

APPLICATION OF DIGITAL SPECTRAL ANALYSIS TO DECOMPOSITION OF POSITRON LIFETIME SPECTRA

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New method of positron annihilation lifetime spectra processing is proposed. It is based on digital spectral analysis, namely, on Prony method. The regularization procedure to solve the inverse problem of finding parameters of useful signals present in the data is formulated. A new approach to determine the number of components in a spectrum is proposed. Results on simulated spectra demonstrate the high-resolution capability of the method.

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1. INTRODUCTION

Positron lifetime spectroscopy is one of the best methods for detecting open-volume defects of atomic dimensions. Positrons implanted into the solid rapidly lose energy via a variety of inelastic processes. The thermalized positrons diffuse in the lattice and finally will be annihilated by an electron. The rest energy of electron-positron pair is converted into energy as two annihilation γ -quanta with energy 0.511 MeV. Detection of any of these γ -quanta is the signal of positron death. The signal of positron birth depends on the source used but in any case the time difference between these two signals determines the positron lifetime for a single event. The lifetime is a random quantity and thus must be described by its probability distribution function.

Many open-volume defects in metals and semiconductors are negatively charged and hence form a local attractive potential for positron. Therefore, in the process of a random walk a positron can be trapped at these defects in a localized bound state. The annihilation rate is determined by the overlapping of positron and electron densities. Negatively charged or neutral open-volume defects have at the same time locally reduced electron density. That is why the lifetime of the positron trapped at these defects increases.

In other words, we expect that for open-volume defects of k different types the probability distribution function $s(t)$ of the positron lifetime t is a weighted sum of $(k + 1)$ components. It was shown [1] that under quite general assumptions this function has a form

$$s(t) = \sum_{j=1}^{k+1} I_j \exp\left\{-\frac{t}{\tau_j}\right\} \quad (1)$$

The aim of the spectrum analysis is the extraction from the experimental signal the set of parameters $\{\tau_j, I_j\}$ for $j = 1, \dots, k + 1$. This analysis allows us to separate out various atomic defect configurations and their relative

abundance.

The experimental spectrum differs from the function $s(t)$ in several aspects.

- 1) Function $s(t)$ is the input signal for detecting system. To obtain the output signal we have to convolve $s(t)$ with impulse response characteristic of the system.
- 2) Output signal is formed with the help of a multi-channel analyzer (MCA), that is, the continuous nature of the signal $s(t)$ is discretized. This transformation has several consequences.

It is a common experience that only one to four components can be resolved, whereas the number of channels is of order of 100. It means that the problem of spectrum analysis is overdetermined and its solution is usually defined in a least square sense.

Events detected in any particular channel form the flow of random events, in the simplest case the Poisson flow. Therefore the spectrum as a whole can be subdivided into useful signal and Poisson noise.

Discretization of a continuous signal by a multichannel analyzer is equivalent (see below) to the procedure of taking sampling values of this signal at some grid points. This grid depends on the signal under study and its properties should be taken into account in spectrum processing.

Various approaches have been used to analyze positron lifetime spectra but up to now the most commonly used are approaches based on the Gauss-Newton non-linear least square fitting [2]. We propose a new approach to the problem and within this approach we studied two problems.

The first concerns the determination of the number of components. The conventional solution is to start with one-component fit and add components as long as the variance of the fit decreases. The weakness of this solution is well known in spectral analysis [3]: the variance of the fit usually does not reach a minimum value

but rather monotonically decreases as the order of the model is increased. Another approach based on optimal linear filtering and the method of maximum entropy was proposed in [4] but its resolution capability is not high enough and this leads to certain difficulties in treating a spectrum with relatively close components.

Another traditional problem concerns the ill-posed nature of the inverse problem [5] under study. This means that the problem is inherently unstable.

2. SAMPLING

Let the spectrum reside on a multichannel analyzer with a time increment Δ per channel. In the experiment the number of counts versus channel number is measured. Therefore we can calculate the total numbers of counts and then for any particular channel $[a, b]$ the probability P of the positron lifetime to fall inside this channel. For the simplest case of a one-component spectrum this probability is equal to the exponential probability distribution function

$$s(t) = Ae^{\alpha t} \quad (2)$$

integrated over t from a to b ,

$$P = \int_a^b s(t) dt = \frac{A}{\alpha} (e^{\alpha b} - e^{\alpha a}). \quad (3)$$

According to the mean value theorem

$$P = \int_a^b s(t) dt = s(c)(b-a) = Ae^{\alpha c}(b-a), \quad (4)$$

where $c \in [a, b]$. From equations (3), (4) we obtain

$$c = a + \frac{1}{\alpha} \ln \frac{e^{\alpha b} - 1}{e^{\alpha a} - 1}. \quad (5)$$

The distance

$$\delta = c - a = \frac{1}{\alpha} \ln \frac{e^{\alpha b} - 1}{e^{\alpha a} - 1} \quad (6)$$

is the same for all channels. It means that the procedure which measures the probability P for the probability distribution function $s(t)$ (2) gives sampled values of this function on an equidistant grid with the same step Δ . Note that the exact position of the grid on the time axis depends on the coefficient α which is usually not known in advance.

Therefore, for the positron lifetime spectrum of the form (1) this procedure will determine the sampled values for different components on different grids. Let us suppose that in the model sampled values for the signal $s(t)$ (1) as a whole are defined on the equidistant grid with step Δ which for simplicity is taken as

$$0, \Delta, \dots, (N-1)\Delta, \quad (7)$$

where N is the number of channels in the spectrum.

It can be easily shown that for a one component signal $s(t)$ (2) two estimators $\hat{\alpha}_1, \hat{\alpha}_2$ of the damping coefficient α will be the same for two grids: (7) and $0 + \delta, \Delta + \delta, \dots, (N-1)\Delta + \delta$ accordingly. But the estimator \hat{A}_2 for the amplitude A we have to increase by a factor $e^{-\alpha\delta}$ ($\alpha < 0$) in order to consider formally the estimators

$$\hat{\alpha}_1 = \hat{\alpha}_2, \quad (8)$$

$$\hat{A}_1 = \hat{A}_2 e^{-\alpha\delta} = \hat{A}_2 \frac{e^{\alpha\Delta} - 1}{e^{\alpha\delta} - 1}, \quad (9)$$

as obtained for the sampled values of the function $s(t)$ (2) taken at the grid points (7). Similar transformations can be applied to all components in the positron lifetime spectrum (1).

3. COMPUTATIONAL PROCEDURE

Experimental positron lifetime spectrum in any channel contains not only the useful signal but also a noise. Therefore let us suppose that the spectrum is defined by the set of sampled values $x[1], x[2], \dots, x[N]$, taken on the grid (7) and try to estimate this spectrum with a general p -member model of complex exponentials

$$\hat{x}[n] = \sum_{j=1}^p A_j \exp\{(\alpha_j + i2\pi f_j)(n-1)\Delta + i\theta_j\} \quad (10)$$

Hereafter, in keeping with the tradition of the signal processing literature, we use brackets around a discrete variable.

In Eq. (10) i is the imaginary unit, $1 \leq n \leq N$, A_j, α_j, f_j and θ_j are amplitude, damping coefficient, frequency and phase constant accordingly for j -th complex exponential. Note that we do not impose any restrictions on the order of model p and values of all these parameters. As a consequence sum (10) may present both pure decreasing exponentials with zero frequencies and undamped sines with zero damping coefficients. Terms of the first type are the same as the components of the useful signal (1) whereas the latter describe the noise part of the spectrum.

In shorthand notation we can write down the function of discrete time (10) as

$$\hat{x}[n] = \sum_{j=1}^p h_j z_j^{n-1}, \quad (11)$$

where

$$h_j = A_j \exp(i\theta_j), \quad (12)$$

$$z_j = \exp\{(\alpha_j + i2\pi f_j)\Delta\}. \quad (13)$$

For N sampled values we have to minimize the sum

$$\rho = \sum_{n=1}^N |\varepsilon[n]|^2, \quad (14)$$

where

$$\varepsilon[n] = x[n] - \hat{x}[n] = x[n] - \sum_{j=1}^p h_j z_j^{n-1}, \quad (15)$$

simultaneously over parameters h_j, z_j and the number of terms p . It is well known that this is an extremely complicated nonlinear problem even for the case where the order p of the exponential model is known in advance.

The traditional way to resolve this problem implies the application of iterative algorithms of multidimensional minimization. This approach has the following shortcomings. These algorithms are tedious, the obtained solution is very sensitive to the choice of starting values of the independent variables and the solution may converge to a local but not to the global extremum in

multidimensional space. However these methods as a rule are very flexible and can be applied to any non-linear approximation of sampled values and not necessarily to the exponential one. On the other hand for the latter approximation there is a group of specially developed methods usually defined as the Prony method [6]. The Prony method is not iterative and reduces the non-linear aspects of the problem to finding roots of polynomials.

The key point of the Prony method is to consider expression (11) as a general solution of some (yet unknown) linear difference equation

$$\sum_{m=0}^p a[m] x[n-m] = 0, \quad (16)$$

with constant coefficients $a[m]$ such that $a[0] = 1$. According to the general theory of these equations the complex constants z_j in Eq. (11) are the roots of characteristic equation

$$\sum_{m=0}^p a[m] z^{p-m} = 0. \quad (17)$$

Thus, the first stage of computational procedure is to determine the coefficients $a[m]$. There are two distinct cases here. If the number of sampled values $x[1], \dots, x[2p]$ is equal to the number of unknown exponential parameters $h_1, \dots, h_p, z_1, \dots, z_p$ we have a well determined problem and are able exactly to determine them from the set of linear equations (16) writing down for $p+1 \leq n \leq 2p$.

In the overdetermined case where $N > 2$ Eq. (16) should be rewritten in the form

$$\sum_{m=1}^p a[m] x[n-m] = e[n], \quad (18)$$

where $e[n]$ is the error of the forward prediction if the coefficients $a[m]$ are treated as forward prediction coefficients. In this case we have to minimize the sum of the squared errors $e[n]$ over all coefficients $a[m]$ so that the latter can be derived as a solution of covariance equations of forward prediction.

In the second stage obtained coefficients $a[m]$ are used to form a polynomial (17). Roots of this polynomial give the following estimators of damping coefficient α_j and frequency f_j for j the exponential

$$\alpha_j = \frac{\ln|z_j|}{\Delta}, \quad f_j = \frac{1}{2\pi\Delta} \arctg \frac{\text{Im} z_j}{\text{Re} z_j}. \quad (19)$$

If the complex constants z_j are known then Eq. (11) becomes linear with respect to the complex exponential parameters h_j . Minimization of sum of the squared errors of the exponential approximation (11) over these parameters leads to a standard normal equation of least square method

$$(\mathbf{Z}^H \mathbf{Z}) \mathbf{h} = \mathbf{Z}^H \mathbf{x}, \quad (20)$$

where superscript "H" denotes Hermitian conjugation,

$$\mathbf{Z} = \begin{pmatrix} 1 & 1 & \dots & 1 \\ z_1 & z_2 & \dots & z_p \\ \vdots & \vdots & \ddots & \vdots \\ z_1^{N-1} & z_2^{N-1} & \dots & z_p^{N-1} \end{pmatrix}, \quad \mathbf{h} = \begin{pmatrix} h_1 \\ h_2 \\ \vdots \\ h_p \end{pmatrix}, \quad \mathbf{x} = \begin{pmatrix} x[1] \\ x[2] \\ \vdots \\ x[N] \end{pmatrix}. \quad (21)$$

The solution of the set of linear equations (20) is the third stage of the computation procedure. Obtained com-

plex parameters are then used for estimations of amplitude A_j and phase constant θ_j of j the exponential (see Eq. (12))

$$A_j = |h_j|, \quad \theta_j = \arctg \frac{\text{Im} h_j}{\text{Re} h_j}. \quad (22)$$

4. ALGORITHMS

At the first stage of computational procedure we have to solve the covariance equations for linear prediction. For this purpose we used the fast algorithm initially proposed in [7] and modified later in [8]. This algorithm resembles the Levinson algorithm for Toeplitz matrices but is much more complicated because a Hermitian matrix of covariance normal equations is not a pure Toeplitz matrix but can only be represented as a product of matrices of this type. This algorithm like the Levinson one has recursive structure and therefore for a given order p allows us to obtain the whole set of least square solutions for all orders less than or equal to p . Moreover, the algorithm gives not only the solution of covariance equations for forward prediction but at the same time the similar solution for backward prediction.

At the second stage we used the Jenkins-Traub method [9] for finding the roots of a polynomial. This method finds roots (one per time), roughly speaking in ascending order of their modulus, then deflates found complex root and decreases the order of polynomial.

At the third stage we have to solve the set of normal equations of the linear least square method. Even for real coefficients of linear prediction the roots of the characteristic Eq. (17) in the general case will be complex and hence we have to solve the set of normal equations with a Hermitian matrix. We used for this purpose the standard algorithm of Cholesky decomposition [10].

5. RESULTS

To test the performance of the program *Q_fit* we used several series of simulated spectra. This approach is similar to the one proposed in [4]. Moreover, in order to compare the results of processing of the spectrum with different programs we used the same set of input parameters as in [4].

For a given set of lifetime values and relative intensities a number of different spectra were simulated which formed the series. Each spectrum in the series consists of a given number of counts. For each count we first selected a state in (1) and then for this state generated one deviate with an exponential distribution function. In both cases the inverse function method was used to transform the random deviate with a uniform probability distribution.

Each spectrum has been convoluted with a spectrometer resolution function which as in [4] was taken in the form of the single Gaussian with FWHM = 270 ps. For convolution we used a fast Fourier transform algorithm after prior setting up of buffer zone of zero-padded values at the end of a spectrum and bounding the Fourier transform of

spectrometer resolution function. Similar algorithms were used for the inverse operation of deconvolution. (The details can be found in [11]).

Series A. 2 components: 150 ps 50%, 250 ps 50%; number of channels 128; time calibration 33 ps/channel; counts per spectrum 2×10^6 ; number of spectra 5.

Let us use this simplest two-component spectrum with well separated and equally intensive components to illustrate the procedure of determining the number of states in (1). First of all we applied the program *Q_fit* to the pure noise-free spectrum when the sampled values were obtained by exact integration of the useful signal (1) over the channels of MCA. For a successively increasing order of exponential model (10) we obtained:

- 1) $p = 1$: pure decreasing real exponential (that is both frequency and phase constant are zero within the computational accuracy) and average lifetime $\tau \approx 195$ ps.
- 2) $p = 2$: the single exponential splits into two pure de-creasing real exponentials whose parameters are exactly equal to the input ones ($\tau_1 = 150$ ps, $I_1 = 0.5$; $\tau_2 = 250$ ps, $I_2 = 0.5$).
- 3) $p = 3, 4, 5$: all terms of the exponential model can be subdivided into two groups. The first group is comprised of two real exponentials, the same as in 2). The second group includes the remaining exponentials of the model and all of them within the computational accuracy have zero intensity.
- 4) $p = 6$: interruption in the subroutine for the fast solution of the covariance equations of linear prediction because in the iterative process the variance of forward prediction became negative.

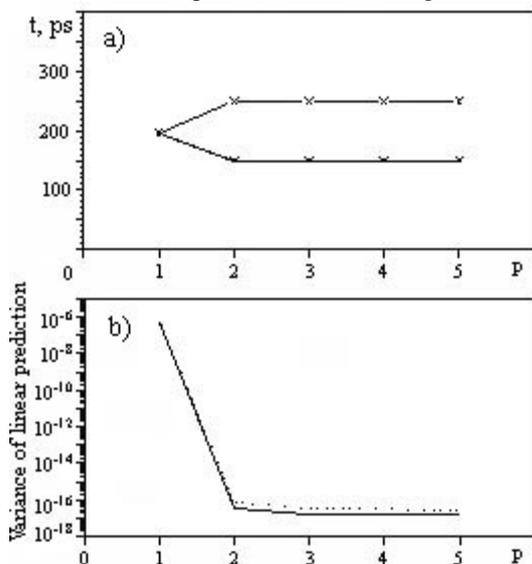


Fig. 1. Application of the program *Q_fit* to the noise-free spectrum of series A: (a) mean lifetime t of pure decreasing real exponentials with nonvanishing energy vs. order p of model; (b) variance of forward (solid line) and backward (dotted line) prediction

In Fig. 1,a mean lifetime values of real exponentials versus the order of exponential model are shown. From this figure it is clear that the spectrum has two components. The same conclusion can be drawn from the vari-

ance of linear prediction versus the order of exponential model dependency (Fig. 1,b). Indeed, for the value $p = 2$ divides into two regions with sharply different diminishing rate of variance of both the forward and backward prediction. That is although for $p = 2$ the variance, strictly speaking, does not reach a minimum we nevertheless are able to figure out when further addition of exponential terms does not lead to any practically sufficient improvement of the approximation.

Unfortunately, for simulated spectra the situation is quite different. In this case the number of counts on each grid point is subjected to the statistical fluctuation, and the spectrum as a whole consists of the useful signal and a noise. It can be seen from Fig. 2 that for such a spectrum the variance of both forward and backward prediction monotonically decrease as the order of the exponential model increases. Thus, we have to use a different approach to determine the number of states in a positron lifetime spectrum.

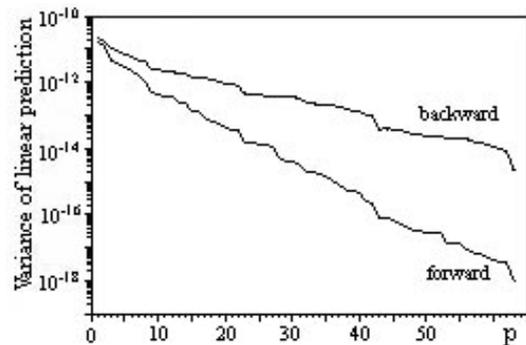


Fig. 2. Variance of linear prediction for one particular data set from series A

Fig. 1,a contains a hint of this approach. As was noted, processing of positron lifetime spectrum belongs to ill-posed inverse problem. As a rule for any particular inverse problem we can define two positive functionals F_A and F_B . The first of them measures the agreement of a model to the data while the second one measures something like the "smoothness" of the desired solution. When F_A by itself is minimized the agreement becomes very good, but the solution becomes unstable. In another extreme case, minimizing F_B by itself gives smooth solution that has nothing at all to do with the measured data. So, the central idea in inverse theory is to minimize the weighted sum of these two functionals.

It can be seen from Fig. 2 that increasing of exponential model order leads to the improvement of the agreement between the solution and the underlying spectrum, that is the solution becomes closer and closer to the nonsmooth initial signal. But the solution sought must look like the useful signal (1). So we have to smooth the derived solution. One way to do this is to maintain in it only a few most energetic pure decreasing exponentials.

The energy of a discrete sequence $x[i]$ ($i = 1, \dots, N$) from a standpoint of signal processing is defined as

$$E = \sum_{i=1}^N |x[i]|^2. \quad (23)$$

Therefore let us subdivide terms of exponential approximation into two groups. The first group will be comprised by pure decreasing real exponentials, that is, by terms j satisfying following conditions

$$\alpha_j < 0, |f_j| < \varepsilon, |\theta_j| < \varepsilon, \quad (24)$$

where ε is the machine-dependent precision parameter which in our calculation was taken as 10^{-5} . Another group will contain the remaining exponentials of the model, that is, terms for which any of the conditions (24) has broken down.

In Fig. 3,a for a particular data set from series A the mean lifetime t for all exponentials from the first group versus order of the exponential model is shown. It is seen from this figure that the first group is relatively small. For the order of exponential model from 1 to 63 the size of this group falls in the range 1 ($p = 1$) to 6 ($p = 56$).

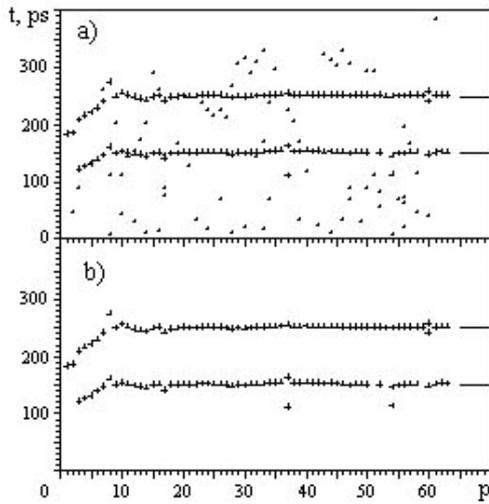


Fig. 3. Application of program Q_fit to the same data set from series A as in Fig. 2: (a) mean lifetime of pure decreasing real terms of the exponential approximation (10) whose energy is $\geq 0.005 E_s$ (large cross) $< 0.005 E_s$ (small cross), where E_s is the energy of the spectrum; (b) mean lifetime of pure decreasing real exponentials with nonvanishing energy. These exponentials form a regularized solution

Let us restrict our consideration to the spectra, which do not contain very weak components. In this case we can proceed further with a classification of exponential terms in approximation (10). For any spectrum we can calculate its energy E_s and then leave in the first group only those exponentials whose energy exceed some prescribed fraction (say, 0.005) of E_s (Fig. 3,b).

Thus far we have classified only individual terms of the exponential model. But there is also a restriction, which should be imposed on the first group of exponentials as a whole. Indeed, these exponentials will form the useful signal (1) and as any probability distribution function this signal should be normalized. Therefore we can impose on each useful component the set of following conditions (master conditions):

- 1) it is real and pure decreasing (Eq. (24));
- 2) its energy is not vanishingly small as compared with the energy of the spectrum;

- 3) for given order p these components as a whole form a normalized probability distribution function.

Let us define the optimal range of the exponential model as a range of p where the number of its terms satisfying master conditions and their mean values are virtually constant. It can be seen from Fig. 3,b that for series A the optimal range is defined by the following inequality $10 \leq p \leq 63$. Thus, we get the conclusion that a particular spectrum from this series consists of two components and these two components form the smoothed desired solution.

What we have just described can be viewed as a way to determine the number of states in a positron lifetime spectrum and at the same time as a regularization procedure applied to the inverse problem under study.

It is worthwhile to illustrate the resolution capability of the computational procedure. The energy spectral density of the exponential approximation (11) may be defined by numerous means depending on the adopted assumptions concerning the behavior of this approximation beyond the sampled range. It is generally believed that following two-side function

$$\hat{x}[n] = \begin{cases} \prod_{j=1}^M e^{-h_j z_j^n}, & n \geq 0; \\ \prod_{j=1}^M e^{-h_j (z_j^*)^{-n}}, & n < 0, \end{cases} \quad (25)$$

provides the best spectral estimations over other such functions. The z -transformation of the function (25) is given by

$$\hat{X}(z) = \prod_{j=1}^p h_j \frac{(z_j - 1/z_j^*) z^{-1}}{1 - (z_j + 1/z_j^*) z^{-1} + (z_j / z_j^*) z^{-2}}. \quad (26)$$

Hence, according to the general recipe, we can calculate the energy spectral density of the exponential approximation

$$E(f) = \left| \Delta \chi \hat{X}(z) \Big|_{z = \exp(i2\pi f \Delta)} \right|^2, \quad (27)$$

where f is the real frequency from the range $-f_c < f < f_c$ and $f_c = 1/(2\Delta)$ is the Nyquist critical frequency.

For the same data set as in Fig. 3 and for the highest possible order of exponential approximation ($p = 63$) the energy spectral density of this approximation is shown in Fig. 4. For comparison in this figure the energy spectral density of a regularized (smooth) solution is also shown. It can be seen that regularization procedure is equivalent to the extraction of noise from the exponential approximation.

The spectral density in Fig. 4 illustrates the resolution capability of the computational procedure taken in the ordinary sense, i.e. as a capability to distinguish two sine waves with close frequencies. But we are interested in a different definition of this term as the capability of the computational procedure to distinguish two pure decreasing exponentials with close damping coefficients. So let us formally treat frequency f as a complex variable $f = f + ia$ and consider the energy spectral density (27) as the section of the right-hand side complex function along the real axis in the complex frequency plane.

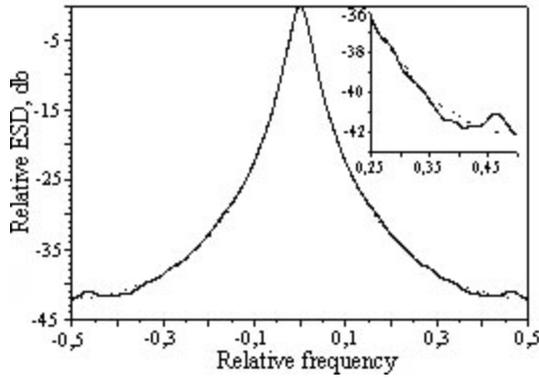


Fig. 4. Energy spectral density of the exponential approximation of data set from series A (solid line) and of the regularized solution (dotted line)

A similar section of this function along imaginary axis is shown in Fig. 5,a for the same order of the exponential model ($p = 63$) as in Fig. 4. For simplicity instead of the damping coefficient α we chose in Fig. 5 the mean lifetime τ ($\tau = -1/\alpha$) as an equivalent independent variable. Formally the function in Fig. 5,a represents the result of an analytic continuation of the function in Fig. 4 from the real axis into the complex frequency plane.

Fig. 5,a clearly demonstrates the high-resolution capability of the computational procedure to distinguish pure decreasing exponentials. (Note that y -axis in Fig. 5, has a logarithmic scale). From our point of view the high resolution power of the computational procedure may be ascribed to the covariance method of linear prediction and to the special character of the basic set used for the exponential approximation. The covariance method does not use a windowing operation and it is commonly adopted that the absence of this operation leads to the increasing resolution power in any spectral method.

It can be seen from the Fig. 3,b that in the range $3 \leq p \leq 7$ estimations of the average value of two pure decreasing exponentials are biased and the bias monotonically decreases as the p is increased. That is why we have to use an exponential model with a relatively high order. But in many spectral methods the higher is the order of the model the higher is the probability of splitting of the spectral line [3]. This is also true for the proposed computational procedure as can be seen from Fig. 5,b. In Fig5,b the same function as in Fig. 5,a is shown but calculated for a different order of the exponential model. For $p = 60$ the first group initially contains four pure decreasing exponentials (Fig. 3,a). One of them with the shortest lifetime has a very low intensity and hence can be eliminated. The three remaining exponentials satisfy the master conditions but this result takes place for a single value of p in the range $10 \leq p \leq 63$ and hence can be taken as accidental. That is why we have to consider the whole range of the exponential model order and make a statistically plausible conclusion, that is, conclusion that takes place for most values of p from this range. It should be noted that the fast algorithm for the solution of covariance equations of linear prediction applied for highest possible order p at

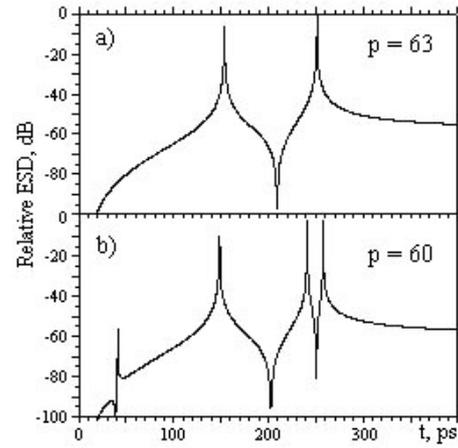


Fig. 5. Resolution capability of the Q_fit program for two values of the model order and the same data set from series A as in Fig. 3

the same time gives solutions for all orders less than or equal to p without any additional computation. So consideration of the whole range of p does not lead to any significant increasing in the time for spectrum processing.

Processing of the series of spectra is naturally subdivided into two successive steps. First, for one particular spectrum from the series the above-described procedure is applied. Results are the number of states in the positron lifetime spectrum and the optimal range of the exponential model order. At the second step these data serve as input parameters for the computational procedure. The program for a given value of p from the optimal range determines the parameters for all terms in the exponential approximation (10), it sorts them into two groups and checks up the first group against the master conditions. If both master conditions are satisfied and the number of states in the regularized solution is equal to the one determined at the first step then this solution is accepted. Otherwise the next value of p from the optimal range is selected.

In Table 1 are listed the results for series A obtained by *MELT* [4], *Posfit* [4] and *Q_fit*. In this table we give the mean values of lifetimes and intensities as well as the standard deviations. Both *MELT* and *Posfit* give reliable but slightly biased estimates. *Q_fit* gives very good unbiased results with low errors. From our viewpoint this distinction in estimates may be due to the different procedures of noise treatment adopted in these methods.

Table 1. Mean values for lifetimes τ_i and weights I_i ($i = 1, 2$) found by *MELT*, *Posfit* [4] and *Q_fit* for simulated spectra from the series A. The errors represent the standard deviations from the mean values

Analysis method	$t_1(dt_1)$, ps $I_1(dI_1)$, %	$t_2(dt_2)$, ps $I_2(dI_2)$, %
Simulation	150 50	250 50
<i>MELT</i> [4]	148.2 (4.0) 46.7 (3.3)	242.4 (4.0) 53.3 (3.3)
<i>Posfit</i> [4]	144.3 (5.6) 45.2 (3.6)	244.8 (3.3) 54.8 (3.6)
<i>Q_fit</i>	149.5 (3.1) 50.2 (1.8)	250.4 (1.6) 50.0 (2.0)

Table 2. Mean values for lifetimes τ_i and weights I_i ($i = 1, \dots, 4$) found by MELT, Posgauss [4] and Q_fit for simulated spectra from the series D. The errors represent the standard deviations from the mean values

Analysis method	$t_1(dt_1)$, ps	$t_2(dt_2)$, ps	$t_3(dt_3)$, ps	$t_4(dt_4)$, ps
	$I_1(dI_1)$, %	$I_2(dI_2)$, %	$I_3(dI_3)$, %	$I_4(dI_4)$, %
Simulation	100	250	600	1000
	25	25	25	25
MELT [4]	103.1 (0.6)	254.9 (4.3)	559.3 (14.1)	947.8 (6.4)
	25.27 (0.35)	23.94 (0.36)	20.92 (0.80)	29.87 (0.94)
Posgauss [4]	100.6 (0.7)	254.9 (4.8)	618.4(13.7)	1012.1 (8.1)
	25.31 (0.40)	25.34 (0.27)	25.89 (0.57)	23.47 (1.02)
Q_fit	100.2 (1.7)	245.9 (14.2)	592.0 (28.0)	998.9 (12.9)
	24.81 (1.32)	24.59 (0.78)	25.34 (0.93)	25.24 (1.78)

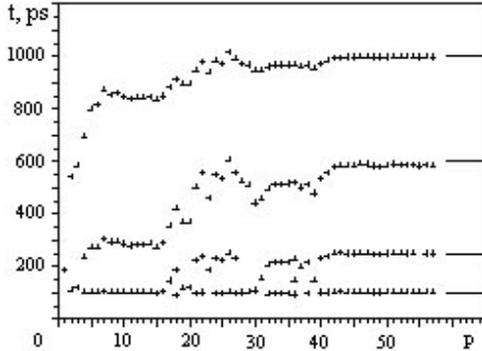


Fig. 6. Mean lifetime of the exponential model terms satisfying the master conditions for one particular data set from series D

The next two series illustrate the ability of the different methods to resolve components with close mean lifetimes.

Series B. 2 components: 150 ps 50%, 220 ps 50%; number of channels 128; time calibration 33 ps/channel; counts per spectrum 2×10^6 ; number of spectra 5. In this series the two components are closer than in series A. All methods are able to resolve the two components and give estimations with the same qualitative characteristics as in **series A**. The main difference consists in sufficient increasing of errors for all methods.

Series C. 2 components: 150 ps 50%, 190 ps 50%; number of channels 128; time calibration 33 ps/channel; counts per spectrum 2×10^6 ; number of spectra 5. "Here MELT, unable to resolve the two components at the optimal entropy weight, showed a wide peak with a mean lifetime of ≈ 169 ps" [4]. Posfit also runs into problems for this series. "The result obtained for the two component fits are not stable: In the first spectrum the two components are reproduced quite well, in two others the result is approximate, while in the remaining two a spurious short or long component appears" [4]. Q_fit are still able to resolve the two components and gives unbiased estimators but the errors have further increased up to the ± 10 % level on the mean lifetimes and to the ± 20 % level on the intensities found.

Series D. 4 components: 100 ps 25%, 250 ps 25%, 600 ps 25%, 1000 ps 25%; number of channels 128; time calibration 58 ps/channel; counts per spectrum 2×10^6 ; number of spectra 10. This is an artificial and

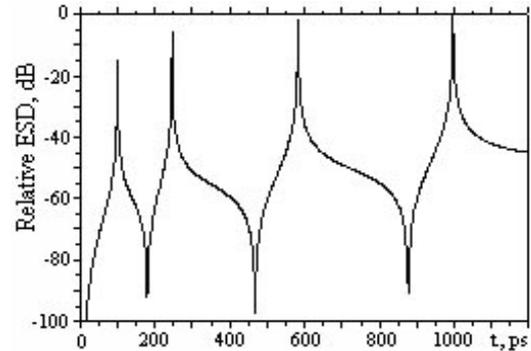


Fig. 7. Resolution capability of the Q_fit program for the same data set from series D as in Fig. 6

complicated data set used mainly to test the performance of the programs. The results found by different methods are summarized in Table 2. All methods resolve four components in spectra but the MELT results show a correlation between two longest components [4]. In this respect both Posgauss ("modified version of Posfit" [4]) and Q_fit performs better. As a whole Q_fit gives estimators with the smallest bias but with significantly larger errors. The scatter in the results of Q_fit may be due to the high resolution capability of this method.

In Fig. 6 for a particular data set from series D the mean lifetime for all exponentials satisfying the master conditions is shown. The tree structure in Fig. 6 is similar to the one for noise-free case but is stretched-out over approximately a ten times longer range of the p -axis. The whole range of the exponential model order in Fig. 6 can be subdivided into three intervals:

- 1) $8 \leq p \leq 15$;
- 2) $32 \leq p \leq 35$;
- 3) $42 \leq p \leq 57$,

where the parameters of pure decreasing energetic exponentials remain virtually constant separated by transition ranges. The first of them corresponds to a three component model, the second to the four components with biased estimations and the third to the four component model with unbiased estimations. So, the more exactly the noise in the spectrum is described the more reliable is the spectrum decomposition.

The optimal range of the exponential model order for the series D is much shorter than for series A and extends from 42 to 57. Fig. 7 illustrates the resolution capability of the computational procedure for one particular order of exponential model from the optimal range. It can be seen from this figure that the useful signal consists of four components, which are determined with high-resolution capability.

CONCLUSION

A new approach to analyze the positron lifetime spectra was proposed. This approach is based on a non-linear least square method but differs from the existing ones in that it uses fast algorithms and a more flexible basic set. This allows to use a model function with a large number of components and thus to describe readily and with reasonable accuracy both useful and noise parts of the spectrum.

These components can further be separated and this separation is equivalent to a regularization procedure. It should be stressed that in contrast to the common practice we did not use any low pass filter. Therefore, the proposed method is free from the undesired consequences of such filtering. As a result a regularized smooth solution gives unbiased estimations of parameters of the useful signal with a high-resolution capability.

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ПРИМЕНЕНИЕ ЦИФРОВОГО СПЕКТРАЛЬНОГО АНАЛИЗА К ЗАДАЧЕ ДЕКОМПОЗИЦИИ СПЕКТРОВ ВРЕМЕНИ ЖИЗНИ ПОЗИТРОНОВ

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Предложен новый метод численного анализа спектров времени жизни позитронов. Он основан на цифровом спектральном анализе, конкретно, на методе Прони. Сформулирована регуляризационная процедура решения обратной задачи нахождения параметров присутствующего в данных полезного сигнала. Предложен новый подход определения числа компонент в спектре. Результаты, полученные на модельных спектрах, демонстрируют высокую разрешающую способность метода.

ЗАСТОСУВАННЯ ЦИФРОВОГО СПЕКТРАЛЬНОГО АНАЛІЗУ ДО ЗАДАЧІ ДЕКОМПОЗИЦІЇ СПЕКТРІВ ЧАСУ ЖИТТЯ ПОЗИТРОНІВ

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Запропоновано новий метод чисельного аналізу спектрів часу життя позитронів. Він базується на цифровому спектральному аналізі, а саме на методі Проні. Сформульовано регуляризационну процедуру вирішення зворотної задачі знаходження параметрів корисного сигналу, який присутній у даних. Запропоновано новий підхід до визначення числа компонент у спектрі. Результати, отримані на модельних спектрах, демонструють високу роздільну здатність метода.