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Methodology of positron lifetime spectroscopy: Present status and perspectives

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Abstract

Achievements in instrumentation for positron lifetime measurements are summarized with emphasis on digital spectrometric systems. The main part of the data presented are based on the long-term exploitation of a conventional positron lifetime spectrometer developed at Charles University in early 90s, on bench-mark testing measurements with this spectrometer working in conjunction with a pair of 8-bit ultra-fast digitizers and on analogous measurements with a new, recently assembled digital positron lifetime spectrometer. Perspectives of further improvements of positron lifetime instrumentation are discussed.

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The positron lifetime (PLT) spectroscopy is a well-proven and in many cases irreplaceable technique for studying the microstructure of a broad class of condensed-matter systems. However, in spite of numerous efforts to improve quality of PLT measurements, these still suffer from a limited timing resolution and a lack of control over various distortions.

In 1953, Bell and Graham, while studying positronium, observed that their time spectra displayed components with positron lifetime of about 100 ps [1]. It is surprising that after five decades the threshold for observation of short positron lifetime components has been reduced only by a factor of three. Indeed, only a few of the existing PLT spectrometers are able to resolve components in the PLT spectra with a lifetime below 30 ps. This demonstrates that the development of the PLT instrumentation stagnates in spite of tremendous progress in photonics and electronics.

During the last decades several important innovations in PLT technique have appeared: (i) the fast photomultipliers (PMTs) Philips XP2020/Q were developed, (ii) BaF_2 scintillators were successfully applied to γ-ray timing spectroscopy [2] and (iii) the extraction of the fast PMT signal from a dynode has been introduced [3]. These steps led to a significant enhancement of the performance of setups for conventional PLT measurements with ^{22}Na positron source. A few years ago, in the pioneering works of Saito et al. [4] and Rytsölä et al. [5], a successful use of ultra-fast digitizers in PLT spectroscopy had been reported. In addition, for the needs of precise timing a *true* constant fraction (CF) method has been introduced [4] as a replacement of the *analog* CF method used in conventional CF discriminators. In testing measurements with ^{22}Na source an impressive timing resolution, represented by FWHM of 140 ps has been achieved [4]. Nevertheless, it was still by far not clear whether the introduction of the digital PLT spectroscopy represents, except for its conceptual simplicity, a real step forward. In fact, the above-mentioned resolution had been achieved before with a conventional *analog* fast-slow setup, as seen in [6], so that the key question whether the use of the ultra-fast digitizers enhances the precision of PLT measurements remained unanswered.

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To clarify this point we have undertaken testing measurements [7] with the above-mentioned analog spectrometer [6] and with its re-configuration into the digital mode of operation. Identical γ detectors, assembled in 1993 from BaF_2 scintillators and Philips PMTs XP2020/Q, were used. Except for this re-configuration all remaining conditions of measurements were preserved. The analog spectrometer after more than 11 years of continuous exploitation displayed in PLT measurements with ^{22}Na source a timing resolution characterized by FWHM = 173.0 ps at a coincidence rate of 125 s^{-1}. However after its re-configuration into the digital mode it displayed a resolution of 136.1 ps or better at a slightly lower coincidence rate of 115 s^{-1}. Moreover, the digitizers made it possible to reduce the number of the distorted detector signals by a factor of 10 [7]. It clearly indicates that the advent of ultra-fast digitizers represents a new milestone in the PLT technique.

Inspired by the previous results [4,7], a new digital PLT spectrometer has been assembled at Charles University. It includes a pair of Acqiris 8-bit 4 GS/s digitizers, sharing a common clock signals. The detectors are formed by fast Hamamatsu PMTs H3378-50 and truncated-cone BaF_2 crystals with diameters of 18/36 mm and thickness of 12 mm. While scanning the data from the new setup, we used the true CF method applied to the *integrated* anode detector signals referred hereafter to as the "iCF method". The way of determining the iCF points is elucidated in Fig. 1, where *normalized* integrals from 2500 detector signals are plotted. Values of reference time t_{iCF} are determined using a parabolic interpolation with a small additional correction for a non-parabolicity of real detector signals. As seen from the insert in Fig. 1, the interpolation used is good enough for reaching almost an ideal determination of the iCF point adjusted at a level of 7%. In measurements with ^{22}Na source the standard error of its determination is estimated to be less than 2 ps, see Fig. 1. In off-line scanning the list-mode data we use a new digital filter to exclude distorted signals. This is achieved by setting an allowed region for the integrated signals. This region is limited by suitably pre-selected majorizing and minorizing functions, as shown in Fig. 1. For each detector a separate digital filter is needed. Gamma-ray energy selection for distinguishing the 1274 keV start and 511 keV stop detector signals is also provided digitally.

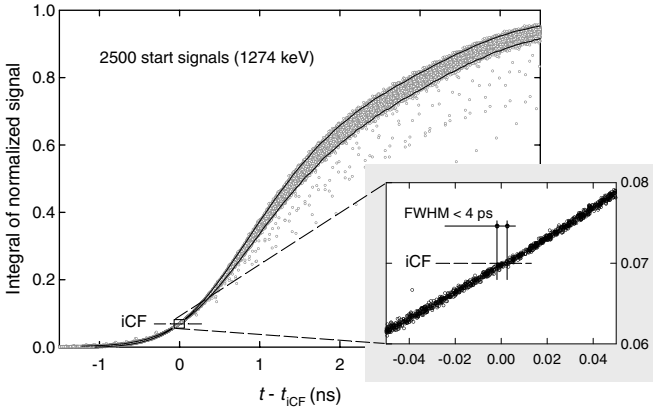

Fig. 1. Integrated normalized digitized detector signals plotted as a function of t–t_{iCF}, where t_{iCF} represents the time mark obtained by the iCF method with the aid of a *corrected* parabolic interpolation. The signals plotted satisfy the condition imposed by the energy window adjusted at the 1274 keV γ line. Solid lines represent majorizing and minorizing functions.

The data from testing measurements with an αFe sample, plotted in Fig. 2, demonstrate how the introduced digital filters work. Spectrum (a) in both panels has been obtained after the filtering, while spectrum (b) is formed exclusively by filtered-out detector signals. These spectra differ significantly from each other. The number of filtered-out events represents about 20% of the area under the unfiltered PLT spectrum. However, it turns out that the dominating part of the filtered-out events is not damaged at all by the pileup or other distortions. In view of randomness of perturbations of the detector signals the PLT spectrum constructed exclusively from the signals that

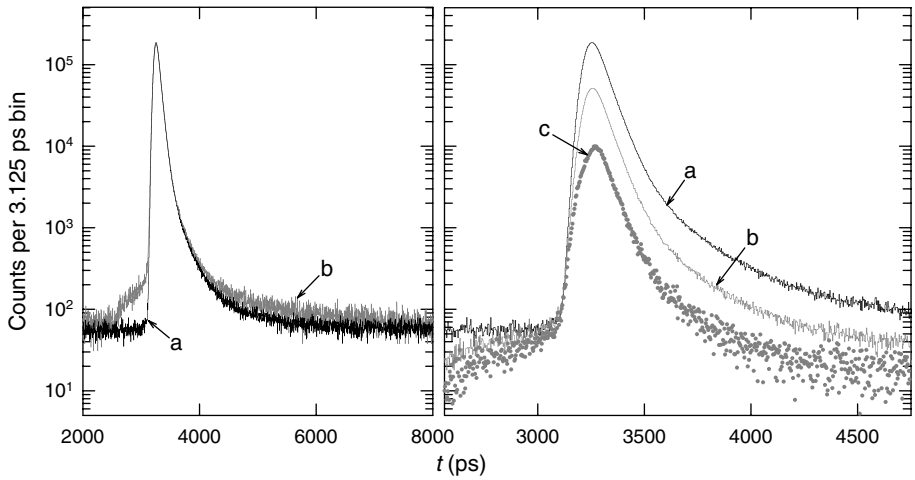

Fig. 2. (a) A PLT spectrum obtained from the measurement with αFe sample with active digital filters; (b) a spectrum reconstructed exclusively from the filtered-out events – in the left panel it is re-scaled for better comparison with the filtered spectrum; (c) a spectrum obtained from the filtered-out signals after corrections for a 78% admixture of undamaged signals.

are really *distorted* should have a shape of a strongly smoothed *true* PLT spectrum, not displaying any sharp structures. Such a spectrum can be obtained from the spectrum (b) in the right-hand side panel of Fig. 2 by subtracting an appropriate fraction of the *filtered* spectrum (a). We found that this can be achieved under the condition that this fraction contributes to the filtered-out events not more than by 78%. In this limiting case the spectrum constructed from the distorted events is illustrated by plot (c) in Fig. 2. The influence of pile-up effects or other distortions could be in part eliminated by postulating of an appropriate, *a priori* unknown shape of timing response function with no guarantee for a success. However, the use of the digital filters solves this problem radically.

The quality of PLT data reached with the new spectrometer is illustrated in Fig. 3. It is to be added that with a very narrow γ-ray energy selection a resolution power of 129 ps has been reached in ^{22}Na measurements with αFe sample. The data from the digital spectrometer make it possible to be analyzed also in a "start-stop/stop-start regime", when two independent PLT spectra are accumulated at a time, taking advantage that both detectors are sources of signals from detection of the 511 keV, as well as 1274 keV γ-rays. As a result, a doubled count rate is achieved, in our case up to 240 s^{-1}, keeping the resolution below 145 ps.

In an attempt to understand the role of various factors affecting the timing resolution we developed an algorithm for simulating the PMT output signals induced by a fictitious *prompt* γ-cascade with energies 1274 and 511 keV. Except for collection of scintillation light from the BaF_2 crystals, this algorithm takes into account all factors responsible for formation of the detector signals as well as for digitizing and subsequent off-line scanning of these signals.

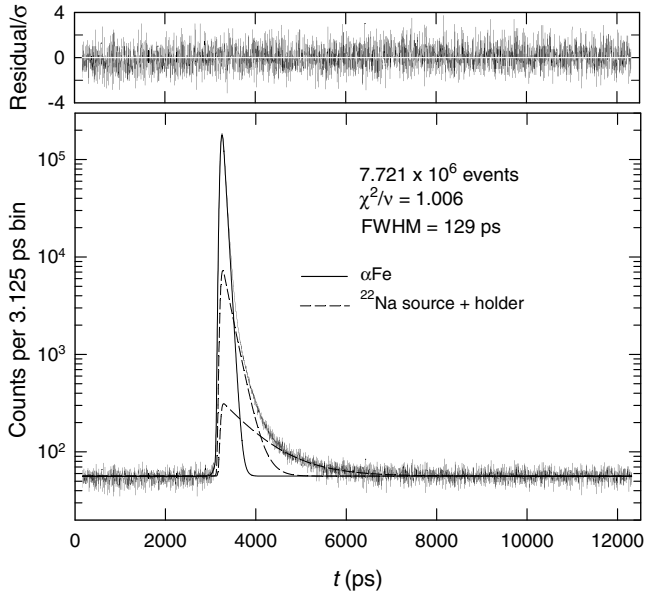

Fig. 3. A PLT spectrum from the measurement with αFe sample and its decomposition. This spectrum has been obtained in the "start-stop" scanning regime when energy windows were adjusted to include the whole 1274 and 511 keV γ lines.

When the digitizers used worked in the regime of an artificially introduced floating baseline they made it possible to determine with high precision the shape of area-normalized single-photoelectron signals. The relative r.m.s. value characterizing fluctuations of size of these signals was determined to be 32.1%. The energy resolution of both detectors at 511 keV was found to be 27.5%. Having at our disposal these data together with the value of time jitter for the H3378-50 PMTs [8], the above-outlined algorithm could be used to simulate functioning of our PLT spectrometer. There is only one free parameter in the simulations – the photocathode yield, induced by the fast part of the BaF_2 signal. To reproduce energy resolution of 27.5% at 511 keV this parameter had to be adjusted at surprisingly low value of 201 photoelectrons per 1 MeV of the deposited energy in a BaF_2 crystal. In this case, for the prompt 1274–511 keV γ-cascade our simulations yielded the timing response function with FWHM $= 142 \pm 1$ ps. This agrees unexpectedly well with what we have observed and it suggests that within the given conditions we achieved the limiting timing resolution.

The detectors used in our new PLT spectrometer are evidently not of top quality, as the number of photoelectrons triggering electron avalanches is relatively low: the above-mentioned value of 201 photoelectrons/MeV is by a factor of 1.7 lower than one would expect from quantum efficiency QE = 19% at 220 nm listed for H3378-50 PMTs [8], and from the expected UV output of 1800 photons/MeV from BaF_2 crystals [9].

The long-term experience with the most frequently used Philips PMTs, Model XP2020/Q, see [6], suggests that even with the know-how in manufacturing of PMTs from the mid 80s a much better timing resolution power of *digital* PLT spectrometers should be reached compared to the best one reported so far. Indeed, in spite of disfavor that our XP2020/Q PMTs deteriorated after the long-term use since 1993, the PLT spectrometer assembled from them and the Acqiris DC-241 digitizers displayed an excellent timing resolution, as shown in [7]. The ≈20% improvement in timing resolution we observed in switching to the digital mode and the fact that we were able to achieve a timing resolution of ≈140 ps in 1993 [6] suggest that it should be possible to achieve a timing resolution close to the magic limit of 100 ps in the digital mode if PMTs equal in quality to 1993 vintage XP2020/Q PMTs were used.

Much better timing resolution power than 140 ps can be obtained with the available BaF_2 crystals and PMTs if these are carefully selected according to the UV light yield, the QE at 190–220 nm and the time jitter. Regarding the data on PMT time jitter, a novel technique, based on the use of sonoluminescence pulsed light sources, can be employed [10]. As for the BaF_2 UV light yield, its determination is not a simple task. Unfortunately, no specifications of this parameter for individual BaF_2 crystals are provided by manufacturers.

It is known that non-stochiometric crystals $Ba_{0.9}La_{0.1}F_{2.1}$ display a fast UV scintillation component with decay time $\tau = 0.41$ ns and a yield of 1260 photons/MeV compared to respective values 0.73 ns and 1800 photons/MeV for pure BaF_2 crystals [9]. The use of the $Ba_{0.9}La_{0.1}F_{2.1}$ crystals would certainly improve the timing resolution.

In an attempt to use faster photonics devices for the needs of PLT spectroscopy we tested timing properties of a detector assembled from a Photek three-plate microchannel-plate (MCP) PMT, Model PMT-325, and a BaF_2 crystal with diameter of 25 mm and thickness of 12 mm. For this purpose we used the above-mentioned new PLT spectrometer with one of the Hamamatsu-based detectors replaced with the detector to be tested. The testing measurement with the hybrid pair of detectors yielded the ^{60}Co prompt peak with FWHM = 149 ps, while for a pair of identical detectors with H3378-50 PMTs we obtained FWHM = 105 ps. It implies that a pair of detectors using the Photek MCP PMTs would yield a ^{60}Co prompt peak with FWHM $\approx$ 183 ps. From this result it follows that the tested MCP PMT is not of use for PLT spectroscopy. A detailed inspection revealed that in spite of a relatively high photocathode QE, specifically 26% at 200 nm, the quantum *detection* efficiency (QDE) of the MCP PMT tested turned to be as low as 3–4%. In other words, only 12% of photoelectrons trigger the electron avalanches in the MCPs. This discrepancy is not yet understood. In view of the large deficiency of "active photoelectrons" the above-mentioned value FWHM $\approx$ 183 ps is, in fact, unexpectedly low. A measurement of ^{137}Cs γ-ray spectrum with NaI(Tl) scintillator coupled with a two-plate MCP PMT led to a surprisingly high resolution power of 8%, see [11], which implies that a very high QDE of the MCP PMTs can be reached. It suggests that the continuing development of MCP PMTs will bring further progress in PLT technique.

Other possibilities are offered by the use of *bare* MCP PMTs as detectors of γ radiation. The principle of the γ detection is very simple: Gamma rays hitting the MCP PMT entrance window produce fast photoelectrons and Compton electrons. Some fraction of these electrons escapes the entrance window before the electrons fully stop. A part of the ejected electrons is fully or partially absorbed by the MCPs, producing a relatively large number of secondary electrons. The secondary electrons trigger, in turn, electron avalanches in MCPs. The γ-ray detection efficiency of bare MCP PMTs is low, of order of 1%, but it can be increased by placing a thin conversion layer, made from high-Z material, in front of the first MCP.

With a pair of *bare* Photek MCP PMTs, Models PMT-325 and PMT-318, using a ^{60}Co source we undertook a testing measurement of timing properties of these photonics elements. As seen from Fig. 4, an unprecedented timing resolution has been achieved. However, in view of the extremely low detection efficiency, the bare MCP PMTs can hardly be used in conventional PLT experiments. Nevertheless, they can be, at least in principle, employed in PLT measurements on intense pulsed positron beams, especially in conditions of extremely short pulses and a large number of the produced positrons per pulse.

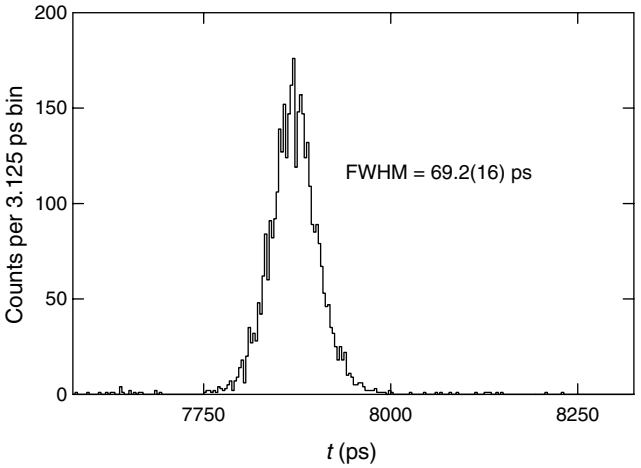

Fig. 4. Timing response function of a pair of *bare* MCP PMT-325 and PMT-318 (measured using a ^{60}Co source).

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References

[1] R.E. Bell, R.L. Graham, Phys. Rev. 90 (1953) 644.
[2] M. Laval, M. Moszyński, R. Allemand, E. Cormoreche, P. Guinet, R. Odru, J. Vacher, Nucl. Instr. and Meth. A 206 (1983) 169.
[3] B. Bengtson, M. Moszyński, Nucl. Instr. and Meth. A 204 (1982) 129.
[4] H. Saito, Y. Nagashima, T. Kurihara, T. Hyodo, Nucl. Instr. and Meth. A 487 (2002) 612.
[5] K. Rytsölä, J. Nissilä, J. Kokkonen, A. Laakso, R. Aavikko, K. Saarinen, Appl. Surf. Sci. 194 (2002) 260.
[6] F. Bečvář, J. Čížek, L. Lešták, I. Novotný, I. Procházka, F. Šebesta, Nucl. Instr. and Meth. A 443 (2000) 557.
[7] F. Bečvář, J. Čížek, I. Procházka, J. Janotová, Nucl. Instr. and Meth. A 539 (2005) 372.
[8] Photomultiplier Tubes and Assemblies, Hamamatsu Photonics, Shizuoka-ken, Japan, 1996.
[9] B.P. Sobolev, E.A. Krivandina, S.E. Drenzo, W.W. Moses, A.C. West, in: M.J. Weber (Ed.), Proceedings of the Material Research Society: Scintillator and Phosphor Materials, San Francisco, 1994, p. 277.
[10] P.V. Vorob'ev, I.N. Nesterenko, A.R. Frolov, T.V. Shaftan, Vol. 1, 1998, arXiv:physics/9811005.
[11] V.V. Anashin, P.M. Beschastnov, V.B. Golubev, L.A. Mironenko, A.A. Sanikov, S.I. Serednyakov, Nucl. Instr. and Meth. A 357 (1995) 103.