
University of California, Riverside, USA
Magnonic condensates and nonlinear dynamics due to spin-currents in nanomagnets
11.08.2016
4. Bradley, Mark
Colorado State University/Departments of Physics and Mathematics, USA
Virtually defect-free ripples and terraced topographies produced by ion sputtering
17.06.2016
5. Chen, Wei
ETH Zurich/Institute of Theoretical Physics, Switzerland
Quantum tunneling of spin and its application in spectroscopy
14.07.2016
6. Fallarino, Lorenzo
CIC nanoGUNE, San Sebastian, Spain
Depth dependent magnetic correlation in metallic and oxide films and multilayers
28.11.2016
7. George, Antony
University of Jena/Institut für Physikalische Chemie, Germany
Atomically thin 2D materials for ultrathin device applications
16.06.2016
8. Guimarães, Filipe
Forschungszentrum Jülich, Germany
Dynamical current-induced spin excitations and their impact on magnetoresistance and Hall effects
12.10.2016
9. Jadidi, Mehdi
University of Maryland, USA
Graphene plasmonics for tunable Terahertz technology
04.05.2016
10. Jariwala, Deep
California Institute of Technology (CalTech), Pasadena, USA
Photocurrent microscopy and spectroscopy in atomically-thin semiconductor devices
05.09.2016
11. Johne, Robert
Max-Planck-Institut für die Physik komplexer Systeme, Dresden, Germany
Controlling the light-matter interaction in semiconductor microcavities
22.09.2016
12. Julin, Jaako
JAMK University of Applied Sciences, Jyväskylä, Finland
Advances and challenges in instrumentation for time-of-flight ERDA: gas ionization chambers and digitizing data acquisition
14.04.2016
13. Kieschnick, Michael
BAD Gesundheitsvorsorge und Sicherheitstechnik GmbH., München, Germany
Combinatorial material science with ion beams
08.04.2016

14. Liu, Enke
Max Planck Institute Chemical Physics of Solids Dresden and Chinese Academy of Sciences/Institute of Physics, Beijing, China
Magnetostructural phase transformations and new magnetic states in shape memory alloys
07.12.2016
15. López-Vidrier, Julian
Albert-Ludwigs-Universität Freiburg/Institut für Mikrosystemtechnik (IMTEK)
Silicon nanocrystal superlattices: From materials to devices
11.08.2016
16. Mantañez Huamán, Liz
Pontificia Universidad Católica del Perú, Lima and Technische Universität Ilmenau, Germany
Synthesis and characterization of wide bandgap semiconductors doped with terbium for electroluminescent devices
19.01.2016
17. Mazzinghi, Anna
LABEC, Florence, Italy
A window on cultural heritage applications at LABEC laboratory
10.08.2016
18. Mchedlidze, Teimuraz
Technische Universität Dresden, Germany
Smart light-induced crystallization of silicon
07.01.2016
19. Münzenrieder, Niko
University of Sussex, UK
Properties, characterisation and application of flexible oxide transistors
07.07.2016
20. Perov, Nikolai
Lomonosov Moscow State University, Russia
Functional magnetic materials and recent activities at Moscow State University Magnetism Department
23.02.2016
21. Prüfer, Thomas
Justus-Liebig-Universität Gießen, Germany
Open circuit voltage of lithium-ion-cells
26.01.2016
22. Rack, Alexander
European Synchrotron Radiation Facility (ESRF), Grenoble, France
Full-field X-ray imaging using hard synchrotron radiation: 4D microtomography and real-time imaging of fast processes
18.03.2016
23. Raschke, Markus B.
University of Colorado, Boulder, USA
Nano-focused multimodal imaging, control, and interaction dynamics: Ultrafast spectroscopy reaching the single molecule limit
12.12.2016
24. Sato, Nana
Keio University/Department of Physics, Tokyo, Japan
Current induced frequency shift of backward volume spin waves
25.02.2016
25. Sawicki, Maciej
Polish Academy of Sciences/Institute of Physics, Warsaw, Poland
On the critical behavior near the Curie Point in dilute ferromagnetic semiconductors
14.03.2016

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26. Scheunert, Gunther
Weizmann Institute of Science, Rehovot, Israel
Scanning probe lithography – Heat assisted magnetic recording in the lab
12.09.2016
 27. Secchi, Maria
University of Trento, Italy
Nanostructure formation on Germanium by ion irradiation
26.01.2016
 28. Shields, Brendan; Appel, Patrick
University of Basel, Switzerland
Nanoscale magnetic imaging with a single crystalline diamond probe
05.07.2016
 29. Shipman, Patrick
Colorado State University, Fort Collins, USA
Topological measures of order for pattern-forming systems
15.06.2016
 30. Tamaru, Shingo
National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba, Japan
Challenges and strategies for using spin torque oscillators for practical microwave applications
13.05.2016
 31. Tanyeli, Irem
Eindhoven University of Technology, The Netherlands
Low energy helium ion irradiation induced surface modification of metals
08.04.2016
 32. Titova, Aleksandra
Lomonosov Moscow State University/Department of Physics, Russia
Soft magnetic properties of Fe-based nanocrystalline films
25.02.2016
 33. van Delft, Falco
Nanovalk, Valkenswaard, The Netherlands
Nano-patterning for manipulation of bio-molecules and cells
05.10.2016
 34. Venanzi, Tommaso
Sapienza - Università di Roma, Italy
Near-field investigation of nanoantennas made of metallic Germanium
24.06.2016
 35. Vert, Matthieu
Université Pierre-et-Marie-Curie Paris, France
Modeling a stochastic process: Conversion of Schelling's segregation model into Markov chains
26.01.2016
 36. Wawro, Andrzej
Polish Academy of Sciences/Institute of Physics, Warsaw, Poland
Co/Mo(Au) multilayers: structure influence on magnetic anisotropy and interlayer coupling
05.10.2016
 37. Wolf, Daniel
Technische Universität Dresden, Germany
Electron tomography – 3D reconstruction of nanoscale materials
27.01.2016

Exchange of researchers

Guests at our Institute

1. Azad, F.
The University of Hong Kong; 01.12. – 16.12.2016
2. Bergen, L.
Technische Universität Wien, Austria; 14.11. – 31.12.2016
3. Bohovicova, J.
University of Technology Bratislava, Slovakia; 27.11. – 09.12.2016
4. Bradley, M.
Colorado State University, USA; 13.06. – 24.06.2016
5. Canpolat, C.
Ankara University, Turkey; 15.04. – 15.07.2016
6. Cheng, F.
Beijing Normal University, China; 01.01. – 29.08.2016
7. Clifford, E.
Dublin Institute of Technology, Ireland; 01.02. – 12.08.2016
8. Devaraj, M.
Indian Institute of Technology Madras, India; 10.09. – 09.10.2016
9. Drovosenko, A.
Moscow University, Russia; 23.11. – 30.11.2016
10. Edgecombe, M.
Ohio State University, USA; 17.05. – 17.08.2016
11. El-Said, A. S.
Mansoura University, Egypt; 07.01. – 15.01.; 02.06. – 23.06.2016
12. Garcia Hemme, E.
Universidad Complutense de Madrid, Spain; 01.09. – 01.12.2016
13. Gas, K.
University Wroclaw, Poland; 09.03.-15.03.; 09.09. – 18.09.2016
14. Gregor-Pawlowski, J.
Krakov University of Technology, Poland; 06.11. – 13.11.2016
15. Holybee, B.
University of Illinois at Urbana Champaign, USA; 17.09. – 02.10.2016
16. Hoy, C. G.
Imperial College London, U. K.; 04.06. – 28.07.2016
17. Huang, K.
Shanghai Institute of Microsystem and Information Technology (SIMIT), Shanghai, China; 19.09. – 25.10.2016
18. Jia, Q.
Shanghai Institute of Microsystem and Information Technology (SIMIT), Shanghai, China; 19.09. – 19.10.2016
19. Krupinski, M.
Institute of Nuclear Physics, Krakow, Poland; 20.07. – 31.07.2016
20. Madeira, T.
Universidade de Lisboa, Portugal; 01.03. – 31.12.2016

21. Marynowska, A.
Institute of Physics, Warsaw, Poland; 01.05. – 07.05.2016
22. Mesko, M.
University of Technology Bratislava, Slovakia; 27.11. – 09.12.2016
23. Mirek, R.
University of Warsaw, Poland; 04.07. – 14.08.2016
24. Nazarov, A.
University of Kyiv, Ukraine; 17.04. – 23.04.2016
25. Nekic, N.
Ruder Boskovic Institute Zagreb, Croatia; 20.06. – 02.07.2016
26. Perzanowski, M.
Institute of Nuclear Physics, Krakow, Poland; 20.07 – 31.07.; 06.11. – 13.11.2016
27. Otalóla, J. A.
Universidad Valparaiso, Chile; 26.08. – 11.09.2016
28. Ou, X.
Shanghai Institute of Microsystem and Information Technology (SIMIT), Shanghai, China; 17.10. – 25.10.2016
29. Sheka, D.
University of Kyiv, Ukraine; 25.01. – 21.02.2016
30. Smith, R.
Loughborough University, U. K.; 17.04. – 23.04.2016
31. Soroka, W.
Lublin University, Poland; 17.10. – 28.10.2016
32. Turek, M.
Lublin University, Poland; 24.04. – 30.04.2016
33. Vasin, A.
University of Kyiv, Ukraine; 17.04. – 23.04.2016
34. Volkov, O.
University of Kyiv, Ukraine; 15.01. – 15.03.2016
35. Yershov, K.
Institute for Theoretical Physics, Kyiv, Ukraine; 02.07. – 28.09.2016
36. Younas, M.
The University of Hong Kong; 11.01. – 25.01.2016
37. Yu, Y.
Harbin Institute of Technology, China; 01.01. – 12.09.2016
38. Zabala, Y.
Institute of Nuclear Physics, Krakow, Poland; 13.11. – 19.11.2016
39. Zhang, Z.
Hefei University, China; 01.04. – 31.12.2016
40. Zhu, J.
Shanghai Institute of Technical Physics (SITP), Shanghai, China; 01.01. – 31.12.2016
41. Zuk, J.
Lublin University, Poland; 24.04. – 30.04.2016

Projects

The projects are listed by funding institution and project starting date. In addition, the institute has several bilateral service collaborations with industrial partners and research institutions. These activities are not included in the following overview.

European Projects

1. 01/2013 – 12/2016 European Union EU
SPRITE – Supporting postgraduate research
Dr. J. v. Borany Phone: 0351 260 3378 j.v.borany@hzdr.de
2. 11/2013 – 10/2017 European Union EU
MatISSE – Materials' Innovations for a Safe and Sustainable nuclear in Europe
Dr. E. Altstadt Phone: 0351 260 2276 e.altstadt@hzdr.de
3. 01/2015 – 12/2018 European Union EU
FRIENDS² – Engineering of New Durable Solar Surfaces
Prof. S. Gemming Phone: 0351 260 2470 s.gemming@hzdr.de
4. 06/2015 – 05/2016 European Union EU
SlovakION – Supporting of Slovak Ion Beam Centre
Dr. J. v. Borany Phone: 0351 260 3378 j.v.borany@hzdr.de
5. 09/2015 – 08/2019 European Union EU
SOTERIA – Safe Long Term Operation of Light Water Reactors
Dr. E. Altstadt Phone: 0351 260 2276 e.altstadt@hzdr.de
6. 10/2015 – 12/2017 European Union EU
SMaRT (ERC Starting Grant) – Shapeable Magnetoelectronics
Dr. D. Makarov Phone: 0351 260 3273 d.makarov@hzdr.de
7. 02/2016 – 01/2020 European Union EU
IONS4SET – Single Electron Transistor
Dr. J. v. Borany Phone: 0351 260 3378 j.v.borany@hzdr.de

Helmholtz Association Projects

1. 03/2011 – 02/2016 Helmholtz-Gemeinschaft HGF
Functional Materials – Helmholtz Young Investigator Group
Dr. Shengqiang Zhou Phone: 0351 260 2484 s.zhou@hzdr.de
2. 07/2011 – 09/2018 Helmholtz-Gemeinschaft HGF
NANONET – International Helmholtz Research School on Nanoelectronics
Dr. A. Erbe Phone: 0351 260 2366 a.erbe@hzdr.de
3. 10/2011 – 09/2016 Helmholtz-Gemeinschaft HGF
MEMRIOX – Virtual Institute – Memory Effects in Resistive Ion-beam Modified Oxides
Dr. P. Zahn Phone: 0351 260 3121 p.zahn@hzdr.de
4. 01/2013 – 12/2017 Helmholtz-Gemeinschaft HGF
W3-Professorship TU Chemnitz
Prof. S. Gemming Phone: 0351 260 2470 s.gemming@hzdr.de
5. 01/2014 – 12/2018 Helmholtz-Gemeinschaft HGF
Spintronics – Helmholtz Young Investigator Group
Dr. A.M. Deac Phone: 0351 260 3709 a.deac@hzdr.de
6. 03/2014 – 02/2017 Helmholtz-Gemeinschaft HGF
Functional Materials – HGF Postdoc Dr. Yu Liu
Prof. M. Helm Phone: 0351 260 2260 m.helm@hzdr.de

7.	11/2014 – 10/2017	Helmholtz-Gemeinschaft	HGF
	Magnetism – HGF Postdoc Dr. K. Schultheiß		
	<i>Prof. J. Faßbender</i>	<i>Phone: 0351 260 3096</i>	<i>j.fassbender@hzdr.de</i>
8.	04/2016 – 03/2017	Helmholtz-Gemeinschaft	HGF
	Helmholtz-Enterprise-Fonds – GridLab		
	<i>Dr. H. Schultheiß</i>	<i>Phone: 0351 260 3243</i>	<i>h.schultheiss@hzdr.de</i>
9.	05/2016 – 04/2019	Helmholtz-Gemeinschaft	HGF
	THz Spectroscopy – HGF Postdoc Dr. A. Singh		
	<i>Prof. M. Helm</i>	<i>Phone: 0351 260 2260</i>	<i>m.helm@hzdr.de</i>
10.	10/2016 – 09/2017	Helmholtz-Gemeinschaft	HGF
	Helmholtz Exzellenznetzwerk – cfaed		
	<i>Dr. A. Erbe</i>	<i>Phone: 0351 260 2366</i>	<i>a.erbe@hzdr.de</i>

German Science Foundation Projects

1.	11/2010 – 12/2016	Deutsche Forschungsgemeinschaft	DFG
	Relaxation dynamics in graphene		
	<i>Dr. S. Winnerl</i>	<i>Phone: 0351 260 3522</i>	<i>s.winnerl@hzdr.de</i>
2.	09/2012 – 06/2017	Deutsche Forschungsgemeinschaft	DFG
	ATOMIX – Atomic mixing in semiconductor layers		
	<i>Dr. M. Posselt</i>	<i>Phone: 0351 260 3279</i>	<i>m.posselt@hzdr.de</i>
3.	01/2013 – 12/2016	Deutsche Forschungsgemeinschaft	DFG
	MWN – Magnetization dynamics in nanostructures		
	<i>Dr. J. Lindner</i>	<i>Phone: 0351 260 3221</i>	<i>j.lindner@hzdr.de</i>
4.	01/2013 – 10/2017	Deutsche Forschungsgemeinschaft	DFG
	Cluster of Excellence – Center for Advancing Electronics Dresden (cfaed)		
	<i>Prof. M. Helm</i>	<i>Phone: 0351 260 2260</i>	<i>m.helm@hzdr.de</i>
5.	02/2013 – 01/2016	Deutsche Forschungsgemeinschaft	DFG
	Nanostructured thermoelectrics		
	<i>Dr. P. Zahn</i>	<i>Phone: 0351 260 3121</i>	<i>p.zahn@hzdr.de</i>
6.	04/2014 – 03/2016	Deutsche Forschungsgemeinschaft	DFG
	Dynano – Spin wave excitations in periodic nanostructures		
	<i>Dr. K. Lenz</i>	<i>Phone: 0351 260 2435</i>	<i>k.lenz@hzdr.de</i>
7.	05/2014 – 04/2019	Deutsche Forschungsgemeinschaft	DFG
	Emmy Noether Junior Research Group – Magnonics		
	<i>Dr. H. Schultheiß</i>	<i>Phone: 0351 260 3243</i>	<i>h.schultheiss@hzdr.de</i>
8.	11/2014 – 10/2017	Deutsche Forschungsgemeinschaft	DFG
	Thermal spin-transfer torques		
	<i>Dr. J. Lindner</i>	<i>Phone: 0351 260 3221</i>	<i>j.lindner@hzdr.de</i>
	<i>Dr. A. M. Deac</i>	<i>Phone: 0351 260 3709</i>	<i>a.deac@hzdr.de</i>
9.	11/2014 – 10/2017	Deutsche Forschungsgemeinschaft	DFG
	All Optical Switching		
	<i>Dr. H. Schultheiß</i>	<i>Phone: 0351 260 3243</i>	<i>h.schultheiss@hzdr.de</i>
10.	10/2015 – 09/2018	Deutsche Forschungsgemeinschaft	DFG
	Ferromagnetic Silicon		
	<i>Dr. Shengqiang Zhou</i>	<i>Phone: 0351 260 2484</i>	<i>s.zhou@hzdr.de</i>

Federally and Saxony State Funded Projects

- | | | | |
|----|---|-----------------------|-------------------------|
| 1. | 01/2014 – 06/2017 | PT Jülich | BMBF |
| | In-situ TEM | | |
| | Prof. J. Fassbender | Phone: 0351 260 3096 | j.fassbender@hzdr.de |
| 2. | 10/2014 – 09/2017 | PT Jülich | BMBF |
| | InTerFEL – High-Field Spectroscopy in the THz Regime | | |
| | Dr. H. Schneider | Phone: 0351 260 2065 | h.schneider@hzdr.de |
| 3. | 02/2016 – 12/2018 | Sächsische Aufbaubank | SAB |
| | PolCarr-Sens – Electrically Polarizable Materials | | |
| | Dr. K. Wiesenhütter | Phone: 0351 260 2065 | k.wiesenhuetter@hzdr.de |

Personnel Exchange Projects and Society Chairs

- | | | | |
|----|---|---|----------------------|
| 1. | 01/2012 – 12/2018 | Institute of Electrical and Electronics Engineers | IEEE |
| | Magnetics Society German Chapter Chair | | |
| | Prof. J. Fassbender | Phone: 0351 260 3096 | j.fassbender@hzdr.de |
| 2. | 04/2015 – 03/2017 | Deutscher Akademischer Austauschdienst | DAAD |
| | Personnel exchange with Chile – ChileConMagnon | | |
| | Dr. K. Lenz | Phone: 0351 260 2435 | k.lenz@hzdr.de |
| 3. | 11/2015 – 04/2017 | Deutscher Akademischer Austauschdienst | DAAD |
| | Visit of Dr. Zhu | | |
| | Prof. M. Helm | Phone: 0351 260 2260 | m.helm@hzdr.de |
| 4. | 01/2016 – 12/2017 | Deutscher Akademischer Austauschdienst | DAAD |
| | Personnel exchange with Poland – Magnetic Anisotropy | | |
| | Dr. K. Potzger | Phone: 0351 260 3244 | k.potzger@hzdr.de |
| 5. | 01/2016 – 12/2017 | Deutscher Akademischer Austauschdienst | DAAD |
| | Personnel exchange with Poland – BEGIN | | |
| | Dr. S. Prucnal | Phone: 0351 260 2065 | s.prucnal@hzdr.de |
| 6. | 01/2016 – 12/2017 | Deutscher Akademischer Austauschdienst | DAAD |
| | Personnel exchange with China – Semiconductors | | |
| | Dr. S. Facsko | Phone: 0351 260 2987 | s.facsko@hzdr.de |
| 7. | 06/2016 – 09/2016 | Deutscher Akademischer Austauschdienst | DAAD |
| | Research visit Dr. K.V. Yershov | | |
| | Dr. D. Makarov | Phone: 0351 260 3273 | d.makarov@hzdr.de |
| 8. | 06/2016 – 05/2018 | Alexander-von-Humboldt-Stiftung | AvH |
| | Humboldt fellowship Dr. Y. Berencen | | |
| | Dr. L. Rebohle | Phone: 0351 260 3368 | l.rebohle@hzdr.de |

Bilateral Projects

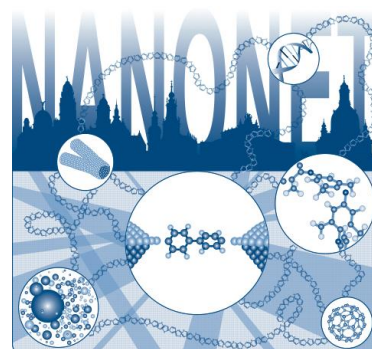
- | | | | |
|----|---|-----------------------|--------------------|
| 1. | 10/2012 – 12/2016 | Carl-Zeiss Microscopy | Industry |
| | Collaboration on analytical ion microscopy | | |
| | Dr. J. v. Borany | Phone: 0351 260 3378 | j.v.borany@hzdr.de |
| 2. | 11/2015 – 01/2019 | TU Hamburg-Harburg | University |
| | ZETA Membrane | | |
| | Dr. J. v. Borany | Phone: 0351 260 3378 | j.v.borany@hzdr.de |
| 3. | 11/2015 – 01/2016 | Robert-Bosch GmbH | Industry |
| | Ion Beam Analysis | | |
| | Dr. J. v. Borany | Phone: 0351 260 3378 | j.v.borany@hzdr.de |
| 4. | 09/2016 – 11/2016 | FEP | Fraunhofer |
| | Flash Lamp Annealing | | |
| | Dr. W. Skorupa | Phone: 0351 260 3612 | w.skorupa@hzdr.de |

Doctoral training programme

International Helmholtz Research School NANO NET

The Institute of Ion Beam Physics and Materials Research is coordinating the International Helmholtz Research School for Nanoelectronic Networks (IHRS NANO NET) supported by the Initiative and Networking Fund of the Helmholtz Association. The project started in October 2012. The total funding is 1.2 Mio. € for a period of six years.

The IHRS NANO NET is an international, interdisciplinary and thematically focused doctoral programme in the field of molecular electronics. The research school aims at attracting and promoting excellence by educating promising doctoral candidates with backgrounds in physics, chemistry, materials science and electrical engineering. During a period of three years PhD candidates benefit from well-structured, comprehensive training curricula and multiple mentorship, while performing cutting edge research projects within one of the 15 NANO NET research groups. Under the supervision of outstanding scientists leading the field of nanoelectronics, the doctoral candidates have the unique opportunity to contribute to the advancement of molecular electronics by developing strategies for the integration of single nano-sized building blocks into large interconnected networks.



The IHRS NANO NET fosters not only professional qualification but also personal development by equipping young graduates with competencies for successful careers in a wide variety of positions in academia and

industry. The NANO NET International Workshop 2016 was conducted in the Czech capital Prague and was attended by about 60 participants from 9 countries. In 2016 we attracted new students: Türkan Bayrak, Jingjing Ye, Filip Kilibarda, Bilal Khan, Alexander Strobel and Hagen Sekulla who are from Turkey, China, Serbia, Pakistan, and Germany, respectively. Three senior students concluded their PhD degree at the TU Dresden: Congratulations to Drs. Mimi Hetti, Banu Iyisan, and Bezu Teschome.

The consortium

- Helmholtz-Zentrum Dresden-Rossendorf (HZDR)
- Technische Universität (TU) Dresden
- Leibniz Institute of Polymer Research (IPF) Dresden
- Fraunhofer Institute for Ceramic Technologies and Systems (IKTS) Dresden
- Nanoelectronic Materials Laboratory (NaMLab) gGmbH Dresden



For further information please contact the NANO NET coordinator, Dr. Peter Zahn (nanonet@hzdr.de) or visit the IHRS NANO NET website: www.hzdr.de/nanonet.

Experimental equipment

Accelerators, ion implanters and other ion processing tools

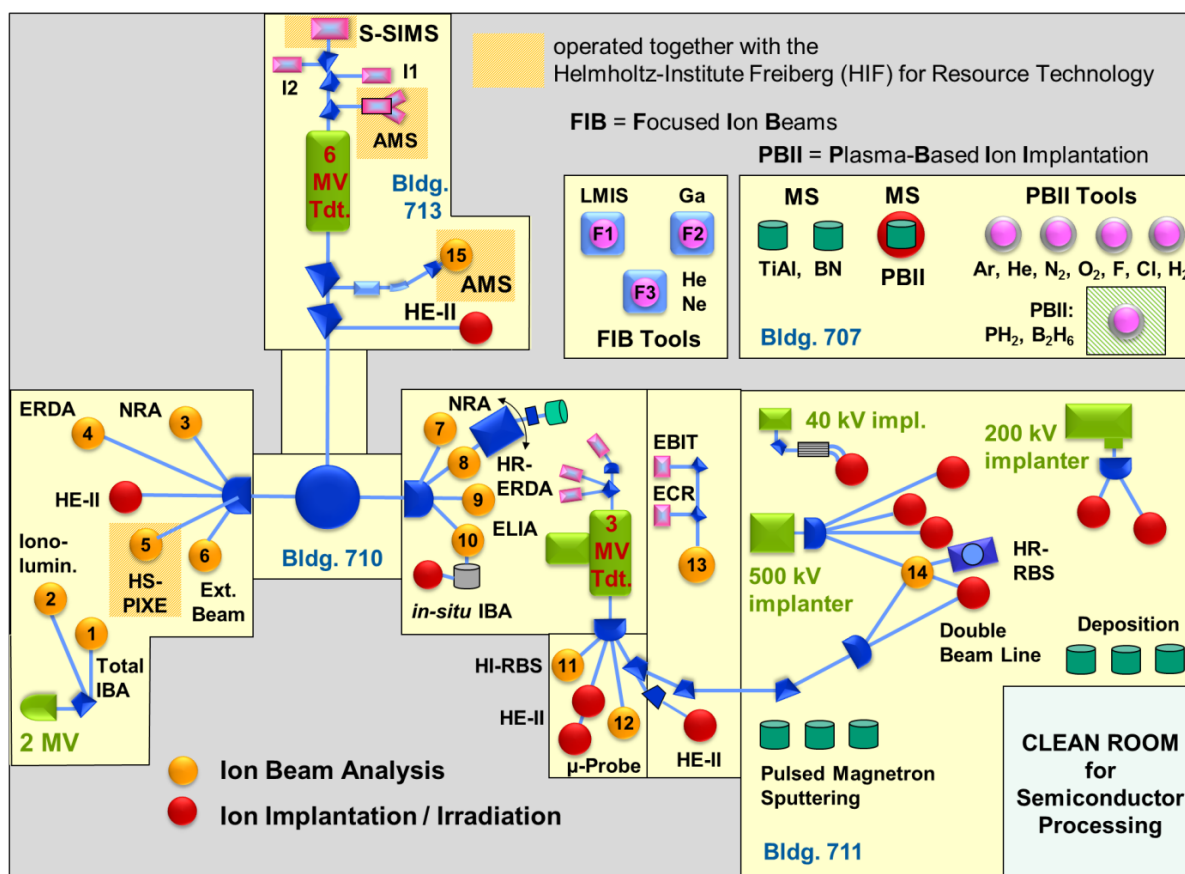
Van de Graaff Accelerator (VdG)	2 MV	<i>TuR Dresden, DE</i>
Tandetron Accelerator (T1)	3 MV	<i>HVEE, NL</i>
Tandetron Accelerator (T2)	6 MV	<i>HVEE, NL</i>
Low-Energy Ion Implanter	0.1 – 40 kV	<i>Danfysik, DK</i>
High-Current Ion Implanter	20 – 200 kV	<i>Danfysik, DK</i>
High-Energy Ion Implanter	20 – 500 kV	<i>HVEE, NL</i>
Plasma Immersion Ion Implantation	5 – 30 kV	<i>UK, DE; Home-built</i>
Mass-Separated Focused Ion Beam (FIB) (15 nm, variable ions)	10 – 30 keV >10 A/cm ²	<i>Orsay Physics, FR</i>
ORION NanoFab FIB Tool (Including GIS and Nanopatterning)	He, Ne ions, 10 – 35 kV, Resolution < 2 nm	<i>Carl Zeiss Microscopy, DE</i>
Highly-Charged Ion Facility	25 eV – 25 keV × Q Q = 1 ... 40 (Xe)	<i>Home-built</i>
Ion-Beam Sputtering	200 – 2000 V	<i>Home-built</i>
UHV Ion Irradiation (Ar, He, etc.)	0 – 5 kV Scan 10 × 10 mm ²	<i>Cremer, DE; VG, USA</i>

Ion beam analysis (IBA)

A wide variety of advanced IBA techniques are available at the MV accelerators (see figure).

RBS	Rutherford Backscattering Spectrometry	(1), (10), (11), (12)	<i>VdG, T1, T2, HIM</i>
RBS/C	RBS + Channelling	(1), (11), (12)	<i>VdG, T1, T2</i>
HR-RBS	High-Resolution RBS	(10), (14)	<i>T1</i>
ERDA	Elastic Recoil Detection Analysis	(1), (4)	<i>VdG, T2</i>
PIXE	Particle-Induced X-ray Emission	(1), (5), (6), (12)	<i>VdG, T1, T2</i>
PIGE	Particle-Induced γ Emission	(6), (12)	<i>T1, T2</i>
NRA	Nuclear Reaction Analysis	(3), (7), (11), (12)	<i>T1, T2</i>
NRRA	Nuclear Resonance Reaction Analysis	(3), (7), (11)	<i>T1, T2</i>
NMP	Nuclear Microprobe	(12)	<i>T1</i>
AMS	Accelerator Mass Spectrometry (focused to long-lived radionuclides: ¹⁰ Be, ²⁶ Al, ³⁶ Cl, ⁴¹ Ca, ¹²⁹ I)	(15)	<i>T2</i>

Some stations are equipped with additional process facilities enabling *in-situ* IBA investigations during ion irradiation, sputtering, deposition, annealing etc.



Schematic overview of the HZDR Ion Beam Center

Other particle-based analytical techniques

SEM	Scanning Electron Microscope (S4800 II)	1 – 30 keV + EDX	Hitachi, JP
TEM	Transmission Electron Microscope (Titan 80-300 with Image Corrector)	80 – 300 keV + EDX, EELS	FEI, NL
TEM	Transmission Electron Microscope (Talos F200X)	20 – 200 keV + SuperX EDX	FEI, NL
HIM	Scanning Ion Microscope (ORION NanoFab with He, Ne ions)	He, Ne ions 10 – 35 kV + RBS, SIMS	Carl Zeiss Microscopy, DE
FIB/SEM	Focused Ion/Electron Cross Beam (NVision 40 with Elphy Plus Litho)	0.5 – 30 keV + EDX, EBSD	Carl Zeiss Microscopy, DE
AES	Auger Electron Spectroscopy	+ XPS	Thermo Fisher Scientific, UK
LEEM	Low-Energy Electron Microscope (Spec-LEEM-III)	0 eV – 4.5 keV Resolution < 6 nm + AES	Elmitec, DE
CEMS	Mössbauer Spectroscopy	⁵⁷ Fe source	Home-built

Photon-based analytical techniques

XRD/XRR	X-Ray Diffractometers	Cu-K α	
	θ - θ Powder D8		<i>Bruker, DE</i>
	θ -2 θ 4-Circle D5005		<i>Siemens, DE</i>
	θ - θ 4-Circle Empyrean		<i>PANalytical, NL</i>
	θ -2 θ 4+2-Circle SEIFERT XRD3003-HR		<i>General Electric, US</i>
SE	Spectroscopic Ellipsometry	250 – 1700 nm	<i>Woollam, US</i>
UV-Vis	Solid Spec 3700 DUV	190 – 3300 nm	<i>Shimadzu, JP</i>
FTIR	Fourier-Transform Infrared Spectrometer	600 – 7000 cm ⁻¹	<i>Nicolet, US</i>
FTIR	Fourier-Transform Infrared Spectrometer	50 – 15000 cm ⁻¹	<i>Bruker, DE</i>
	Ti:Sapphire Femtosecond Laser	78 MHz	<i>Spectra Physics, US</i>
	Femtosecond Optical Parametric Osci.		<i>APE, DE</i>
	Ti:Sapphire Femtosecond Amplifier	1 kHz, 250 kHz	<i>Coherent, US</i>
	Femtosecond Optical Parametric Amplifier		<i>Light Conversion, LT</i>
THz-TDS	Terahertz Time-Domain Spectroscopy	0.1 – 4 THz	<i>Home-built</i>
Raman	Raman Spectroscopy	> 45 cm ⁻¹	<i>Jobin-Yvon-Horiba, FR</i>
	In-situ Raman Spectroscopy	> 100 cm ⁻¹	<i>Jobin-Yvon-Horiba, FR</i>
PL	Photoluminescence (10 – 300 K)	300 – 1600 nm	<i>Jobin-Yvon-Horiba, FR</i>
TRPL	Time-Resolved Photoluminescence	τ = 3 ps – 2 ns τ > 5 ns	<i>Hamamatsu Phot., JP</i> <i>Stanford Research, US</i>
EL	Electroluminescence	300 – 1600 nm	<i>Jobin-Yvon-Horiba, FR</i>
	Optical Split-Coil Supercond. Magnet	7 T	<i>Oxford Instr., UK</i>
PR	Photomodulated Reflectivity	300 – 1600 nm	<i>Jobin-Yvon-Horiba, FR</i>
PLE	Photoluminescence Excitation	300 – 1600 nm	<i>Jobin-Yvon-Horiba, FR</i>
OES	Optical Emission Spectroscopy	250 – 800 nm	<i>Jobin-Yvon-Horiba, FR</i>

Magnetic thin film deposition and analysis

PLD	Pulsed Laser Deposition		<i>SURFACE, DE</i>
MFM	Magnetic Force Microscope	~ 50 nm resol.	<i>VEECO;DI, US</i>
SQUID MPSM	Supercond. Quantum Interference Device	\pm 7 T	<i>Quantum Design, US</i>
SQUID VSM	Vibrating Sample Magnetometer	\pm 7 T	<i>Quantum Design, US</i>
MOKE	Magneto-Optic Kerr Effect (in-plane)	\pm 0.35 T	<i>Home-built</i>
MOKE	Magneto-Optic Kerr Effect (perpend.)	\pm 2 T	<i>Home-built</i>
FR-MOKE	Frequency-Resolved Magneto-Optic KE	\pm 1.1 T	<i>Home-built</i>
SKM	Scanning Kerr Microscope		<i>Home-built</i>
	Kerr Microscope		<i>Evico Magnetics, DE</i>
TR-MOKE	Time-Resolved MOKE (Pump-Probe)		<i>Home-built</i>
VNA-FMR	Vector Network Analyzer Ferromagnetic Resonance	50 GHz	<i>Agilent, DE;</i> <i>Home-built</i>
Cryo-FMR	Variable-Temperature Ferromagnetic Resonance	3 – 300 K	<i>Attocube, DE;</i> <i>Home-built</i>

ME	Magnetoellipsometer		<i>LOT, DE; AMAC, US</i>
μBLS	Brillouin Light Scattering Microscope	± 0.8 T, 532 nm & 491 nm	<i>Home-built</i>
SKM	Scanning Kerr Microscope with RF Detection (Spectrum Analyzer)	± 0.5 T, 40 GHz	<i>Home-built</i>
MT-50G	High Frequency Magneto-transport Setup	± 1.5 T, 50 GHz 250 ps	<i>Home-built</i>

Other analytical and measuring techniques

STM/AFM	UHV Scanning Probe Microscope (variable T)		<i>Omicron, DE</i>
AFM	Atomic Force Microscope (Contact, Tapping, Spreading)		<i>Bruker, US</i>
AFM	Atomic Force Microscope (with c-AFM, SCM-Module)		<i>Bruker, US</i>
	Dektak Surface Profilometer		<i>Bruker, US</i>
	Micro Indenter/Scratch Tester		<i>Shimatsu, JP</i>
MPMS	Mechanical Properties Measurement System – Stretcher		<i>Home-built</i>
MS	Mass Spectrometers (EQP-300, HPR-30)		<i>HIDEN, DE/US</i>
	Wear Tester (pin-on disc)		<i>Home-built</i>
LP	Automated Langmuir Probe		<i>Impedans, IE</i>
HMS	Hall Measurement System	2 – 400 K, ≤ 9 T	<i>LakeShore, US</i>
	Van-der-Pauw HMS Ecopia	300 K & LNT, 0.5 T	<i>Bridge Technol., US</i>
MTD	Magneto-Transport Device	300 K, ≤ 3 T	<i>Home-built</i>
RS	Sheet-Rho-Scanner		<i>AIT, KOR</i>
DLTS	Deep Level Transient Spectroscopy	+ I-U/C-V 10 – 300 K, 1 MHz	<i>PhysTech, DE</i>
IV / CV	Photocapacitance (+I-V/G-V)	250 – 2500 nm	<i>Home-built</i>
IV / CV	I-V and C-V Analyzer		<i>Keithley, US</i>
IV / CV	I-V and C-V Semi-Automatic Prober	-60 – 300 °C	<i>Süss, DE; Keithley, US</i>
IV	I-V Prober	4.2 – 600 K	<i>LakeShore, Agilent, US</i>

Deposition and processing techniques

Physical Deposition	Sputtering DC/RF, Evaporation	<i>Nordiko, UK</i>
	Electron Beam Evaporation System	<i>Leybold Optics, DE</i>
	Thermal Evaporation	<i>Bal-Tec, LI</i>
	High Power Impulse Magnetron Sputtering	<i>Melec, DE</i>
Molecular Beam Epitaxy	III-V Semiconductors	<i>Riber, FR</i>
Chemical Vapour Deposition	Plasma Enhanced CVD (for a-Si, a-Ge, SiO ₂ , SiON, Si ₃ N ₄)	<i>Oxford Instr., UK</i>
Atomic Layer Deposition	Al ₂ O ₃ , HfO ₂ , SiO ₂	<i>Ultratech, US</i>
Dry Etching	ICP-RIE, CF ₄ , SF ₆ , C ₄ F ₈	<i>Sentech, DE</i>
	RIBE, Ø 6", Ar, CF ₄	<i>Roth & Rau, DE</i>

Etching/Cleaning	incl. Anisotropic Selective KOH Etching	
Photolithography	Mask-Aligner, 2 µm-Level	Süss, DE
	Direct Laser Writer DWL 66FS	Heidelberg Instr., DE
	MicroWriter	LOT QD, DE
Electron Beam Lithography	Raith 150-TWO: Ø 6", 10 nm resol.	Raith, DE
	e-Line Plus: Ø 4", 10 nm resol.	Raith, DE
Thermal Treatment	Room Temperature – 2000 °C	
Furnace		InnoTherm, DE
Rapid Thermal Annealing JETFIRST 100		JIPELEC, FR
Rapid Thermal Annealing AW 610		Allwin21, USA
Flash-Lamp Units (0.5 – 20 ms)		Home-built;FHR/DTF, DE
RF Heating (Vacuum)		JIPELEC, FR
Laser Annealing (CW, 808 nm, 450 W)		LIMO, DE;
Laser Annealing (30 ns pulse, 10 Hz, 308 nm, 500 mJ)		COHERENT, USA
Bonding Techniques	Ultrasonic Wire Bonding	Kulicke & Soffa, US
Cutting, Grinding, Polishing		Bühler, DE
TEM Sample Preparation	Plan View and Cross Section incl. Ion Milling Equipment	Gatan, US

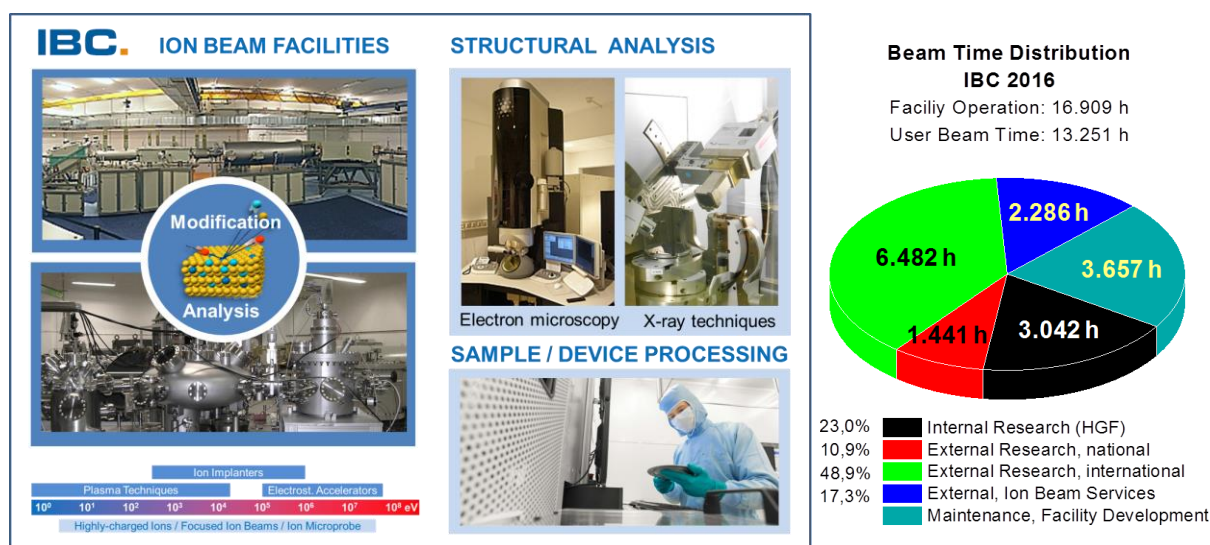
Hot cells laboratory

Mechanical testing of neutron irradiated structural materials	Max. Total Activity 5 TBq (⁶⁰ Co), -150 ... +315 °C	
Fracture mechanics testing	Max. Load ±50 kN	MTS, US
Charpy impact testing	300 J	WPM Leipzig, DE
Small punch test	10 kN	Hegewald & Peschke, DE
Specimen preparation	Electrical Discharge Machining	AGIE, DE
Depth sensing nanoindentation/ AFM	UNAT, Load Range 1 – 500 mN	ASMEC, DE

User facilities and services

Ion Beam Center (IBC)

The Ion Beam Center (IBC) at HZDR combines various machines (electrostatic accelerators, ion implanters, plasma-based equipment) into a unique facility primarily used for ion beam modification and ion beam analysis of materials. The available energy range spans from a few eV to almost 100 MeV with a respective interaction depth in solids between 0.1 nm to 10 μm . In addition to standard broad beams also focused (down to 1 nm) and highly-charged (charge state up to +45) ion beams are available. In addition to these ion beam facilities, structural analysis (electron microscopy and spectroscopy, X-ray scattering techniques) and sample or device processing under clean-room conditions can be utilized at IBC to provide a “complete” user service. As a part of the IBC, the IBC together with the Helmholtz Institute Freiberg for Resource Technology (HIF, www.hzdr.de/HIF) operates the DREAMS (DREsden AMS = accelerator mass spectrometry) facility which is used for the determination of long-lived radionuclides (^{10}Be , ^{26}Al , ^{35}Cl , ^{41}Ca and others) with ultimate sensitivity. A schematic overview of the IBC including the description of the main beam lines and experimental stations is given on page 97 of this Annual Report. In 2016 about 13.300 beam time hours were delivered for about 450 users from 22 countries worldwide performing experiments at IBC or using the capabilities for ion beam services.



IBC activities are efficiently integrated into various Helmholtz programmes within the research field “Matter”, but also in the Helmholtz cross-programme activities “Mineral Resources”, “Materials Research for Energy Technologies”, and “Helmholtz Energy Materials Foundry”. From 2013 the IBC has been recognized as a large-scale facility within the “BMBF Verbundforschung” programme promoting long-term collaborations with universities. In addition, from 2015 the IBC has the status of a Helmholtz LK–II user facility.

The IBC demonstrates its outstanding status within the international community of ion beam facilities by the following features:

- the IBC has provided ion beam technology as a user and competence centre for ion beam applications for more than 30 years. With respect to user beam time hours the IBC is internationally leading and has been supported by numerous national and European grants and by industry.
- the IBC activities cover both ion beam modification as well as ion beam analysis (IBA). Experienced staff is present to support IBC users and to enhance the experimental capabilities to their needs.

- the operation of IBC is accompanied by a strong in-house research at the affiliated host “Institute of Ion Beam Physics and Materials Research”, both in experiment and theory. This allows in-depth research in targeted research areas as well as the exploration of new application fields for ion beam technology.

Following the rules of a European and national user facility, access for scientific experiments to IBC is provided on the basis of a proposal procedure (www.hzdr.de/IBC) via the common HZDR user facility portal **HZDR-GATE** (gate.hzdr.de). Due to the availability of multiple machines and versatile instrumentation, continuous submission of IBC proposals can be offered (no deadline). The scientific quality of the proposals is steadily evaluated and ranked by an external international User Selection Panel. For successfully evaluated proposals, users get free access to IBC facilities for their experiments which can be often realized within three months after proposal submission. The use of the IBC facilities includes the scientific and technical support during planning, execution and evaluation of the experiments. For AMS samples preparation two chemical laboratories are available.

The IBC strongly supports the commercial exploitation of ion beam technology of partners from industry which is essential for materials science applications. For ion beam services the HZDR Innovation GmbH (spin-off of the HZDR) – www.hzdr-innovation.de – provides a direct and fast access to the IBC facilities based on individual contracts. The close collaboration between IBC and HZDR Innovation GmbH is considered as a Helmholtz reference model aiming to develop innovative strategies for an effective contribution of large-scale facilities for economic innovation. Currently, about 18% of the total IBC beam time is provided for commercial ion beam services.

The IBC pursues two strategic objectives for further development. On one hand, it is focused on the successful operation as an international user facility to sustain its position as a leading European ion beam center in the fields of materials and interdisciplinary research. This requires the continuous development and extension of ion beam technologies. Special emphasis is directed towards new R&D areas and communities which so far have not been aware of the advantages of ion technology. On the other hand, cutting edge scientific challenges, in particular for the modification and understanding of the function of materials at the nanoscale down to the atomic level, is addressed in order to achieve new functionalities and to design novel materials. These challenges comprise:

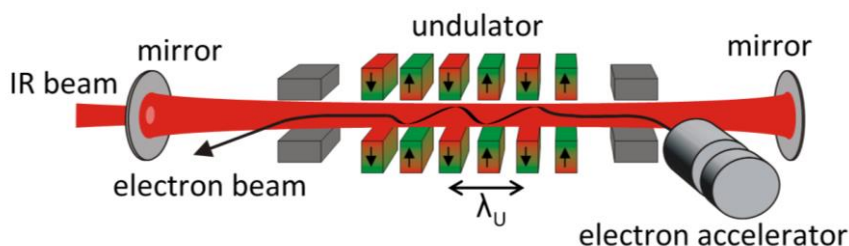
- the extended application of low-energy ions for modification and analysis of ultra-thin films, surfaces, 2D materials and molecular layers based on a new low-energy ion nano-engineering technology platform which has recently started with the installation of first tools,
- the investigation of material states and modifications far from equilibrium induced by high-energy density excitations with cluster and highly-charged ions,
- the exploration of new IBA approaches with unprecedented lateral resolution, ultimate detection limit and high sample throughput, e.g. for resource technology. With projects like “PIXE-Camera” and “Super-SIMS”, both in close collaboration with the HIF, the interdisciplinary work of IBC will be substantially extended towards the application of ion beam analysis methods in geosciences and resource technology.

Recently, some new ion beam tools or end-stations have been commissioned which will attract new users by cutting-edge experimental instrumentation. The ion microscope *ORION NanoFab* (He/Ne ions, 10 – 40 keV) is now in routine operation with unique possibilities for surface imaging, nano-fabrication and, for the first time, elemental analysis using ion beam analysis methods. The cluster tool at the 6 MV accelerator – in operation since 2015 – allows *in situ* deposition and analysis investigations at temperature of up to 800 °C. Analytical techniques comprise Rutherford backscattering spectrometry, Raman spectroscopy and Vis-NIR spectroscopic ellipsometry. In 2016, the ion microprobe at HZDR at the 3 MV tandemron has been basically upgraded (electronics, control software etc.) which now delivers the possibility to use – in parallel or sequential – several IBA techniques with a spatial resolution of about 3 µm and a user friendly operation mode. Results of DREAMS work contributed to outstanding publications in *Nature* and *Science* related to geomorphology and cosmogenic events.

For more detailed information please contact Dr. Johannes von Borany (j.v.borany@hzdr.de) or Dr. Stefan Facsko (s.facsko@hzdr.de) and visit the IBC webpage: www.hzdr.de/IBC.

Free Electron Laser FELBE

ELBE is an acronym for the free-electron laser (FEL) at the Electron Linear accelerator with high Brilliance and low Emittance (ELBE) located at the Helmholtz-Zentrum Dresden-Rossendorf, Germany. The heart of ELBE is a superconducting linear accelerator operating in cw mode with a pulse repetition rate of 13 MHz. The electron beam (40 MeV, 1 mA max.) is guided to several laboratories where secondary beams (particle and electromagnetic) are generated. Two free-electron lasers (U27-FEL and U100-FEL) produce intense, coherent electromagnetic radiation in the mid and far infrared, which is tunable over a wide wavelength range (4 – 250 μm) by changing the electron energy or the undulator magnetic field. Main parameters of the infrared radiation produced by FELBE are as follows:



Wavelength λ	4 – 22 μm 18 – 250 μm	FEL with undulator U27 FEL with undulator U100
Pulse energy	0.01 – 2 μJ	depends on wavelength
Pulse length	1 – 25 ps	depends on wavelength
Repetition rate	13 MHz	3 modes: • cw • macropulsed (> 100 μs , < 25 Hz) • single pulsed (Hz ... kHz)

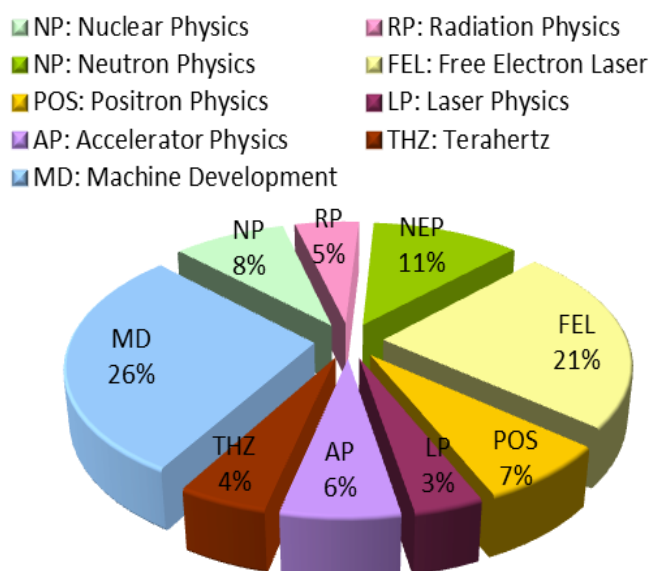
The free electron laser is a user facility. Applications for beam time can be submitted twice a year, typically by April 15 and October 15.

Typical applications are picosecond pump-probe spectroscopy (also in combination with several other femtosecond lasers, which are synchronized to the FEL), near-field microscopy and nonlinear optics. The FELBE facility also serves as a far-infrared source for experiments at the High-Field Laboratory Dresden (HLD) involving pulsed magnetic fields up to 70 T.

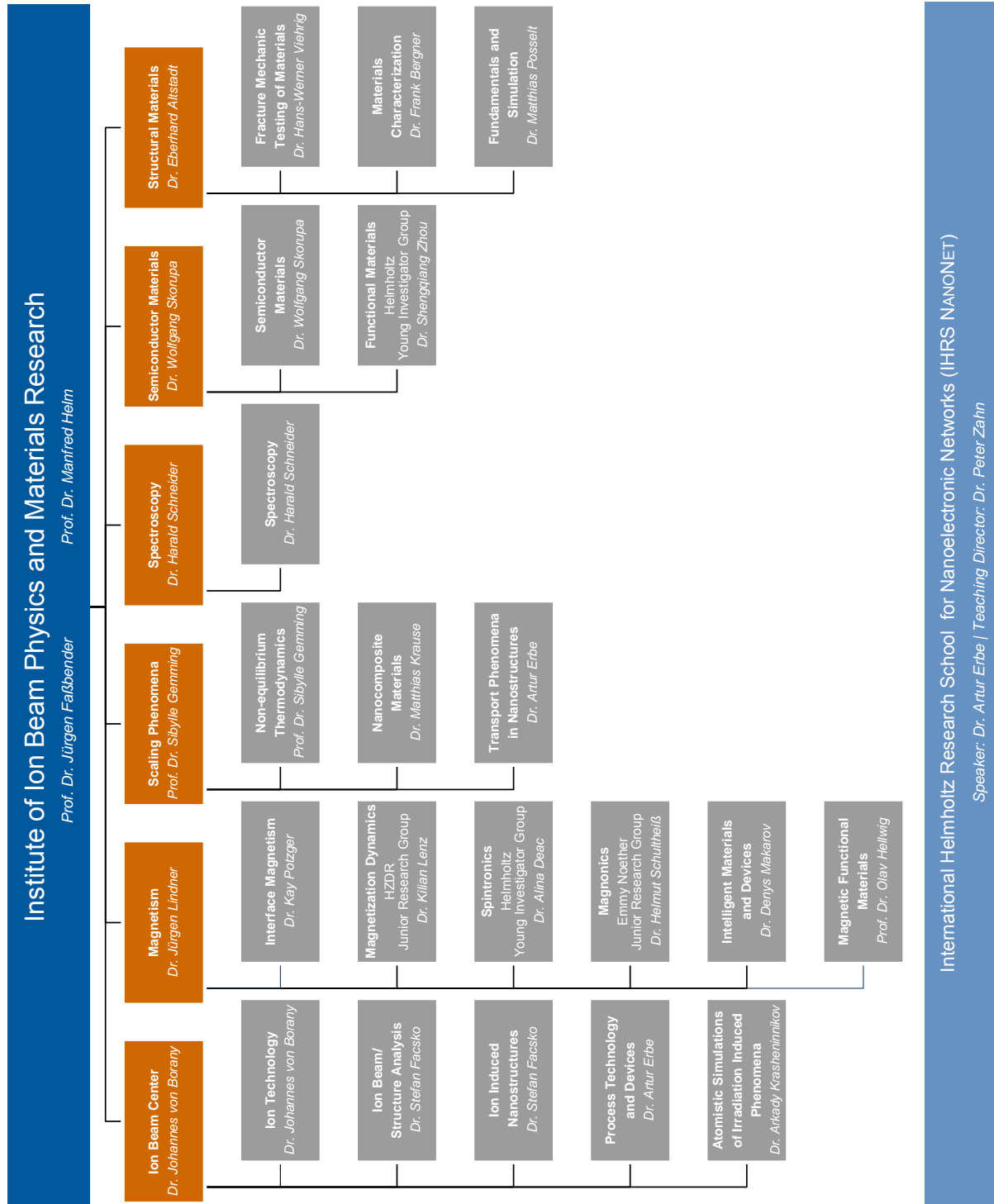
The statistics shows that the FEL used 1255 hours beam time of the ELBE accelerator. This corresponds to 21 % of total beam time, which is again distributed among internal and external users.

For further information please contact:
Prof. Manfred Helm (m.helm@hzdr.de)
or visit the FELBE webpage
www.hzdr.de/FELBE.

Beamtime Distribution at ELBE



Organization chart



List of personnel 2016

DIRECTORS		OFFICE	
Prof. Dr. M. Helm, Prof. Dr. J. Faßbender		S. Gebel, S. Kirch	
SCIENTIFIC STAFF			
Permanent staff		Non-permanent	
Dr. C. Akhmadaliev	Dr. G. Müller	Dr. R. Bali	Dr. Y. Liu (P)
Dr. E. Altstadt	Dr. F. Munnik	Dr. Y. Berencén (P)	Dr. D. Makarov
Dr. F. Bergner	Dr. M. Posselt	Dr. R. Böttger	Prof. Dr. W. Möller (P)
Dr. L. Bischoff	Dr. K. Potzger	Dr. V. Cantelli	Dr. I. Mönch (P)
Dr. J. von Borany	Dr. L. Rebohle	Dr. A. Deac	Dr. M. Neubert (P)
Dr. E. Dimakis	Dr. H. Schneider	Dr. M. Engler	Dr. N. Nishida (P)
Dr. A. Erbe	Dr. W. Skorupa	Dr. D. Erb	Dr. J. Osten (P)
Dr. S. Facsko	Dr. A. Ulbricht	Dr. C. Fowley	Dr. P. Pandey (P)
Prof. Dr. S. Gemming	Dr. H.-W. Viehrig	Dr. J. Ge (P)	Dr. A. Pashkin
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Dr. V. Heera	Dr. S. Winnerl	Dr. M. Ghorbani Asl	Dr. R. Rana
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Dr. M. Krause	Dr. S. Zhou	Dr. C. Heintze	Dr. A. Scharf
Dr. J. Lindner		Dr. R. Heller	Dr. H. Schultheiß (P)
		Prof. Dr. O. Hellwig	Dr. K. Schultheiß (P)
		Dr. G. Hlawacek (P)	Dr. T. Sebastian (P)
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		Dr. J. Julin	Dr. S. Stienen
		Dr. A. Kákay	Dr. O. Volkov (P)
		R. Kaltofen (P)	Dr. X. Wang (P)
		Dr. T. Kosub	Dr. K. Wiesenhütter (P)
		Dr. A. Krashennikov	Dr. R. Wilhelm
		Dr. K. Lenz	Dr. D. Wolf (P)
		Dr. M. Lenz	Dr. R. Yankov (P)
		Dr. K. Li (P)	Dr. O. Yildirim (P)
		Dr. B. Liedke	

TECHNICAL STAFF**Permanent staff**

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H. Lange	A. Schneider	
U. Lucchesi	A. Scholz	

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S. Klengel
T. Schönherr (P)
A. Weißig (P)
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B. Duan	S. Kretschmer	B. Teschome	

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