Appl. Phys. 16, 413—416 (1978)

Applied
Physics

Another Method of Deconvoluting Positron Annihilation Spectra Obtained by the Solid-State Detector

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Received 25 August 1977/Accepted 19 February 1978

Abstract. The use of various methods processing Doppler broadened annihilation spectra taking into account the detector resolution function and the restoration of the electron-positron pair momentum distribution is discussed. A statistic regularisation method is proposed. The method is found to be effective particularly for the study of electron momentum distribution in metals.

PACS: 06, 78.70

The production of Ge(Li) detectors with relatively high energy resolution (~ 1 keV) has resulted in extensive investigations of the annihilation behaviour of slow positrons, using the Doppler broadening of annihilation line (511 keV). The relation between the observed Doppler energy shift ΔE and the moment component of the positron-electron pair P_z along the observation axis is to a good approximation equal to

$$\Delta E = \frac{P_z}{2} E_0 , \tag{1}$$

where $E_0 = 511$ keV and P_z in mc units. The correlation between the energy distribution $f(\Delta E)$ of annihilation photons and the momentum distribution $N(P)$ of the positron-electron pair will be

$$f(\Delta E) = \int\limits_{2\frac{\Delta E}{E_0}}^{\infty} N(P)/P dP \tag{2}$$

(assuming an isotropic momentum distribution). Since the experimental energy spectrum is smeared by the instrumental resolution function $R(E)$, (2) can be written as

$$f(\Delta E) = \int\limits_0^{\infty} \left[\int\limits_{-PE_0/2}^{PE_0/2} R(E'-E) \frac{dE'}{P} \right] N(P) dP , \tag{3}$$

where $f(\Delta E)$ denotes the experimental spectrum.

As seen from (2) and (3), this method provides, in principle, the same information as the angular correlation method. However, some essential advantages such as the considerably greater rate of information storage, possible use of positron sources of rather low activity ($\sim 10\,\mu$C instead of a source strength of the order of 100 mC in the angular correlation system), simplicity of the experimental procedure. A disadvantage is the poorer resolution of Ge(Li) detectors compared to angular correlation technique (~ 1.0 keV or ~ 4 mrad in the angular scale). Thus, a correct method permitting allowance for smearing of the experimental spectrum by resolution function would increase the efficiency of the use of the Ge(Li) detectors considerably.

1. The Basic Relations

The instrumental resolution function $R(z)$ becomes a solution of the first-order Fredholm equation

$$f(x) = \int\limits_{-\infty}^{+\infty} R(x'-x)\phi(x')dx' , \tag{4}$$

where $f(x)$ is the experimental spectrum, $\phi(x)$ is the non-distorted spectrum.
The algebraic form of (4) is

$$f_j = \sum_{i=1}^{n} R_{ji}\phi_i , \quad j = 1, \ldots, n . \tag{5}$$

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0340-3793/78/0016/0413/$01.00

All earlier methods of solving (4) and (5) consist of the development of a certain interative process with step-wise control of the convergence of the square sum of the experimental $f(x)$ function deviations from the convolution of the i-th approximation $\phi^i(x)$ [1–5]. Vector $\boldsymbol{\phi}$ which minimizes the χ^2 value $\left[\chi^2=n^{-1}\sum_{i=1}^{n}(f_i^{(K)}-f_i^{\exp})^2\right]$, is taken as the problem solution. Further, the authors of [4, 5] approximated the obtained curve by the sum of some analytic functions (e.g. Gaussian) and use the smooth curve obtained as a primary approximation to the next iterative process. The procedure is repeated until the minimum of χ^2 is obtained.

Though the authors of [4, 5] managed to obtain a fair correlation of the deconvoluted spectra from the Ge(Li) detector and of the angular correlation spectra, their method seems to have some essential drawbacks, such as:

1) The method ignores the statistic nature of the experimental function f and consequently makes it impossible to estimate the deconvolution error.

2) The method is valid only for curves with the best statistics (more than 10^6 counts in the peak). Otherwise a preliminary smoothing of the spectrum is needed, which would be an incorrect procedure.

3) If the primary approximation is chosen wrong, the convergence might not be obtained.

4) In general, χ^2 cannot be a true criterion in such problems, since the initial approach, (4), is incorrect.

These difficulties arise from the poor stipulation of the system, i.e. from the strong dependence of the solution $\boldsymbol{\phi}$ on variations in f, the coefficient errors, R_{ij}, and the miscalculations.

For the reason it seems more expedient to use the statistical regularisation method for solution of (4), developed in [7, 8]. We have used it earlier in processing the angular correlation spectra [9].

2. The Statistical Regularisation Method

The method implies treatment of (4) as a mathematical statistics problem and the regularizer (the a priori information) is introduced as some probability distribution for $\boldsymbol{\phi}$.

Let f_j be measured with a dispersion S_j^2, independent of different j and with normal distribution. Then the conditional probability that at the given $\boldsymbol{\phi}$ vector f can be measured in

$$P(f/\phi)=\prod_{j=1}^{n}\frac{1}{\sqrt{2\pi}\cdot S_j}\exp\left[-\frac{\left(f_j-\sum_{i=1}^{n}R_{ji}\phi_i\right)^2}{2S_j^2}\right]. \tag{6}$$

This relation can be rewritten in matrix form

$$P(\boldsymbol{f}/\boldsymbol{\phi})=C_1\exp[-\tfrac{1}{2}(\boldsymbol{\phi},B\boldsymbol{\phi})+(\boldsymbol{a},\boldsymbol{\phi})], \tag{7}$$

where $B=R^+WR$, $\boldsymbol{a}=R^+W\boldsymbol{f}$

$$W_{ij}=\delta_{ij}/S_j^2 \qquad (i,j=1,\dots,n),$$

$C_1=\text{const}$, independent of $\boldsymbol{\phi}$. Since the only a priori information about $\phi(x)$ is that it is a smooth function, the priori statistic ensemble will be [7]

$$P_\alpha(\boldsymbol{\phi})=C_2\alpha^{\frac{n}{2}}\exp\left[-\frac{\alpha}{2}(\boldsymbol{\phi},\Omega\boldsymbol{\phi})\right], \tag{8}$$

where Ω is a positively defined symmetric matrix such that the functional $(\boldsymbol{\phi},\Omega\boldsymbol{\phi})$ gives a finite difference approximation to the integral $\int(d^2\phi/dx^2)^2dx=\text{const}$, α is the regularisation parameter, $C_2=\text{const}$, independent of $\boldsymbol{\phi}$.

A distribution for $\boldsymbol{\phi}$ is given by the Bayes expression [7]

$$P(\boldsymbol{\phi}/\boldsymbol{f},\alpha)=\frac{P(\boldsymbol{f}/\boldsymbol{\phi})P_\alpha(\boldsymbol{\phi})}{\int P(\boldsymbol{f}/\boldsymbol{\phi})P_\alpha(\boldsymbol{\phi})d\boldsymbol{\phi}}. \tag{9}$$

Substituting (6) and (8) into (9) we obtain

$$P(\boldsymbol{\phi}/\boldsymbol{f},\alpha)=C_3\exp\{-\tfrac{1}{2}[\boldsymbol{\phi},(B+\alpha\Omega)\boldsymbol{\phi}]+(\boldsymbol{a},\boldsymbol{\phi})\}, \tag{10}$$

where $C_3=\text{const}$, independent of $\boldsymbol{\phi}$. The normal distribution obtained for the probability $P(\boldsymbol{\phi}/\boldsymbol{f},\alpha)$ gives the most exhaustive solution of the problem with α in given a priori. The mathematical expectation of $\boldsymbol{\phi}$ over a distribution, (10), is taken as the deconvolution function

$$\langle\boldsymbol{\phi}\rangle_\alpha=(B+\alpha\Omega)^{-1}\boldsymbol{a}. \tag{11}$$

The deconvolution error is defined as the root of $\boldsymbol{\phi}$ dispersion:

$$\sigma_i=\sqrt{(B+\alpha\Omega)_{ii}^{-1}} \qquad (i=1,\dots,n). \tag{12}$$

Thus, (11) and (12) give the most probable fair solution at given vectors $\boldsymbol{f}$ and $\boldsymbol{S}$. It is important that this solution is obtained by using the minimal and reasonable a priori information about the smoothness of function $\phi(x)$.

However, since in most practical cases the parameter α is an unknown algorithm a solution of (4) has, in this case, been found [7]. In computer realization this algorithm consists in search for the most probable $\alpha=\alpha_0$ and in finding the solution with $\alpha=\alpha_0$ according to (11) and (12).

3. Results and Conclusions

To illustrate the efficiency of the program the annihilation spectra in water and benzene were measured at $t°=22°$ C. Each experimental spectrum was obtained using a digitally stabilized Ge(Li) system with resolution (FWHM) 1.33 keV. The statistics in each curve peak were $\sim 10^6$ counts. The detector resolution was determined by recording the 514 keV γ-ray peak of ^{85}Sr with statistics of $\sim 3 \cdot 10^4$ counts. For convenience, the resolution function was described as the sum of two Gaussians (92.9%, $\theta = 1.34$ keV; 7.1%, $\theta = 3.32$ keV, 0.987 keV shift to the left) and was used as such in all calculations. It should be noted that in general it can be used in a tabular form.

Before the processing of each spectrum the background was subtracted [5].

$$B_i = B_L - \left(\sum_{j=1}^{i} f_j/A\right)(B_L - B_H), \tag{13}$$

where B_i stands for the background in the i-th channel, B_L for the background in the low energy region of the spectrum, B_H for the background in the high energy region of the spectrum, and A for the sum of counts beneath the curve.

This approximate description of the background seems to be quite sufficient for practical purposes and does not affect the ultimate result.

The program was tested in model experiments. The curves with a "narrow" component corresponding to annihilation of p-Ps atoms were naturally the most difficult for deconvolution.

The model experiments were conducted as follows. Setting the $\phi(x)$ function (angular correlation curves of some substances were used as a rule) smeared it by a nucleus equation (4), and adding the normally distributed error we got "the experimental" function $f(x)$ from

$$f_j = \sum_{i=1}^{n} R_{ji}\phi_i + \left(\sum_{i=1}^{n} R_{ji}\phi_i\right)^{1/2} \xi_j, \quad j = 1, \dots, n, \tag{14}$$

where ξ_i is a normally distributed value with mathematical expectation 0 and dispersion 1.

Carrying out back deconvolution we compared the obtained result with the initial function ϕ. The restoration was considered good if the results coincided within the convolution error. The statistics of $\sim 10^6$ counts in the peak appeared to be sufficient for restoration of even narrow curves (with a strong narrow component). Broad curves (without a narrow component) could be restored with statistics of $\sim 10^5$ counts in the curve peak.

The deconvolution of non-distorted curves from experimental spectra of benzene and water is shown in Fig. 1. [15% of positron annihilation in Ni foil covered the source and 12% of positron annihilation in the source material (NaCl) were taken into consideration in Ge(Li) experiments.]

Comparison of the results obtained by other methods of solving (4) seemed to be of interest. The method of

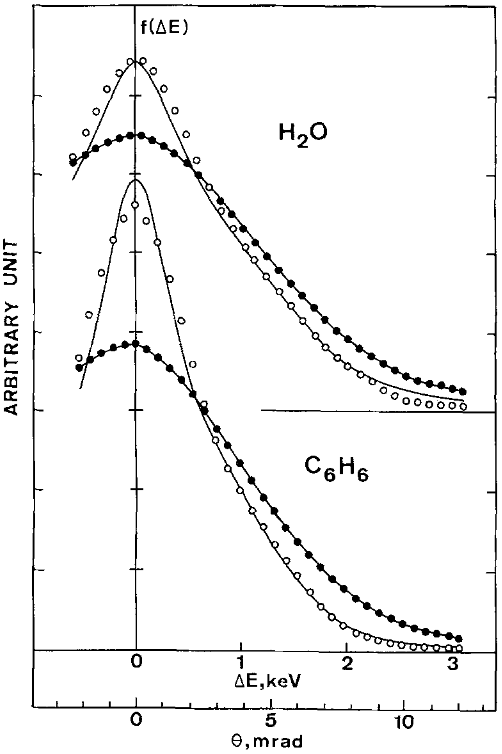

Fig. 1. Doppler broadened annihilation line in benzene and water. ● The experimental curve, ○ the deconvoluted line, – solid line, angular correlation curve, corrected on the resolution function [11, 12]. The uncertainties are smaller than the points. All curves are normalized to equal areas

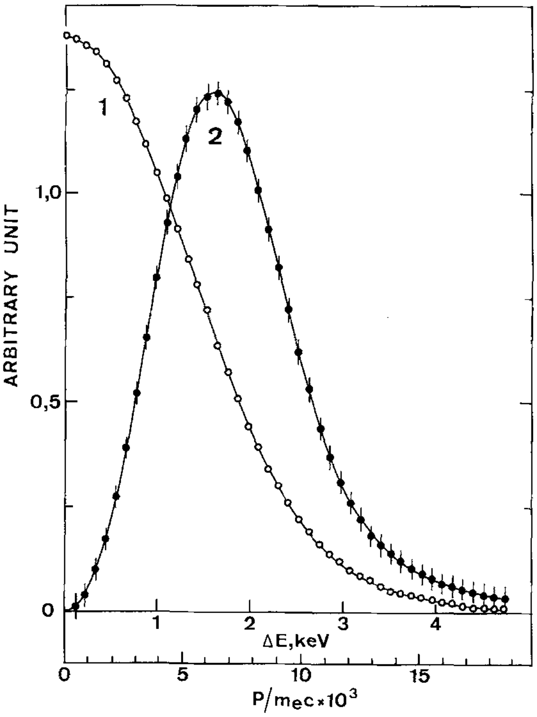

Fig. 2. Curve *2* is the model momentum distribution: *1* is the experimental energy spectrum, *2* is the deconvoluted curve

ordered minimization of average risk developed by Vapnik and Chervonenkis [10] was used, because this method, as well as the statistical regularisation method, were most suitable for the purpose. The restoration curves appeared to coincide with those in Fig. 1 to an extent such that they could not be differentiated. Since the two methods implying different treatment gave the same results the solution obtained seems to be true.

It will be noted that in a number of cases (for example, the problem of positron annihilation in metals) information about the momentum distribution of annihilating positron-electron pair provides more information than the energetic photon spectrum. Here, (3) should be used.

However, model experiments show that for curves with an intense narrow component (~ 15 to 20%) both methods gave a smooth solution even with good statistics ($\sim 10^6$ to 10^7 counts in the peak), although they represent very well the broad part of the curve. At the same time very good deconvolution was obtained in the peak (Fig. 2). Thus this method might appear to be very effective, for instance in the study of metals.

It will be noted that the above drawback of the statistic regularisation method does not seem to be essential in principle, in this case, and is rather a consequence of approximate computer realization of the full algorithm of finding solution with an "a priori" unknown regularization parameter α, as the limited computer efficiency compelled us to seek the solution only with a single, most probable α_0.

Acknowledgements. The authors wish to thank Dr. V. N. Vapnik for the kindly offered program and Dr. V. Z. Nozik for the valuable discussions.

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