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Suppressed Auger scattering and tunable light emission of Landau-quantized massless Kane electrons

D. B. But,^{1,2} M. Mittendorff,^{3,4} C. Consejo,¹ F. Teppe,¹ N. N. Mikhailov,⁵ S. A. Dvoretiskii,⁵
C. Faugeras,⁶ S. Winnerl,³ M. Helm,³ W. Knap,^{1,2} M. Potemski,⁶ and M. Orlita^{6,7,*}

¹Laboratoire Charles Coulomb, UMR CNRS 5221,
University of Montpellier, Montpellier 34095, France

²International Research Centre CENTERA, Institute of High Pressure Physics,
Polish Academy of Sciences, 01-142 Warsaw, Poland

³Helmholtz-Zentrum Dresden-Rossendorf, PO Box 510119, 01314 Dresden, Germany

⁴Universität Duisburg-Essen, Fakultät für Physik, 47057 Duisburg, Germany

⁵A.V. Rzhanov Institute of Semiconductor Physics, Siberian Branch,
Russian Academy of Sciences, Novosibirsk 630090, Russia

⁶Laboratoire National des Champs Magnétiques Intenses,
CNRS-UGA-UPS-INSA-EMFL, 25 rue des Martyrs, 38042 Grenoble, France

⁷Charles University, Faculty of Mathematics and Physics,
Ke Karlovu 5, 121 16 Prague 2, Czech Republic

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The Landau level laser has been proposed a long time ago as a unique source of monochromatic radiation, widely tunable in the THz and infrared spectral ranges using an externally applied magnetic field. In spite of decades of efforts, this appealing concept never resulted in the design of a reliable device. This is due to efficient Auger scattering of Landau-quantized electrons, which is an intrinsic non-radiative recombination channel that eventually gains over cyclotron emission in all materials studied so far: in conventional semiconductors with parabolic bands, but also in graphene with massless electrons. The Auger processes are favored in these systems by Landau levels (or their subsets) equally spaced in energy. Here we show that this scheme does not apply to massless Kane electrons in gapless HgCdTe alloy, in which undesirable Auger scattering is strongly suppressed and the sizeable cyclotron emission observed, for the first time in the case of massless particles. The gapless HgCdTe thus appears as a material of choice for future technology of Landau level lasers.

When a magnetic field is applied to a solid, the continuous density of electronic states transforms into a set of discrete energy levels, known as Landau levels (LLs). Electrons excited in such a ladder may recombine with emission of photons. This process can be viewed as an inverse of cyclotron resonance¹ and it is referred to as cyclotron emission²⁻⁵. The idea to construct the LL laser by achieving stimulated cyclotron emission² is as old as the experimental realization of the very first laser itself⁶. The tunability represents a great advantage of this concept. The strength of the magnetic field defines the spacing between LLs, and therefore, also the emission frequency of the laser. This frequency typically corresponds to the far infrared (terahertz) spectral range. The successful realization of the LL laser^{2,7-10} would thus bridge

the so-called terahertz gap^{11,12}, which still exists despite a considerable effort of several generations of physicists.

There is, however, a fundamental difficulty that has been recognized during the very first attempts to test the concept of a LL laser. In conventional materials, with quadratically dispersing electronic bands, and therefore, with equally spaced LLs, $E_N^S = \hbar\omega_c(N + 1/2)$ for $N = 0, 1, 2$, cyclotron emission is an intrinsically weak process^{2,3,13}. This is due to two independent processes which compete with cyclotron emission: reabsorption of cyclotron radiation² and Auger inter-LL recombination^{14,15} (Fig. 1a). The latter becomes at higher pumping rates extremely efficient for electrons within the equidistant LL ladder, and in consequence, the excited LLs become depleted with a characteristic rate proportional to the number of electrons in this level ($\tau^{-1} \propto n$). Stronger pumping, either optical or electrical, in systems with parabolic bands does not enhance cyclotron emission, but instead, increases the probability of Auger scattering. The inversion of population thus cannot be achieved. Electrons promoted via Auger processes to higher LLs then relax via non-radiative channels, most often by emission of optical phonons³.

To overcome this obstacle, various systems with non-equidistant LLs have been proposed as appropriate materials for a LL laser^{2,16}. These may be found, for instance, in different narrow-gap materials or in the valence band of zinc-blende semiconductors. So far, stimulated cyclotron emission was achieved only in bulk germanium in which a non-equidistant spacing of valence-band LLs was created by crossed electric and magnetic fields¹⁷, alas in the regime close to the electrical breakdown of this material. In other attempts, quantum cascade structures have also been tested to achieve efficient cyclotron emission^{18,19}.

Renewed impetus to investigate cyclotron emission came along with the fabrication of graphene, which was considered as an ideal system with non-equidistant LLs

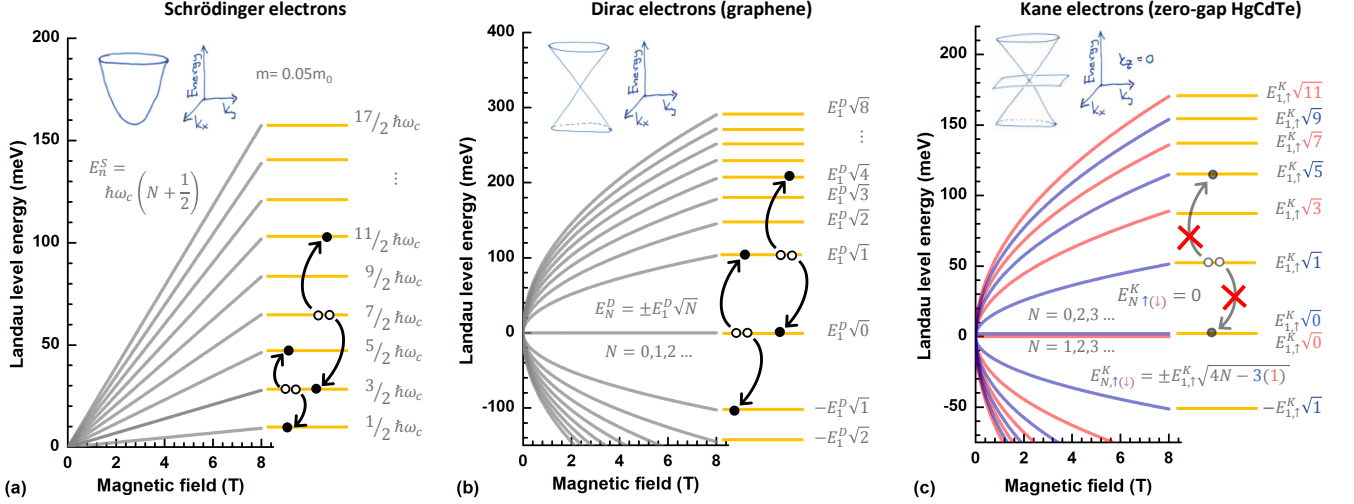


FIG. 1. Landau level ladders and possible paths for Auger scattering of massive and massless electrons. The B -dependence and spacing of Landau levels for conventional massive (Schrödinger) electrons, Dirac electrons in graphene and massless Kane electrons (at $k = 0$), in part (a), (b) and (c), respectively. The arrows in LL ladders correspond to the Auger scattering processes discussed in the main text.

for the implementation of the LL laser^{10,20}. Its conical band may be viewed as an extreme case of non-parabolicity and implies a specific sequence of Landau levels, $E_N^D = \pm v\sqrt{2e\hbar BN}$ for $N = 0, 1, 2, \dots$, with the \sqrt{N} spacing, typical of massless Dirac electrons (Fig. 1b). However, to the best of our knowledge, no cyclotron emission, let alone the Landau level laser based on graphene, has been reported so far, despite pertinent theoretical predictions and numerous experimental attempts^{10,20–26}.

This surprising lack of optical emission seems to be still a consequence of Auger scattering. Initially, such processes were expected to vanish in Landau-quantized graphene, but finally they were experimentally proven to be present and very efficient^{27,28}. The reason behind is simple. The LL spectrum in graphene: $E_N^D \propto \sqrt{N}$ for $N = 0, 1, 2, \dots$, includes subsets of equidistant levels. For instance, LLs with indices $N = 0, \pm 1, \pm 4, \pm 9, \dots$ or $0, \pm 2, \pm 8, \pm 18, \dots$, are equally spaced and may thus support, similar to conventional materials, efficient Auger recombination (Fig. 1b). Notably, such a conclusion is valid not only for graphene, but for any other system with a conical band described by the Dirac Hamiltonian for particles with a zero rest mass.

Fortunately, one may find other systems with massless electrons, described by a Hamiltonian that differs from the Dirac one. In such systems, equidistantly spaced LLs may be avoided and undesirable Auger scattering possibly suppressed. In this paper, we test Landau-quantized 3D massless Kane electrons^{29–32} against the efficiency of Auger scattering and against the possibility of efficient LL emission. This type of massless electrons appears in conventional bulk zinc-blende semiconductors when their energy band gap – the parameter responsible for the gen-

eration of the non-zero band mass of electrons and holes, and therefore, also Schrödinger-like behavior of these carriers – vanishes, for instance, in the ternary compound HgCdTe explored in this paper.

In magnetic fields, the conical band of 3D massless Kane electrons splits into LLs, or more precisely, Landau bands, which resemble those of massless Dirac electrons³³:

$$E_{N,\sigma}^K(k) = \pm v\sqrt{2e\hbar B(N - 1/2 + \sigma/2) + \hbar^2 k^2}, \quad (1)$$

where $N = 1, 2, 3, \dots$ is the LL index, k the momentum along the applied magnetic field and v the velocity parameter. However, the splitting due to spin is given by $\sigma = \pm 1/2$ and differs from the one known for genuine Dirac-type electrons ($\sigma = \pm 1$). As a result, the LL spectrum of massless Kane electrons (Fig. 1c): $E_N^K \propto \sqrt{N}$ for $N = 0, 1, 3, 5, \dots$, does not include any subsets of equally spaced LLs at $k = 0$, where the maxima in the density of states are. In contrast, 3D Dirac electrons with $\sigma = \pm 1$ display at $k = 0$ the spacing $E_N^D \propto \sqrt{N}$ where $N = 0, 1, 2, 3, \dots$, which is identical to that of 2D graphene.

Moreover, there exists a set of narrowly spaced LLs, which originates from the weakly dispersing (nearly flat) band. In the simplest approach, these LLs do not disperse with B : $E_{n,\sigma}^K = 0$ for $n = 0, 2, 3, 4, \dots$. Excitations from this flat band to conduction-band LLs constitute a dominant contribution to the optical response at low photon energies and follow the standard selection rules, $N \rightarrow N \pm 1$, for electric-dipole transitions^{31,32,34}.

To test Landau-quantized electrons in gapless HgCdTe for the presence of Auger scattering, we have employed a degenerate pump-probe technique, using the free-electron

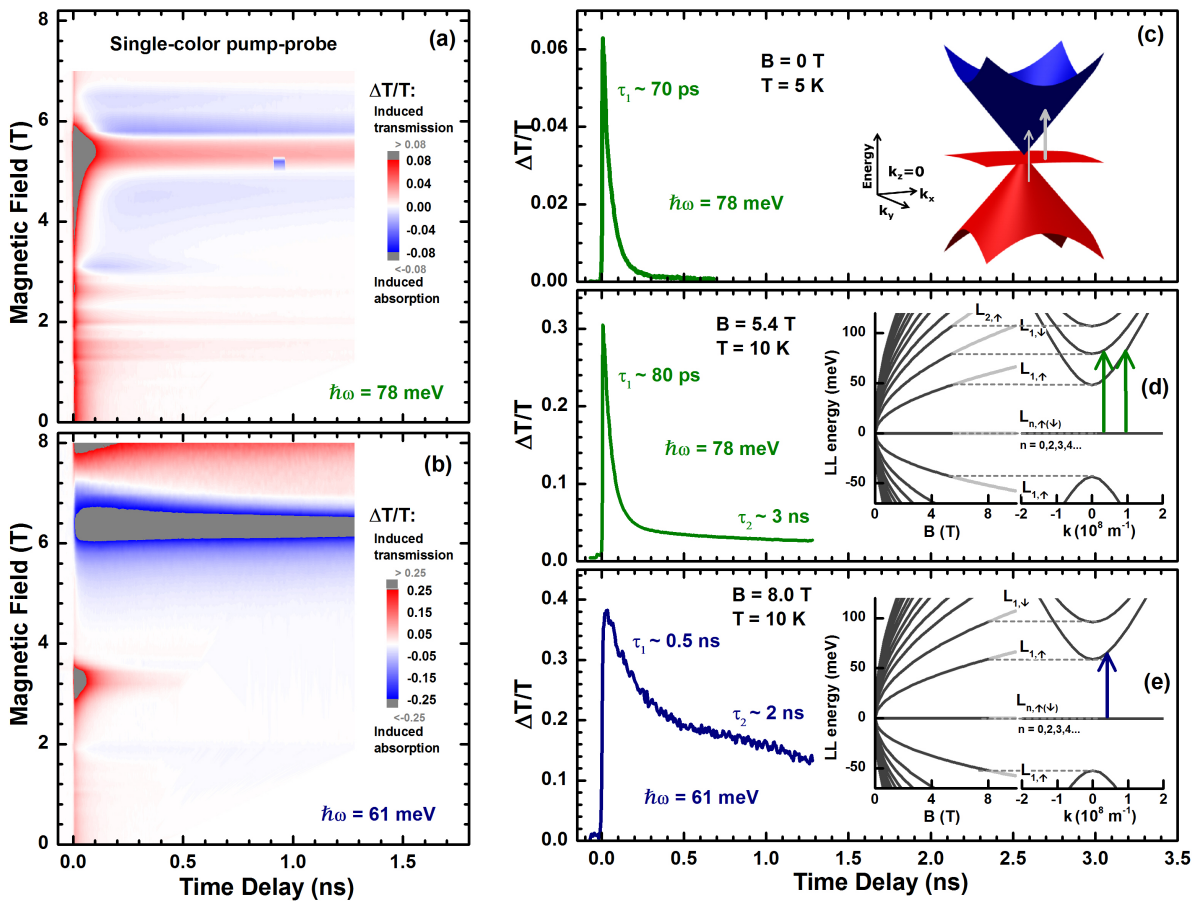


FIG. 2. **Pump-probe experiments and Auger scattering of Landau-quantized massless Kane electrons.** (a) and (b) False-color maps of induced transmission $\Delta T/T$ measured at $T = 10$ K and for laser photon energies $\hbar\omega = 78$ meV (upper panel) and 61 meV (lower panel) as a function of the time delay and magnetic field, showing both, pronounced induced transmission ($\Delta T/T > 0$) and absorption ($\Delta T/T < 0$). (c-e) Selected pump-probe transients. The inter-LL excitations in (d) and (e) follow the standard selection rules for electric-dipole transitions which are active in linearly polarized light, $N \rightarrow N \pm 1$, and schematically shown in the corresponding insets.

laser emitting ~ 3 -ps-long pulses in the mid-infrared range. The results are plotted in Figs. 2a and b, in the form of false color-maps, for two different photon energies of the laser. These maps show pump-probe transients, *i.e.*, the pump-induced relative change of transmission, $\Delta T/T$, as a function of the applied magnetic field and time delay between pump and probe pulses. One immediately concludes that $\Delta T/T$ undergoes a fairly complex evolution in both, its amplitude and sign, on a characteristic time scale extending up to nanoseconds.

To illustrate the observed behavior in greater detail, three selected pump-probe traces are plotted in Figs. 2c-e. At $B = 0$, the observed induced transmission (bleaching) is primarily due to electrons excited from the fully occupied heavy-hole-like flat band to the upper conical band (inset of Fig. 2c). The contribution of excitations from the lower cone is considerably weaker. The induced transmission exhibits a nearly mono-exponential decay, with a characteristic time, $\tau = 70$ ps, which is comparable to that in other gapless systems with conical bands,

such as graphene³⁵. This relatively fast relaxation may be associated with the emission of phonons.

When the magnetic field is applied, one observes a pronounced change in $\Delta T/T$ transients. For particular values of B , the second component develops, with a characteristic decay time in the nanosecond range. The appearance of this slow component clearly coincides with the resonant pumping into the bottom of Landau bands. This is illustrated in Fig. 2d, where the $\Delta T/T$ trace collected at $B = 5.4$ T and $\hbar\omega = 78$ meV is plotted. In this particular case, electrons are pumped from the flat band to two lowest lying conduction-band LLs, see the inset of Fig. 2d. The faster component, with a relaxation rate not much different from the zero-field one ($\tau \approx 80$ ps), can be associated with the relaxation of electrons excited into the lowest Landau band ($E_{1,\uparrow}^K$). **This relaxation of electrons from states with finite-momentum towards $k \approx 0$ is probably due to emission of phonons.** Most likely, electrons relax via emission of acoustic phonons. The slow component, appearing only when electrons are

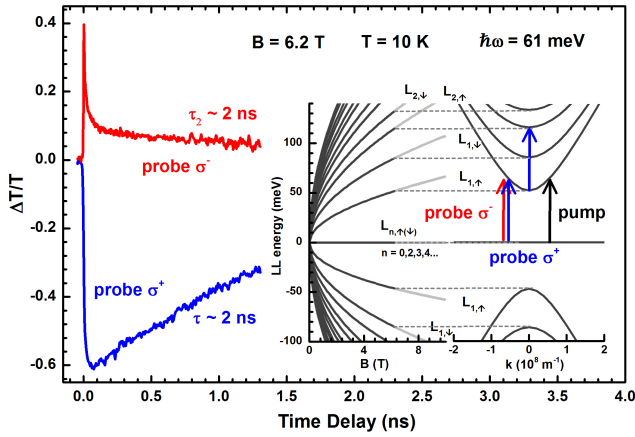


FIG. 3. **Pump-probe experiments using circularly polarized probe beam.** Pump-probe transients collected at $B = 6.2$ T using the laser photon energy of $\hbar\omega = 61$ meV and circularly polarized probe beam. The involved excitations, which follow the standard electric-dipole selection rules for right and left circularly polarized radiation: $N \rightarrow N + 1$ and $N \rightarrow N - 1$, respectively, are schematically shown in the inset.

198 pumped resonantly from/to states around $k \approx 0$, reflects
 199 the existence of long-living electrons at the bottom of the
 200 $E_{1,\downarrow}^K$ Landau band. Another example of the slowly decay-
 201 ing component in the pump-probe transient is shown in
 202 Fig. 2e. This $\Delta T/T$ trace was collected at $B = 8$ T and
 203 $\hbar\omega = 61$ meV, when only the lowest LL in the conduction
 204 band ($E_{1,\uparrow}^K$) is pumped close to zero-momentum states.

205 Experiments with circularly polarized radiation repre-
 206 sent another way to visualize how the relaxation of photo-
 207 excited electrons in gapless HgCdTe is slowed-down when
 208 the magnetic field is applied. Pump-probe transients col-
 209 lected using a linearly polarized pump pulse, but with a
 210 circularly polarized probe pulse are depicted in Fig. 3.
 211 In this particular case, electrons are excited by the pump
 212 pulse from the flat band to the lowest lying LL only,
 213 as schematically shown by the vertical arrow in the in-
 214 set of Fig. 3. The transient recorded using σ^- -polarized
 215 probe shows positive $\Delta T/T$, with an initial fast decay
 216 due to the relaxation of electrons to the bottom of $E_{1,\uparrow}^K$
 217 level and slower component indicating a nanosecond-long
 218 lifetime of excited electrons around $k \approx 0$. Different be-
 219 havior, *i.e.*, strong induced absorption $\Delta T/T < 0$, is
 220 observed when the probing beam is σ^+ polarized. In this
 221 latter case, the induced absorption is primarily due to the
 222 $E_{1,\uparrow}^K \rightarrow E_{2,\uparrow}^K$ transition which becomes activated due to
 223 electrons promoted to the $E_{1,\uparrow}^K$ level by the pump pulse.
 224 This excitation corresponds to the fundamental cyclotron
 225 mode, which becomes resonant with the laser photon en-
 226 ergy ($\hbar\omega = 61$ meV) just at the selected magnetic field
 227 of $B = 6.2$ T.

228 Let us now discuss the main findings of our pump-
 229 probe experiments. These show that electrons in Landau-
 230 quantized gapless HgCdTe relax with relatively long de-
 231 cay times, at the scale of nanoseconds. In addition, the
 232 presented pump-probe experiments were performed at

233 relatively high photon fluences, $\sim 0.5 \mu\text{J}\cdot\text{cm}^{-2}$. These
 234 translate, for the chosen photon energy, into the photon
 235 flux of 10^{14} cm^{-2} per pulse. Due to the relatively large
 236 absorption coefficient³¹, which in the linear regime, at
 237 $\hbar\omega \approx 70$ meV and $B = 0$, reaches $\lambda \approx 3 \times 10^3 \text{ cm}^{-1}$, a
 238 significant part of the flux is absorbed in the explored 3-
 239 μm -thick layer of gapless HgCdTe. This corresponds to
 240 more than 10^{16} cm^{-3} electrons promoted from the flat
 241 band to the upper conical band by a single pump pulse.
 242 Since there is no decay observed at the scale of the pulse
 243 duration (~ 3 ps), this number serves as a rough estimate
 244 of the electron density in the sample just after the pump
 245 pulse.

246 One may compare this carrier density and the deduced
 247 relaxation time, with the results obtained in conven-
 248 tional semiconductors, where a strong decrease of relax-
 249 ation time with the electron density has been reported:
 250 $\tau \propto n^{-1}$, see Refs. 3, 13, and 36. In conventional semi-
 251 conductors, the nanosecond decay times are only found
 252 at very low carrier densities in the excited LL, in the
 253 range of 10^{12} - 10^{13} cm^{-3} . For the carrier density close
 254 to 10^{16} cm^{-3} in a parabolic-band semiconductor, one ex-
 255 pects the electron relaxation time to drop down to the
 256 sub-picosecond range³ due to efficient Auger processes.
 257 This is more than three orders of magnitude less than
 258 relaxation times observed in gapless HgCdTe in a mag-
 259 netic field. This indicates that Auger scattering is indeed
 260 strongly suppressed for Landau-quantized massless Kane
 261 electrons as compared to other materials explored so far.
 262 We interpret this suppression as a direct consequence of
 263 the specific LL spectrum, which does not include any
 264 subset of equidistant levels (around $k \approx 0$).

265 Since the rate inter-LL Auger scattering follows the de-
 266 generacy of Landau levels ($\propto B$) and the strength of in-
 267 teraction between Landau-quantized electrons ($\propto \sqrt{B}$)³⁷,
 268 we expect similarly long lifetimes also at lower magnetic
 269 fields. Let us also note that the observed lifetime of
 270 electrons, even though unusually long as compared to
 271 any so-far explored Landau-quantized semiconductor or
 272 semimetal³, remains still significantly shorter than spon-
 273 taneous cyclotron radiative lifetime. This latter time
 274 can be for massless electrons estimated using the simple
 275 formula¹⁰, $\tau_{\text{sp}}^{-1} \sim \alpha\omega_c(v/c)^2$, where α is the fine structure
 276 constant, c speed of light in vacuum and the character-
 277 istic ω_c cyclotron frequency ($\tau_{\text{sp}} \approx 10^{-6}$ s in the THz
 278 range for $v = 10^6$ m/s). Hence, there still exist other
 279 (non-radiative) channels which dominate the recombina-
 280 tion, such as electron-phonon interaction giving rise to
 281 emission of phonons.

282 The observed slowing-down in the relaxation dynamics
 283 of electrons induced by the magnetic field, with the the
 284 overall lifetime of photo-excited electrons in the nanosec-
 285 ond range, call for cyclotron emission experiments. We
 286 have performed such experiments in the THz spectral
 287 range, which is the most relevant one for applications of
 288 the future Landau level laser technology. To generate
 289 cyclotron emission, the sample was placed in a super-
 290 conducting coil at liquid helium temperature and electri-

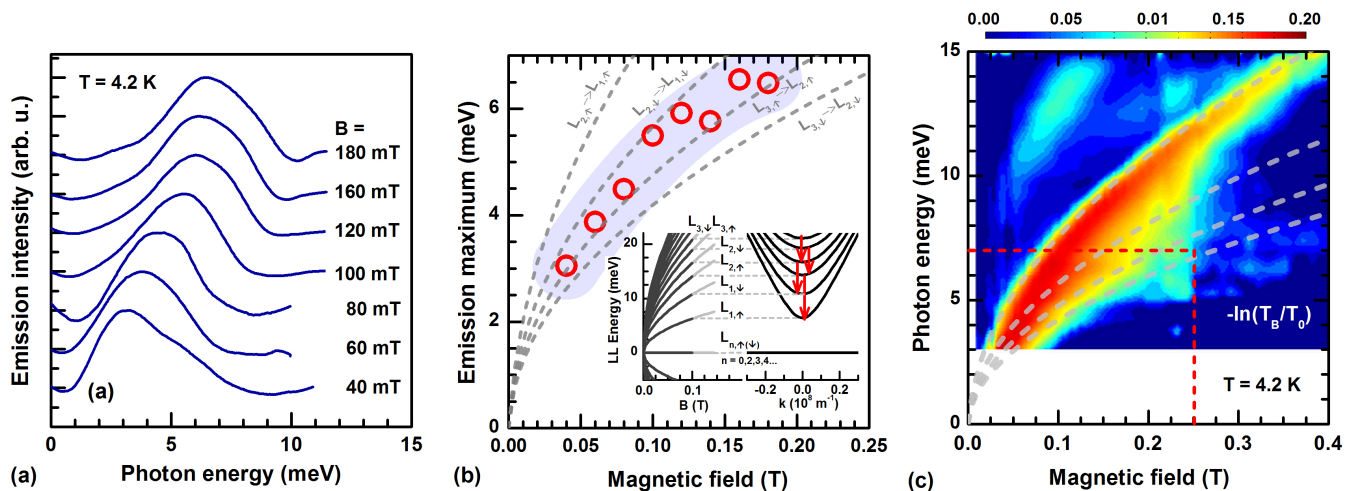


FIG. 4. **Cyclotron-emission and absorption of 3D massless Kane electrons.** (a) Cyclotron emission spectra measured on gapless HgCdTe sample kept in the liquid helium at selected values of the magnetic field. (b) Experimentally deduced maxima of emission compared with theoretically expected cyclotron modes (dashed lines), see the text. The grey area correspond to the error bar of cyclotron emission maxima. The LL spectrum with schematically depicted emission lines (by vertical arrows) is shown in the inset. (c) The false-color map of magneto-absorbance, primarily due to cyclotron resonance absorption, reprinted from Ref. 31. The dashed lines show the theoretically expected positions of cyclotron modes, the same ones as in (b). The red dashed lines shows the range of magnetic fields and photon energies in which cyclotron emission was explored.

291 cally pumped, using ms-long current pulses, see Meth- 323
 292 ods section. The emitted radiation was analyzed using 324
 293 a photoconductive InSb detector, with a spectrally 325
 294 narrow (cyclotron-resonance-like) response, tunable by a 326
 295 specially dedicated coil. The collected cyclotron emission 327
 296 spectra are plotted in Fig. 4a for selected values of the 328
 297 magnetic field applied to the sample. 329

298 A brief inspection of the emission spectra leads us to 332
 299 a conclusion that they feature a single emission band. 333
 300 Its position in the spectrum is tunable in the THz range 334
 301 by a relatively low magnetic field (tens of millitesla) and 335
 302 it roughly follows a \sqrt{B} dependence, which is typical of 336
 303 massless electrons (Fig. 4b). This also agrees well with 337
 304 results of cyclotron absorption experiments performed on 338
 305 the same sample, see Fig. 4c and Ref. 31. However, a 339
 306 closer look at the lineshape suggests that several emis- 340
 307 sion modes actually contribute. Theoretically, one indeed 341
 308 expects several cyclotron emission modes in the interval 342
 309 given by the linewidth (FWHM ~ 4 meV). In Fig. 4b, 343
 310 the dashed lines show the positions of cyclotron emis- 344
 311 sion modes with the final states of electrons in the four 345
 312 lowest lying conduction-band LLs: $E_{N+1,\uparrow(\downarrow)}^K \rightarrow E_{N,\uparrow(\downarrow)}^K$ 346
 313 for $N = 1$ and 2. It is thus the spacing of these cyclotron 347
 314 modes, together with primarily elastic scattering 348
 315 processes, which is responsible for the observed width of 349
 316 the emission band. The mutual intensities of individual 350
 317 modes then determine the position of the maximum of 351
 318 the emission band. At higher B , the relative weight of 352
 319 modes from higher LLs increases, which reflects the dis- 353
 320 tribution of electrons among LLs established by electrical 354
 321 pumping and which leads to a slowing-down of the \sqrt{B} 355
 322 dependence.

323 Even though the cyclotron emission observed in the 324
 325 used configuration is primarily due to spontaneous re- 326
 327 combination, it is worth to discuss conditions required 328
 329 to obtain stimulated emission and gain. To achieve 329
 330 the light amplification comparable to, *e.g.*, quantum 330
 331 cascade lasers³⁸, the gain coefficient has to approach 331
 332 $g = \Delta n \cdot \sigma \sim 10 \text{ cm}^{-1}$, where Δn stands for the popu- 332
 333 lation inversion and $\sigma = \lambda^2 / (2\pi) \cdot (\tau_{\text{tot}} / \tau_{\text{sp}})$ is the stimu- 333
 334 lated emission cross-section³⁹. The typical spontaneous 334
 335 cyclotron emission lifetime of massless electrons in the 335
 336 THz range reaches $\tau_{\text{sp}} \sim 1 \mu\text{s}$ and the total lifetime, 336
 337 which dominantly reflects elastic scattering, may be esti- 337
 338 mated (its lower limit) from the linewidth of the emission 338
 339 line: $\tau_{\text{tot}} \sim 1 \text{ ps}$. Taking the characteristic wave length of 339
 340 $\lambda = 300 \mu\text{m}$, we obtain the cross-section $\sigma \sim 10^{-10} \text{ cm}^{-2}$. 340
 341 This implies the necessity to achieve the population in- 341
 342 version $\Delta n > 10^{11} \text{ cm}^{-3}$. Our simple estimate, see Sup- 342
 343 plementary materials⁴⁰, indicates that such a popula- 343
 344 tion inversion should be achievable at least in the pulsed 344
 345 mode.

343 To conclude, we have demonstrated, for the first time, 343
 344 cyclotron emission of massless electrons. This emission 344
 345 was observed in gapless HgCdTe – a system hosting 345
 346 3D massless Kane electrons. The existence of sizeable 346
 347 cyclotron emission is directly related to their particu- 347
 348 lar Landau level spectrum, which comprises only non- 348
 349 equidistantly spaced levels. The systems hosting massless 349
 350 Kane electrons are thus promising candidates for an ac- 350
 351 tive medium of a Landau level laser, which would, in this 351
 352 particular case, operate in the THz and infrared spectral 352
 353 ranges and would be widely tunable by very low magnetic 353
 354 fields. 354

355 Methods

356 *Sample growth.* The sample was grown using standard
357 molecular-beam epitaxy on a (013)-oriented semi-insulating
358 GaAs substrate. The growth sequence started with ZnTe and
359 CdTe transition regions, followed by the MCT epilayer with
360 gradually changing cadmium content x . The prepared MCT
361 layer contains a region with $x \approx 0.17$ of thickness $d \approx 3.2 \mu\text{m}$.
362 For more details about the explored sample see Ref. 31.

363 *Pump-probe spectroscopy.* The free-electron laser FELBE
364 provided frequency-tunable Fourier-limited radiation pulses.
365 In the experiments described in this paper, photon energies
366 of $\hbar\omega = 61$ and 78 meV were chosen. The pulse duration was
367 about 3 ps, the repetition rate was 13 MHz. The pulses were
368 split into pump and probe pulses by a pellicle beam splitter.
369 The polarizations of pump and probe beams were controlled
370 independently. Frequency-tunable quarter-wave plates (from
371 Alphalas GmbH) were used for the generation of circularly
372 polarized radiation. Both the pump and probe beam were
373 focused on the sample in the magnet cryostat by an off-axis
374 parabolic mirror (effective focal length: 178 mm). The spot
375 size on the sample was ~ 0.5 mm (FWHM). The pump fluence
376 was $\sim 0.5 \mu\text{J}\cdot\text{cm}^{-2}$, the fluence of the probe beam was about
377 10% of the pump fluence. The time delay between pump and
378 probe pulses was varied using a mechanical delay stage.

379 *Emission measurements.* To measure radiation emitted
380 due to inter-LL recombination of electrons, the radiation was
381 guided, using a copper light pipe, to a InSb photoconductive
382 spatially separated from the coil inside which the sample was
383 placed. The used detector has narrow-band (~ 60 GHz) and
384 field-tunable response due to cyclotron resonance absorption.
385 This tunability is ensured by a specially dedicated supercon-
386 ducting coil and allows us to analyze the radiation in the
387 spectral range of 0.4 - 2.5 THz. All emission experiments oper-
388 ate in a pulsed mode (using current pulses for pumping), with

389 the pulse duration of 7 ms and with the peak-to-peak value
390 of electric field up to 12 V/cm. The duty circle was tuned in
391 such a way that the average power consumption of emitter did
392 not exceed 13 mW (to avoid sample heating). The signal on
393 the InSb detector was collected using a conventional lock-in
394 technique.

395 Additional information

396 The authors declare no competing financial interests. Cor-
397 respondence and requests for materials should be addressed
398 to M.O.

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409 Author contributions

410 The experiment was proposed by M.O. and M.P. The sam-
411 ple was grown by N.N.M. and S.A.D. Time-resolved and cw
412 magneto-optical experiments were carried out by M.O., M.M.,
413 S.W., C.F. and M.H. The cyclotron emission experiments
414 were performed by D.B., C.C., F.T. and W.K. All coau-
415 thors discussed the experimental data and interpretation of
416 results. M.O. and M.P. wrote the manuscript, all coauthors
417 commented on it.

418 Data and code availability

419 The data that support the findings of this study as well as
420 the code for modelling of cyclotron mode energies are available
421 from the corresponding author on reasonable request.

422 * milan.orlita@lncmi.cnrs.fr

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