

**A new system for real-time data acquisition and pulse parameterization
for digital positron annihilation lifetime spectrometers with high
repetition rates**

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1 **A new system for real-time data acquisition and pulse**
2 **parameterization for digital Positron Annihilation**
3 **Lifetime Spectrometers with high repetition rates**

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16 **ABSTRACT:** We present a new system for high repetition rate and real-time pulse analysis
17 implemented at the Monoenergetic Positron Source (MePS) at the superconducting electron
18 LINAC ELBE at Helmholtz-Zentrum Dresden-Rossendorf. Dedicated digital signal processing
19 and optimized algorithms are employed allowing for high bandwidth throughput, online pulse
20 analysis and filtering. Positrons generated from radioisotopes and from bremsstrahlung pair
21 production by means of highly intense accelerator-based positron beams serve as a microstructure
22 probe allowing material characterizations with respect to chemical, mechanical, electrical, and
23 magnetic properties. Positron annihilation lifetime events with up to 13 MHz repetition rate are
24 being processed online without losses while performing signal selections for pile-up reduction,
25 online energy calibration, and – for radioisotope-based measurements – identification of start and
26 stop events.

27 **KEYWORDS:** Data acquisition concepts; Digital signal processing (DSP); Online farms and online
28 filtering; Data processing methods; Data reduction methods; Detection of defects;

29

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47

48 1. Introduction

49 1.1. Positron Annihilation Spectroscopy

50 Positron annihilation spectroscopy (PAS) is a highly sensitive method for characterizing atomic
51 imperfections, such as lattice defects or dislocations [1]. Furthermore, with the help of the hydrogen-
52 like bound state of positron and electron, called positronium, PAS allows determining the sizes of open
53 and closed micro- and mesopores (0.3 - 100 nm) via the positron lifetime decay [2]. The fate of a positron
54 embodies generation, implantation into matter, thermalization, diffusion, formation of positronium (if
55 possible) and finally annihilation. Positrons are commonly generated either by β^+ decays of radioactive
56 isotopes such as ^{22}Na or by pair production by energetic x-rays from electron bremsstrahlung or γ -rays
57 from neutron-capture reactions in fission reactors. Following their generation, positrons are implanted
58 into the material under study with various energies, where they approach thermal equilibrium with the
59 sample body through inelastic collisions, various excitation mechanisms or scattering by phonons [3].
60 Subsequently, positrons diffuse inside the crystal lattice in a 3-dimensional random-walk process until
61 annihilating with an electron by emitting two characteristic 511 keV photons [4] or they form
62 positronium with an electron from the thermalization track [5], and finally annihilating through two or
63 three gamma quanta depending on the relative spin orientation [6]. Detection of these released gamma
64 quanta forms the basis of positron experiments and reflects material characteristics due to their angular,
65 energy or annihilation lifetime distribution.

66 **1.2. Beam-based PALS**

67 The investigation of defect concentration, defect type, pore size or pore size distribution are main topics
68 of positron annihilation lifetime spectroscopy (PALS), where the time difference of generation and
69 annihilation provides information about the electron density at the annihilation site or the free volume.
70 Particularly, investigations of thin films or multi-layered systems become increasingly relevant for
71 fundamental science and industrial applications. Dedicated instruments that are capable of investigating
72 not only the surface or a small section of the sample at atomic levels are in high demand. Mono-energetic
73 positron beams with energies from a few dozens of eV up to 20 keV allow investigating depth profiles
74 of defects or pores from the surface to depths of sub-micrometer.

75 Intense positron beams, which yield quite high intensities, are found at nuclear research reactors or at
76 electron accelerators. See for example: NEPOMUC at the research reactor in Munich (Germany) [7],
77 [8], POSH at the research reactor Delft (the Netherlands) [9], TIPS research reactor in Austin (Texas)
78 [10], PULSTAR in Raleigh (North Carolina) [11] or KUR in Kyoto (Japan) [12] as well as accelerator
79 based beam lines like: the positron beam at Lawrence Livermore National Laboratory (California) [13],
80 the micro-beam at National Institute of Advanced Industrial Science and Technology (AIST) in Tsukuba
81 (Japan) [14], the positron beam facility at the High Energy Accelerator Research Organization (KEK)
82 in Tsukuba (Japan) [15] and, last but not least, the monoenergetic positron source (MePS) at Helmholtz-
83 Zentrum Dresden-Rossendorf (Germany) [16].

84 **2. Digital data acquisition and online pulse analysis**

85 This section presents the progress and advantages of a fully digital signal processing on the positron
86 beam-based MePS system. Although a large part of the problems could be met by choosing C++ as a
87 high-level programming language and Qt as a platform for graphical user interfaces, the performance of
88 the system was strongly related to the algorithm. We plan to make the next version of the source code
89 available in a mature version via an open source platform. This has not yet been done for the current
90 version. In the following, we show how technical and software adaptations are used to speed up the
91 processing and improve the quality of the positron lifetime spectra.

92 **2.1. General information on digitization**

93 Digitization in the context of PALS refers to the transformation of a continuous signal, e.g., anode
94 voltage of a photo-multiplier tube coupled to a scintillating crystal, into time-discrete values by
95 employing high-speed analogue-to-digital convertors.

96 The development of digital systems for recording positron annihilation lifetime spectra started in early
97 2000er. Groups in Japan [17], [18] and Finland [19], [20] recorded the first fully-digital lifetime spectra
98 based on fast digital oscilloscopes. In the following years, many other laboratories [21]–[26] performed
99 a transition from analogue to digital technology. The advantages of digital data acquisition became
100 increasingly visible. The improved quality and time resolution of the lifetime spectra [27], the simple

101 installation of a station, the free choice of time windows or channel dispersions, the use of filter
102 algorithms [28], the increased long-term and temperature stability [29], as well as the high functionality
103 and flexibility are still reasons for more and more groups to upgrade their systems. In digital systems,
104 especially once digitized pulse becomes immune to distortion, electronic noise or other operational
105 fluctuations [30]. In addition, the raw data of the detector can be archived for offline analysis, giving
106 the user better control over analysis parameters [30]. Thus, systematic studies based on list mode data
107 can help to evaluate parameters without interference from repeated measurements.

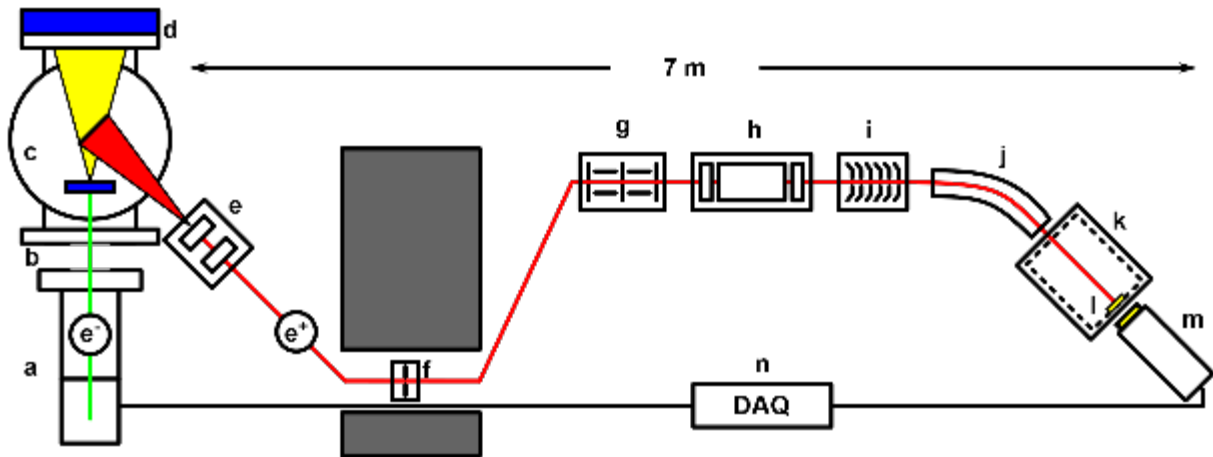
108 However, in most cases the conversion from analogue to digital data processing took place at source-
109 based and not beam-based systems. The reason for the delayed implementation in beam-based systems
110 is primarily given by the increasingly large amount of data traffic due to the higher event rates at reactor-
111 or accelerator-driven systems. High-resolution and high-speed digitizers generate larger data packages,
112 which have to be transmitted to the analysis computer and have to be handled fast enough in order to
113 perform an online analysis or pulse parameterization. This increasingly limits the maximum bandwidth
114 or counting rate of a system, and it demands higher data processing rates than analogue components.

115 **2.2. Introduction into MePS**

116 In order to illustrate the goal of digitization, some key information about MePS, the Mono-energetic
117 Positron Source, are described in the following. The MePS system results from a joint project of Martin
118 Luther University Halle-Wittenberg and Helmholtz Zentrum Dresden-Rossendorf and is one of the user
119 beamlines at the large-scale research facility ELBE Center for High-Power Radiation Sources. A
120 schematic depiction is shown in Figure 1.

121 Positron generation at MePS based on pair production caused by a pulsed, superconducting electron
122 accelerator called ELBE [31], Figure 1 (a). Bunches of electrons with an energy of about 35 MeV
123 impinge through a beryllium window Figure 1 (b) onto a water-cooled tungsten converter creating
124 bremsstrahlung, Figure 1 (c). The high-intensity bremsstrahlung in turn generates electron-positron pairs
125 in a tungsten foil located behind the converter, enabling positrons to emerge from the tungsten surface
126 with a defined energy of 3 eV [32] in a process called moderation [33]. Subsequently, the moderated
127 positrons are accelerated by a DC electric field to a transport energy of 2 keV and focused into a
128 magnetic guiding field via an electrostatic lens Figure 1 (e) and transported towards the sample chamber,
129 Figure 1 (k).

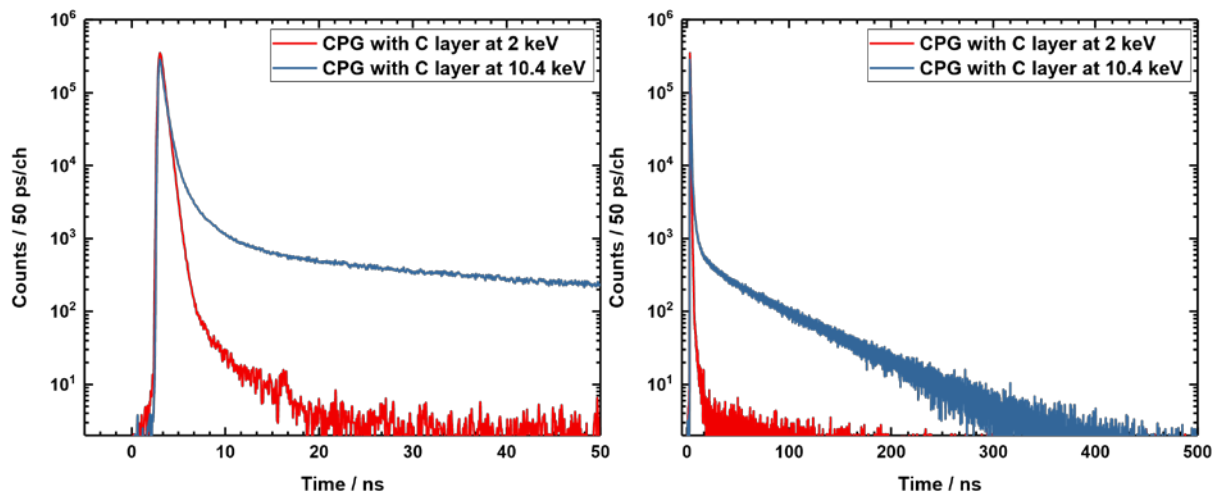
130 Consequently, a pulsed electron beam generates a pulsed positron beam. While the electron beam has
131 about 10 ps pulse width, the energy dispersion of the positron beam leads to a temporal broadening
132 which is compensated by a chopper and buncher section close to the sample chamber but prior to post-
133 acceleration, Figure 1 (g), (h) and (i).



134

135 *Figure 1: Schematic depiction of the MePS system Systems: (a) electron source ELBE, (b) beryllium window, (c)*
 136 *converter and moderator, (d) aluminum block, (e) extraction lens, (f) aperture wagon, (g) chopper, (h) buncher,*
 137 *(i) accelerator, (j) bent tube, (k) sample chamber with Faraday cage, (l) sample, (m) scintillation detector, (n)*
 138 *digital data acquisition.*

139 The operating frequency of ELBE and thus of MePS can be freely selected with a divider of 2^n of 13
 140 MHz ($n = 0, \dots, 8$). Usually, a repetition rate of 1.625 MHz ($n = 3$) is selected for positron annihilation
 141 lifetime experiments in order to avoid pulse overlap for longer annihilation lifetimes. Therefore, it is
 142 possible to measure relatively short positron lifetimes e.g., carbon with 393 ps, and relatively long
 143 positron lifetimes e.g., micro or mesoporous films with 5 to 100 ns, within the same setup at different
 144 implantations energies in 60 s per energy [34].



145

146 *Figure 2: Positron annihilation lifetime spectrum for controlled porous glass (CPG) with 280 nm carbon layer*
 147 *on top, measured at 2 keV (red) and 10.4 keV (blue) positron implantation energy at MePS.*

148 The RF signal serves as the time-start for determining the positron lifetime. The time stop is defined by
 149 detecting the annihilation radiation, which is registered by a CeBr_3 scintillator (2.54 cm thickness; 5.08
 150 cm diameter) coupled to a *Hamamatsu R13089* photomultiplier tube (PMT) located behind the sample
 151 chamber. Due to the large detector efficiency and the high number of impinging positrons, count rates

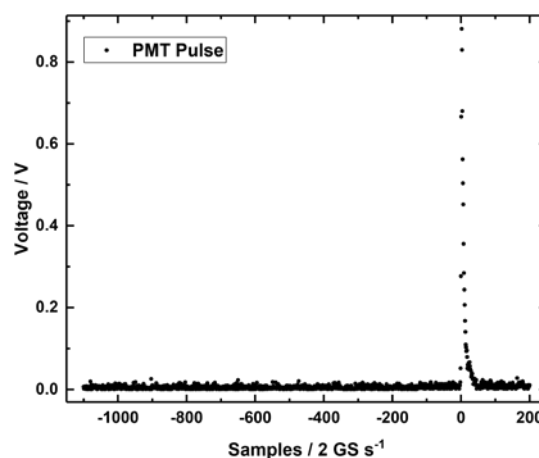
152 of about 170 kcps at 1.62 MHz are reached. The time difference between RF timing signal and
153 annihilation measures the positron lifetime shifted by a constant transport time. The main challenge of
154 digitization was to implement an online data processing that is able to detect the pulses of the PMT,
155 distinguish signal from background, assign them to the appropriate RF timing signal, calculate the time
156 difference and display the time difference histogram i.e., positron annihilation lifetime spectrum.

157 **2.3. High performance PALS**

158 To address these challenges, we use three levels of digitization. The first level is a digitizer with FPGA-
159 based trigger logic that allows channels to be triggered individually while it uses only a small time region
160 around the pulse to be recorded and transmitted to the analysis computer. The second level is a
161 sophisticated processing algorithm that minimizes the calculation time of the pulse maximum and
162 timestamp without compromising precision. The third and final level is a parallelized calculation that
163 exploits the full potential of the processor by implementing a waterfall-like data processing on multiple
164 cores.

165 **2.3.1. 1st Level: Data collection**

166 At the beginning of the electronic data processing, an analogue-to-digital converter converts the
167 continuous-time and continuous-amplitude signal from the detector – here a photomultiplier tube
168 connected to a CeBr3 scintillator – to a discrete-time and discrete-amplitude digital signal with a given
169 bit depth. In conventional digital systems, a fixed time window is recorded with respect to a trigger and
170 transmitted to the analysis computer. Consequently, large amounts of data are being transferred for
171 further analysis when large time windows have to be covered, as it is the case for long annihilation
172 lifetimes. As an example, a digitizer with 2 GS/s sample rate transmits 1300 data points per event per
173 channel for a time window of 650 ns, see Figure 3. Given a common PMT signal spanning of about 20
174 ns implies that 97% of the recorded data points contain no information about the signal. Furthermore, it
175 is up to the quality of the trigger, how many signals or whether a signal at all is recorded.



176

177 *Figure 3: Inverted PMT signal for 2 GS/s sample rate and a 650 ns long time window.*

178 Taking the sample rate of 2 GS/s, a digitizer resolution between 9 and 16 Bit, and two channels (time
179 reference and detector signal) results in a sustained data transfer rate of about 7.5 GB/s, which can barely
180 be handled in an economically feasible way.

181 In order to reduce the data traffic, we employed the *Teledyne SPDevices ADQ14-DC-2X-MTCA* digitizer
182 with 2 GS/s, 14 bit and pulse detection performed on an integrated FPGA. The firmware enables the
183 transmitted points to be reduced to a predefined range around the pulse (20 ns, 40 points), see Figure 4
184 left. In addition, the input channels of the digitizer running independently with each detected pulse
185 getting an individual rough timestamp of the internal FPGA clock. Consequently, only essential data for
186 further data processing are transmitted. The 1st level data collection decreases the data traffic by a factor
187 of 20.

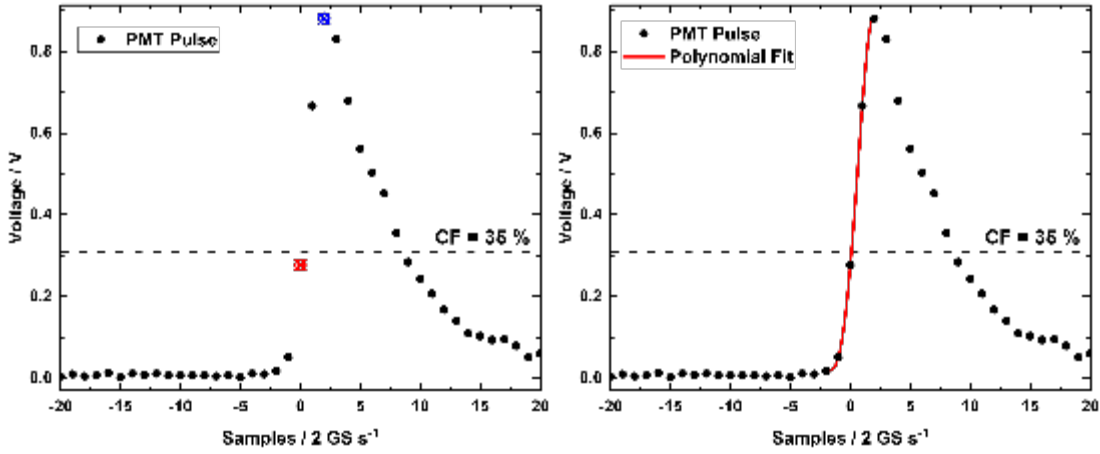
188 **2.3.2. 2nd Level: Data processing**

189 The information that needs to be determined from the tailored pulse are the amplitude (proportional to
190 the energy deposition in the detector) and the time-of-arrival. The pulse amplitude selection allows
191 identifying start and stop events in conventional radioactive source-based positron annihilation lifetime
192 experiments and filtering against background or scattered events. Furthermore, the amplitude serves a
193 constant fraction timing (CFT) algorithm for accurate determination of the time-of-arrival [35]. Hui et
194 al. [24] showed the effect of different interpolation or fitting algorithms in dependence of the digitizer's
195 sampling rate and the selected CFT level on the timing resolution of a PALS system. However, the
196 disadvantages of those individual approaches are: complex numerical functions and a large temporal
197 width of the pulse in order to adjust the shape or interpret the region between digitization points.

198 With the aim of developing a powerful and functional algorithm, we abstained from using a full peak
199 interpolation, because tests showed that the improvements in timing resolution with and without a full
200 peak interpolation were negligible. Instead, we used only the pulse maximum and a simplified algorithm
201 for the rising edge. A fourth order polynomial fit in five points was chosen with the central point of the
202 polynomial set next to the calculated CFT level, see Figure 4 right.

$$y[x] = a_0 + a_1x + a_2x^2 + a_3x^3 + a_4x^4 \quad (1)$$

203



204

205 *Figure 4: Inverted PMT timing signal for 2 GS/s and 20 ns with pulse maximum (blue dot), central point closest*
 206 *to CF level (red dot), 4th order polynomial fit (red line).*

207 This approach has the advantage, that the required coefficients could be simplified by numerically
 208 shifting the time zero point to the CFT intersection point.

209 Solving the Vandermonde matrix for (1) leads to:

$$\begin{pmatrix} a_0 \\ a_1 \\ a_2 \\ a_3 \\ a_4 \end{pmatrix} = \frac{1}{24t_{SI}^4} \begin{pmatrix} 24t_{SI}^4 y_3 \\ 2t_{SI}^3 (y_1 - 8y_2 + 8y_4 - y_5) \\ -t_{SI}^2 (y_1 - 16y_2 + 30y_3 - 16y_4 + y_5) \\ 2t_{SI} (-y_1 + 2y_2 - 2y_4 + y_5) \\ y_1 - 4y_2 + 6y_3 - 4y_4 + y_5 \end{pmatrix} \quad (2)$$

210 with t_{SI} being the sample period. In order to reduce the problem to a root-finding we shift the coordinate
 211 system towards the CFT level.

$$\mathbf{a}_0^* = \mathbf{a}_0 - y_{CFD} \quad (3)$$

212 To avoid numerically demanding root functions or case selections associated with the analytical solution
 213 of a fourth degree polynomial, which also requiring large resources of the processor, we implemented
 214 the iterative Householder method [36] for root-finding of scalar real functions.

$$\mathbf{x}_{n+1} = \mathbf{x}_n + \alpha \frac{\frac{\partial^{\alpha-1}}{\partial \mathbf{x}^{\alpha-1}} \left(\frac{1}{f[\mathbf{x}_n]} \right)}{\frac{\partial^\alpha}{\partial \mathbf{x}^\alpha} \left(\frac{1}{f[\mathbf{x}_n]} \right)} \quad (4)$$

215 Here, α is the degree of the Householder method (e. g. $\alpha = 1$ corresponds to the Newton method, $\alpha = 2$
 216 to the Halley method). With increasing order of the applied polynomial, the convergence rate increases
 217 and the accuracy of the numerical solution can already be sufficient for the first iterations. However,
 218 this advantage is accompanied by an increasing complexity of the iteration function. Fortunately, this is
 219 a negligible problem for our application. On one hand, the solution of the first iteration of the

220 Householder method has sufficient precision for the application in positron annihilation lifetime
 221 spectroscopy, and on the other hand, by shifting the CF point to a virtual time zero point, the first
 222 iteration could be carried out with the initial value x equals 0. Using (4) with $x_0 = 0$, the polynomial (1),
 223 the coefficients from (2) and (3) we obtain a simplified form:

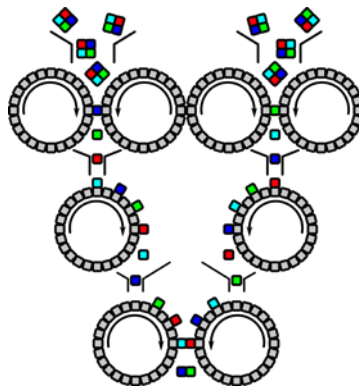
$$x_1 = \frac{a_0^*(2 a_0^* a_1 a_2 - a_0^{*2} a_3 - a_1^3)}{a_1^4 - 3 a_0^* a_1^2 a_2 + 2 a_0^{*2} a_1 a_3 + a_0^{*2} (a_2^2 - a_0^* a_4)} \quad (5)$$

224 By that, the numerical root determination reduces to a single term, which only requires simple arithmetic
 225 operations. Thus, this approach avoids complex functions such as roots or higher exponents as well as
 226 case selections, since the choice of the start value and the form of the polynomial almost guarantees
 227 convergence at the searched intersection point. Combining the fine time stamp from the CF timing and
 228 the rough time stamp from the FPGA clock, one obtains a global time stamp (6) for each pulse, which
 229 can be combined or compared with other pulses, for example to determine the positron annihilation
 230 lifetime.

$$t = t_{\text{FPGA}} + x_1 \quad (6)$$

231 2.3.3. 3rd Level: Data allocation and parallelization

232 Once the detector signals being processed and a time stamp being calculated, the independent time
 233 stamps of RF and detector have to be correlated in time. To further accelerate the processing and to use
 234 the full potential of the processor, three major steps were processed in parallel: data collection,
 235 processing and allocation. Since the individual steps depend on each other, a waterfall-like logic was
 236 implemented. We employed a buffered data collection in order to efficiently parallelize the data
 237 processing.

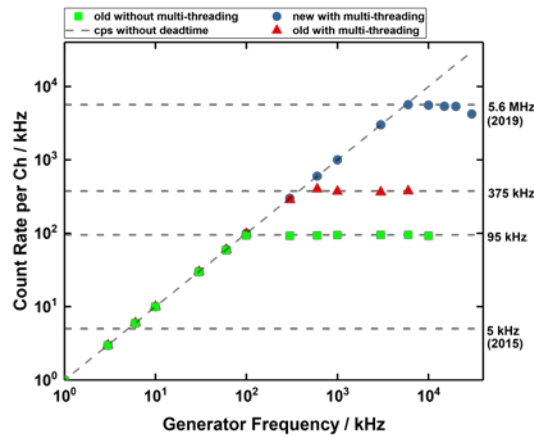


238
 239 *Figure 5: Schematic depiction of the multithreading process for a digitizer with four channels.*

241 **2.4. Performance tests**

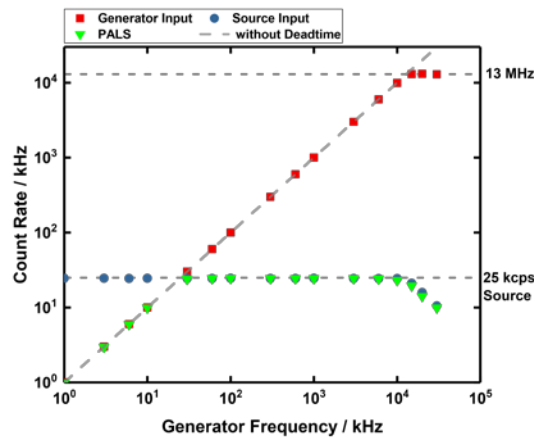
242 In order to quantify the efficiency of the individual optimisation phases, we employed a signal generator
243 with variable frequency as a reference and split it into two input channels. Figure 6 shows the generator
244 frequency and the corresponding processing rate of the respective algorithm. It can be seen that the
245 change from a standard digitizer (*Acqiris DC282*) to an FPGA-based peak detection system (*Teledyne*
246 *SPDevices ADQ14-DC-2X*) with the same pulse analysis algorithm (cubic spline interpolation), allowed
247 going from 5 kcps to almost 100 kcps.

248 Further optimisation was obtained through implementation of the aforementioned waterfall-like
249 parallelisation of data processing. Multithreading of the software increased the performance up to
250 375 kcps. Additionally implementing the Householder root-finding drives the processing rate up to a
251 maximum of 5.6 Mcps per channel.



252
253 *Figure 6: Pulse processing rate per channel as function of the generator frequency: with pulse detection but*
254 *without multi-threading (green), with pulse detection and with multi-threading (red), with pulse detection, with*
255 *multi-threading and Householder approach (blue)*

256 In order to test the optimised algorithm and hardware in case of an uncorrelated time signal distribution,
257 one input was coupled to the pulse generator and the second one to a stochastic signal of a radioactive
258 source, see Figure 7. It can be seen that the time differences of the test source can be processed free from
259 dead-time-related dips or a memory overflow; the source count rate of 25 kcps dominates the system
260 rate up to a generator frequency of 13 MHz. Consequently, the hardware and software can be used to
261 determine the occurrence of rare events coupled with respect to a high frequency time reference.
262 Consquently, this System can also be adapted for other systems or experiments as well as for positron
263 annihilation lifetime spectroscopy. An implementation for prompt gamma-ray timing in proton-beam
264 based tumor therapy for in-vivo dose verifications [37] is under way.



265

266 *Figure 7: Pulse processing rate for a stochastic signal with 25 kcps and a variable frequency generator.*

267 *Processing rate source input (PMT) (blue), processing rate generator input (red) and combined processing rate*

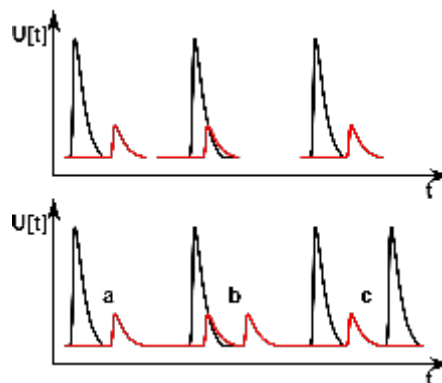
268 *compared to a PALS experiment (green)*

269 2.5. Super-Singles Filter

270 The occurrence of detected or undetected pile-up events, namely the arrival of more than one time signal

271 in a given time window (see Figure 8) has to be prevented in order to avoid a systematic bias. The *super-*

272 *singles filter* suppresses events in close temporal vicinity.



273

274 *Figure 8: Schematic depiction of a pileup event with a virtual (top) and an actual image (bottom): a) Unique*

275 *start-stop allocation, b) Stop-stop error, c) Start-start error.*

276 In contrast to conventional trigger logics, this approach not only checks the recorded time window, but

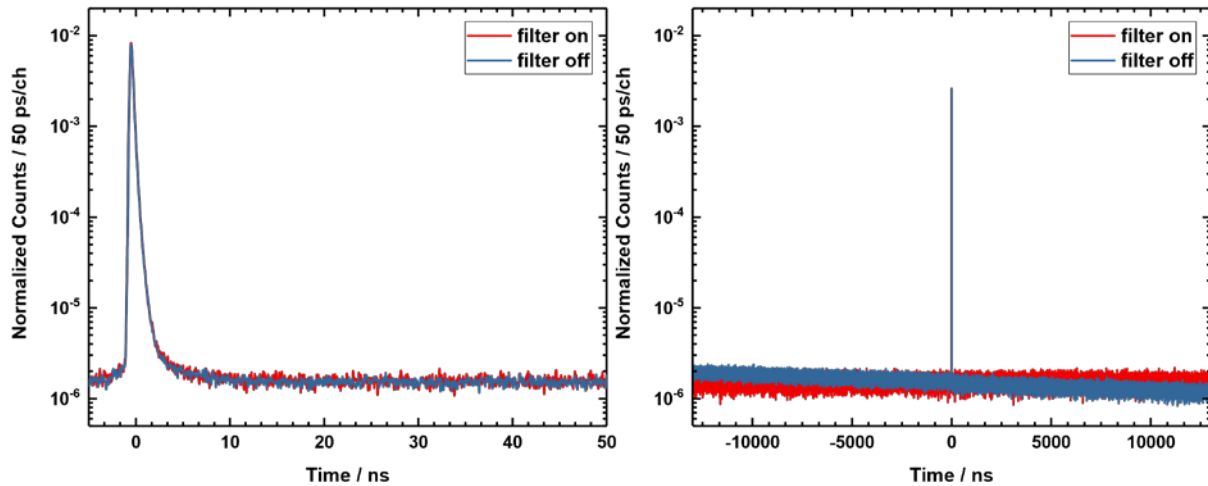
277 also the chronologically preceding and following pulses in the same channel. Thus, excluding the

278 occurrence of other pulses besides the searched start or stop signal, which would falsify an unambiguous

279 assignment and the calculation of the time difference. Tests on source-based PALS systems with large

280 time window show the result of a pile-up contaminated lifetime measurement and in particular the

281 uniformly distributed random background when the super-singles filter is applied, see Figure 9 right.



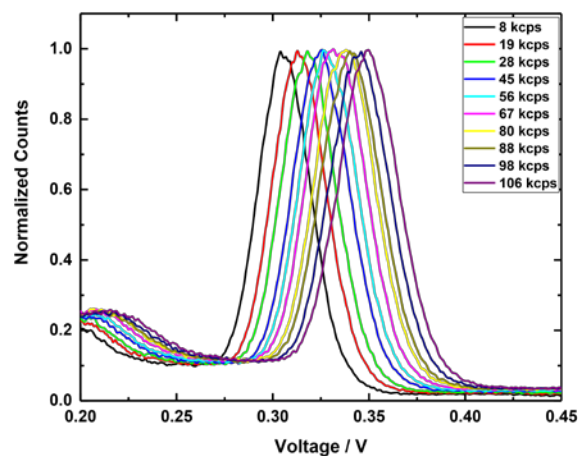
282

283 *Figure 9: Normalized positron lifetime spectrum for a silicon reference material at a source-based setup with*
 284 *and without super-singles filter. Small time window (left) and large time window (right)*

285 Inherently, the systematic bias appears negligible at narrow time intervals but the effect on the positron
 286 annihilation lifetime calculation cannot be neglected, since an adequate background calculation, either
 287 prior or after the main component of the spectrum, is essential. Consequently, taking this effect into
 288 account could prevent ghost components, incorrect lifetimes or false intensities.

289 2.6. Automated energy calibration

290 An underestimated source of error in PALS measurements are wrong or fluctuating energy windows.
 291 Here, the range in which a signal is grouped as a start or stop should be adequately adjusted. Changes
 292 of the detector gain within a measurement (unstable bias supply voltages, changing count rates,
 293 temperature variations) lead to improperly selected energy windows and possible systematic errors.
 294 Especially, changing detector count rates cause shifts in the signal amplitudes as shown in Figure 10.



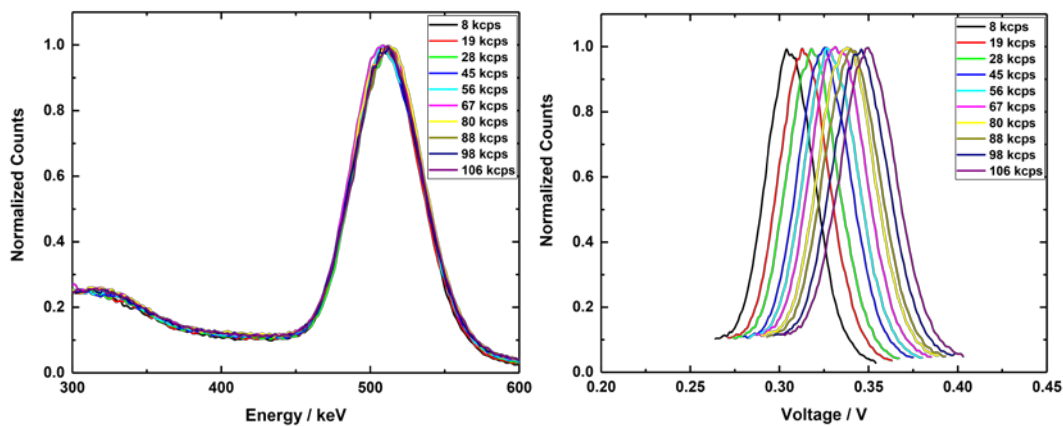
295

296 *Figure 10: Pulse height spectra of the 511 keV annihilation radiation for different count rates.*

297 Such linearity errors predominantly result from changing voltages when the electrode voltage is supplied
 298 from a resistive divider. The current given by the electrons at the last dynodes is no more negligible with

309 respect to the current through the voltage divider. The disturbance of the voltage distribution throughout
300 the divider results in an increased gain with increasing count rate [38]. As a consequence, problems arise
301 when the mostly voltage-defined windows of the pulse assignment at PALS experiments are set to fixed
302 values, like analogue setups, while the count rate varies due to changes in the solid angle, beam steering
303 or sample charging. In worst case, this leads to an almost complete shift of the annihilation peak out of
304 the defined limits.

305 To realign the focus, a dynamic window determination was implemented. This no longer sets the
306 window limits based on the voltage, instead performs a simplified energy calibration prior to each new
307 measurement, allowing defined energy windows, see Figure 11 right.



308
309 *Figure 11: Shifted Pulse height spectrum into annihilation peak (left) dynamic window adjustment (right).*

310 **3. Summary**

311 We demonstrate, how an efficient online data reduction, a numerically effective algorithm for pulse
312 parameterization and utilization of multi-core parallelization led to a significantly improved data
313 processing for positron annihilation lifetime spectroscopy. The approach constitutes the MePS setup as
314 one of the first fully digital measurement systems considering beam-based positron annihilation lifetime
315 spectroscopy facilities. The advantages include variable timing windows, arbitrarily selectable channel
316 dispersions, acquisition times of less than 60 seconds per spectrum with 10^7 events each, absence of
317 systematic bias due to pile-up events or energy window fluctuations, and, in addition, processing rates
318 of up to 5.6 Mcps per channel for dynamic defect evolution studies. The presented system can be applied
319 in future readily in neutron time -of-flight measurements [39], time-of-flight mass spectrometry or
320 prompt gamma-ray timing in proton therapy [37] and been instrumental in the past in a variety of
321 measurements ranging from materials for hard coatings [40], new vistas of the influence of atomic
322 defects in voltage-driven magneto-ionics [41]–[43], material damages in heavily irradiated reactor
323 materials [44] to superconductors for particle accelerators [45] and defect-related magnetism in FeAl
324 alloys [46].

325 **4. References**

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