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ELSEVIER

Study of the Van Cittert and Gold iterative methods of deconvolution and their application in the deconvolution of experimental spectra of positron annihilation

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Abstract

The study of deconvolution by Van Cittert and Gold iterative algorithms and their use in the processing of experimental spectra of Doppler broadening of the annihilation line in positron annihilation measurement is described. By comparing results from both algorithms it was observed that the Gold algorithm was able to eliminate linear instability of the measuring equipment if one uses the 1274 keV ^{22}Na peak, that was measured simultaneously with the annihilation peak, for deconvolution of annihilation peak 511 keV. This permitted the measurement of small changes of the annihilation peak (e.g. *S*-parameter) with high confidence. The dependence of γ-ray-like peak parameters on the number of iterations and the ability of these algorithms to distinguish a γ-ray doublet with different intensities and positions were also studied.

1. Introduction

Positron annihilation (PA) as one of the non-destructive nuclear methods is used for the study of structure of different materials and it is especially efficient in determining free volumes or defects in bulk material. The main characteristics, that are determined by PA, are lifetimes of positrons in matter, Doppler broadening of annihilation line (DBAL), and angular correlation of annihilation γ-rays. The last two characteristics give us information about the momentum distribution of electrons in matter. The more common angular correlation method is approximately one order more accurate, but it is more time consuming and requires a more intensive positron source. On the other hand DBAL is simple and not time consuming. It is mainly used for determining the relative changes in the annihilation peak (e.g., *S*-parameter, defined as the ratio between area of central part of the peak to the full area of the peak) depending on temperature, pressure etc. After deconvolution of the annihilation peak using a resolution function of measuring system, one can obtain the momentum distribution of electrons in the sample. If we are only interested in relative changes of the *S*-parameter, deconvolution is not necessary. But as we have seen in our work, deconvolution can partly eliminate the spurious effects due to instability of the measuring system. Another possibility to eliminate an instability of the system is the use of an expensive spectrum stabilizer unit.

Generally, deconvolution algorithms have recently found many applications in various domains of experimental science. They have been applied to such different problems as improving resolution in spectroscopy and measuring the thickness of multilayer structures. There is also the long-standing problem of determination of position and intensities in γ-ray multiplets [1].

2. Theory

The relationship between a measured value $x(t)$ and the raw result of measurement $y(t)$ can be described by a convolution-type integral equation

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$$y(t) = \int_{-\infty}^{\infty} x(\tau)h(t-\tau)\,\mathrm{d}\tau\,, \tag{1}$$

where $h(t)$ is an impulse response. Knowledge of the instrumental function $h(t)$ is usually required. For a discrete system, Eq. (1) can be written as

$$y(i) = \sum_{k=0}^{N-1} h(i-k)x(k) \qquad i = 0, 1, \ldots, 2N-2\,, \tag{2}$$

where N is the number of samples of vectors $\boldsymbol{h}$, $\boldsymbol{x}$.

The impulse response has a finite length. Therefore we will assume that $h(i) = 0$ for $i < 0$ and $i \geqslant N$. Then Eq. (2) can be written in a matrix form:

$$\begin{bmatrix} y(0) \\ y(1) \\ y(2) \\ \vdots \\ \vdots \\ \vdots \\ \vdots \\ y(2N-2) \end{bmatrix} = \begin{bmatrix} h(0) & 0 & 0 & \ldots \\ h(1) & h(0) & 0 & \ldots \\ h(2) & h(1) & h(0) & \ldots \\ \vdots & \vdots & \vdots & \ddots \\ h(N-1) & h(N-2) & h(N-3) & \ldots \\ 0 & h(N-1) & h(N-2) & \ldots \\ 0 & 0 & h(N-1) & \ldots \\ \vdots & \vdots & \vdots & \ddots \end{bmatrix} \begin{bmatrix} x(0) \\ x(1) \\ x(2) \\ \vdots \\ x(N-1) \end{bmatrix}, \tag{3}$$

or

$$\boldsymbol{y} = \mathbf{H}\boldsymbol{x}\,. \tag{4}$$

It means that the columns of $\mathbf{H}$ are represented by vectors $\boldsymbol{h}$ mutually shifted by one position. Multiplying both sides of Eq. (4) by $\mathbf{H}^{\mathrm{T}}$ gives

$$\mathbf{H}^{\mathrm{T}}\boldsymbol{y} = \mathbf{H}^{\mathrm{T}}\mathbf{H}\boldsymbol{x}\,, \tag{5}$$

or

$$\boldsymbol{y}_1 = \mathbf{H}_1\boldsymbol{x}\,, \tag{6}$$

where $\mathbf{H}_1$ is Toeplitz matrix [2,3].

Solution of the linear equation system (6) (vector $\boldsymbol{x}$), under the condition that the output vector $\boldsymbol{y}$ and the matrix of the impulse response $\mathbf{H}$ is known, is a problem of deconvolution. The output vector of system $\boldsymbol{y}$ is affected by noise that accompanies each measurement. The existence of this noise strongly affects the process of deconvolution, and can lead to difficulties in solving the linear equation system (6).

The above-formulated problem of input reconstruction is as a rule ill-conditioned, i.e. the estimates $\hat{\boldsymbol{x}}(t)$ of $\boldsymbol{x}(t)$ satisfying Eq. (1) are extremely sensitive to errors in the measured data $\boldsymbol{y}(t)$. It is expressed by the fact, that the matrix $\mathbf{H}_1$ is almost singular. The direct inversion of $\mathbf{H}_1$ for solving $\boldsymbol{x}$ cannot lead to a stable solution.

Therefore, in order to solve this problem, the method of regularization must be included. This means that original problem is replaced by an approximate one which solutions are significantly less sensitive to errors in the data $\boldsymbol{y}(t)$.

The Van Cittert iterative method of deconvolution is widely applied in different areas, for example in spectroscopy or in image processing [2,4]. The Van Cittert algorithm of deconvolution is described in detail in Ref. [4], so we will describe it only very briefly.

Its basic form for a general linear discrete system is

$$\boldsymbol{x}^{k+1} = \boldsymbol{x}^k + \mu(\boldsymbol{y} - \mathbf{A}\boldsymbol{x}^k)\,, \tag{7}$$

where **A** is system matrix, k represents the number of iterations and μ is the relaxation factor. The convergence condition of Eq. (7) is that the diagonal elements of the matrix **A** satisfy

$$A_{ii} > \sum_{j=0, j\neq i}^{N-1} A_{ij}\,, \qquad i = 0, 1, \ldots, N-1\,. \tag{8}$$

It is obvious that such a diagonal element dominance is rare in physical problems. However, the deconvolution algorithm in Eq. (7) can be modified in a such way that it will satisfy the conditions of convergence. Hence, Eq. (7) becomes

$$\boldsymbol{x}^{k+1} = \mu\boldsymbol{y} + (\mathbf{E} - \mu\mathbf{A})\boldsymbol{x}^k = \mu\boldsymbol{y} + \mathbf{D}\boldsymbol{x}^k\,, \tag{9}$$

where **E** is a unit matrix and

$$\mathbf{D} = \mathbf{E} - \mu\mathbf{A}\,. \tag{10}$$

Under the condition that $\boldsymbol{x}^0 = \mu$, the successive substitutions give

$$\boldsymbol{x}^k = \mu\boldsymbol{y} + \mu\mathbf{D}\boldsymbol{y} + \ldots + \mu\mathbf{D}^{k-1}\boldsymbol{y} + \mu\mathbf{D}^k\boldsymbol{y} = \mu(\mathbf{E} + \mathbf{D} + \ldots + \mathbf{D}^k)\boldsymbol{y}\,. \tag{11}$$

Assuming that $\lambda_0, \lambda_1, \ldots, \lambda_{N-1}$ are eigenvalues of **A**, then $(1 - \mu\lambda_0), (1 - \mu\lambda_1), \ldots, (1 - \mu\lambda_{N-1})$ are eigenvalues of **D**. If

$$\lim_{k\to\infty} (1 - \mu\lambda_i)^k = 0\,, \qquad i = 0, 1, \ldots, N-1\,, \tag{12}$$

then

$$\lim_{k\to\infty} \mathbf{D}^k = [0]\,,$$

and

$$\lim_{k\to\infty} \boldsymbol{x}^k = \mathbf{A}^{-1}\boldsymbol{y} = \boldsymbol{x}\,. \tag{13}$$

From equation (12) this implies that the necessary and sufficient conditions of convergence are

$$|\,1 - \mu\lambda_i\,| < 1\,, \qquad i = 0, 1, \ldots, N-1\,. \tag{14}$$

If we define λ_i and its conjugate λ_i^* as

$$\lambda_i = a_i + \mathrm{j}b_i\,, \qquad \lambda_i^* = a_i - \mathrm{j}b_i\,,$$

then the convergence condition (14) becomes

$$\mu[\mu(a_i^2 + b_i^2) - 2a_i] < 0\,, \qquad i = 0, 1, \ldots, N-1\,. \tag{15}$$

Inequality (15) gives two bounds for μ

$$\mu = 0\,, \qquad \mu = 2\frac{a_i}{a_i^2 + b_i^2}\,, \qquad i = 0, 1, \ldots, N-1\,. \tag{16}$$

N conditions determine the bounds of the μ coefficient. Unfortunately these conditions are not fulfilled for all cases. However, if the system matrix **A** is positive definite the convergent solution always exists. So we settle the algorithm in a such way that the eigenvalues λ_i will be positive, real numbers.

Let us return to Eq. (5). Matrix $\mathbf{H}^{\mathrm{T}}\mathbf{H}$ is symmetric, so its eigenvalues are real. The eigenvalues of matrix $(\mathbf{H}^{\mathrm{T}}\mathbf{H})(\mathbf{H}^{\mathrm{T}}\mathbf{H})$ are squares of eigenvalues of matrix $\mathbf{H}^{\mathrm{T}}\mathbf{H}$ and therefore must be positive. Eq. (5) becomes

$$(\mathbf{H}^{\mathrm{T}}\mathbf{H}\mathbf{H}^{\mathrm{T}})\boldsymbol{y} = (\mathbf{H}^{\mathrm{T}}\mathbf{H}\mathbf{H}^{\mathrm{T}}\mathbf{H})\boldsymbol{x}\,, \tag{17}$$

and the iterative algorithm of deconvolution becomes

$$\boldsymbol{x}^{k+1} = \boldsymbol{x}^k + \mu[(\mathbf{H}^{\mathrm{T}}\mathbf{H}\mathbf{H}^{\mathrm{T}})\boldsymbol{y} - (\mathbf{H}^{\mathrm{T}}\mathbf{H}\mathbf{H}^{\mathrm{T}}\mathbf{H})\boldsymbol{x}^k], \tag{18}$$

or

$$\boldsymbol{x}^{k+1} = \boldsymbol{x}^k + \mu[\boldsymbol{y}' - \mathbf{H}'\boldsymbol{x}^k]. \tag{19}$$

By this we ensure the existence of a common interval of solution for inequality (14) and convergence of the deconvolution algorithm. Eigenvalues λ_i are real, positive numbers, so from Eq. (16) for μ we can write

$$0 < \mu < \frac{2}{\lambda_{\max}}, \tag{20}$$

where $\lambda_{\max}$ is the greatest eigenvalue of $\mathbf{H}'$

$$\lambda_{\max} = \max(\lambda_0, \lambda_1, \ldots, \lambda_{N-1}). \tag{21}$$

Now we determine the maximum eigenvalue $\lambda_{\max}$. For eigenvalues of system (17) we can write

$$\mathbf{H}'\boldsymbol{x} = \lambda_i \boldsymbol{x}, \qquad i = 0, 1, \ldots, N-1. \tag{22}$$

If x_j is the biggest absolute element in $\boldsymbol{x}$, then from Eq. (22) we get

$$\sum_{m=0}^{N-1} \mathbf{H}'_{jm} x_m = \lambda_i x_j \tag{23}$$

or

$$\lambda_i = \frac{\sum_{m=0}^{N-1} \mathbf{H}'_{jm} x_m}{x_j}. \tag{24}$$

Then

$$\lambda_i \leqslant \sum_{m=0}^{N-1} |\mathbf{H}'_{jm}|, \qquad i = 0, 1, \ldots, N-1. \tag{25}$$

In practical cases we do not know the biggest element in $\boldsymbol{x}$. We determine the value of $\lambda_{\max}$ as the maximum value from λ_i, determined by Eq. (25) e.g., from the sum of absolute values of rows in matrix $\mathbf{H}'$.

This is the base of the Van Cittert algorithm of deconvolution. Now we introduce, in analogy with Eq. (24), a local variable relaxation factor

$$\mu_i = \frac{x_i^k}{\sum_{m=0}^{N-1} \mathbf{H}'_{im} x_m^k}, \tag{26}$$

and we use it in Eq. (19). For the ith element of vector $\boldsymbol{x}^{k+1}$ we get

$$x_i^{k+1} = x_i^k + \frac{x_i^k}{\sum_{m=0}^{N-1} \mathbf{H}'_{im} x_m^k} \left[y_i - \sum_{m=0}^{N-1} \mathbf{H}'_{im} x_m^k \right]. \tag{27}$$

or

$$x_i^{k+1} = \frac{y_i}{\sum_{m=0}^{N-1} \mathbf{H}'_{im} x_m^k} x_i^k . \tag{28}$$

Eq. (28) is the Gold algorithm of deconvolution [5]. It is an extension of Van Cittert's iterative method. The advantage of Gold's method is that the solution x is positive.

3. Theoretical simulation

To test an applicability of Van Cittert's and Gold's methods of deconvolution for γ-rays spectra we performed a theoretical simulation. We convoluted the experimental resolution function (RF) obtained from a positron annihilation experiments with different kinds of Gaussians describing γ-ray peaks. The simulated spectra were used as an input to both Gold's and Van Cittert's iterative deconvolution procedures. Simulated spectra included a noise component due to an experimental resolution function.

3.1. The dependence of output peak parameters from the deconvolution on the number of iterations

Because the deconvolutions are using the iterative method, it was interesting to study the dependence of the output from the deconvolution on the number of iterations (N). N was varied in the range 200–50 000. 2000 iteration steps lasted 40 min on a PC486, 33 MHz for 200 channels of the spectrum (a version with elimination of the multiplication of zero matrix terms took approx. 15 min).

We analyzed the peak which was a convolution of a Gaussian with FWHM = 13 channels and a resolution function with a FWHM = 10.4. The resulting peak had a FWHM = 16.7, which was a value in the range of our experimental values for the FWHM of annihilation peaks measured in DBAL. The deconvoluted peak was fitted and the result should be one Gaussian with a FWHM = 13.

Fig. 1 shows the dependences of the χ^2 value and FWHM on the number of iterations for the algorithm of Van Cittert and Gold. As we can see from this figure, the resulting deconvolution fit for the Van Cittert algorithm was much better for two Gaussians than for one until 14 000 iterations, then the χ^2 became practically the same.

For the Gold algorithm the χ^2 of the deconvolution fit was very good for one Gaussian and reached a minimum value for 2000 iterations. The increase of χ^2 with the number of iterations (for both Van Cittert and Gold methods) is a consequence of the round-off errors which arise during the computations with real numbers represented in standard single precision (REAL in Fortran or FLOAT in C) and a large number of these

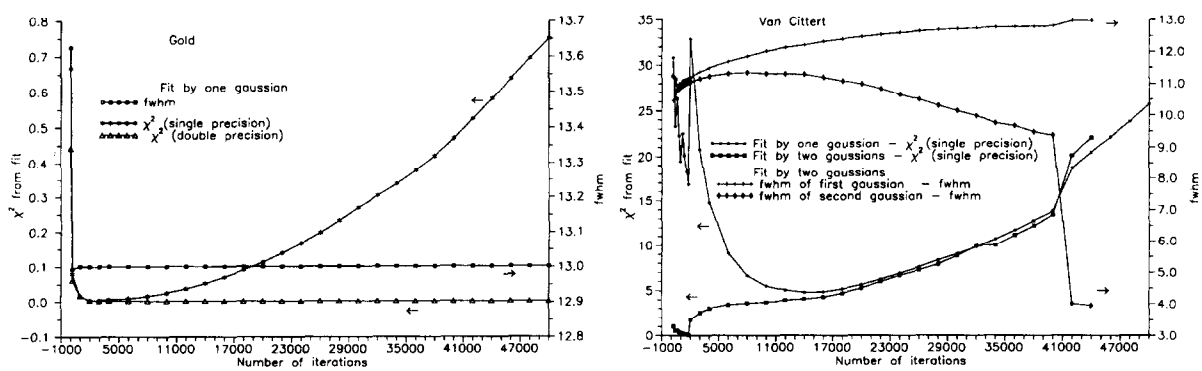

Fig. 1. Dependence of χ^2 and FWHM from fit of deconvoluted peak on number of iteration steps.

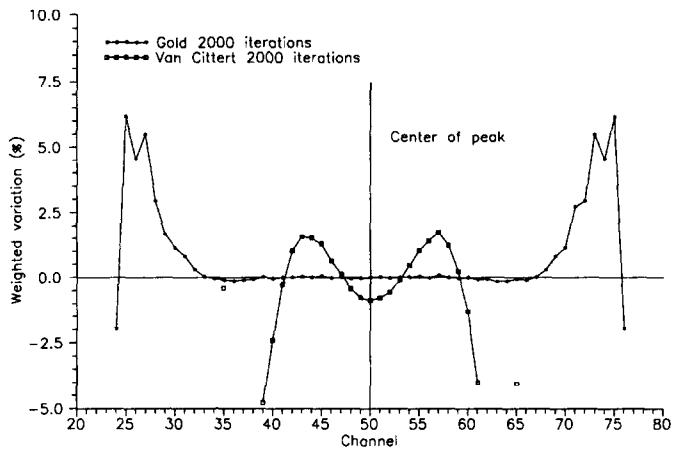

Fig. 2. Weighted variations of deconvoluted peak and original Gaussian.

operations. It was verified by a calculation (for the Gold method) using the double precision floating-point representation of real numbers. The results of the double-precision calculation are shown in Fig 1. The fast convergence and the stability of the solution can be observed. It clearly shows the possible errors arising from using a finite computer word length of and a large number of iteration steps.

For the Van Cittert algorithm, it can be seen that by increasing the number of iterations the first Gaussian became more and more dominant and approached the input Gaussian in convolution. The same dependence can be observed in the intensity of Gaussians.

For the Gold algorithm, the results were much better. From Fig. 1, it is clear that the FWHM from the fit of the deconvoluted peak reached the correct value of 13 for the number of iterations larger than 1000. The variance of the FWHM from this fit was $< 0.03\%$ and of the amplitude $< 0.005\%$ for $N \geqslant 1000$.

For the Van Cittert algorithm and $N \geqslant 2000$, the variance of the FWHM from the fit was $< 0.3\%$ and of the amplitude $< 0.05\%$.

In Fig. 2 the weighted differences of the original input Gaussian and fitted results of the deconvoluted peak after $N = 2000$ iteration steps are shown. The difference is evident. For the Gold deconvolution the variance was still less than 7.5% even for the ± 25 displaced channels from the center of the peak. For the large number of iterations ($N > 30\,000$) there was already deformation in deconvoluted peak, which could be seen as sharp peaks in the dependence of weighted differences. This deformation is a consequence of round-off errors in the computer.

Fig. 3 shows typical results of the deconvolution procedures. The peak, deconvoluted by the Gold's algorithm, is almost identical with the original Gaussian after carrying out 2000 iterations. Van Cittert's procedure showed negative values and there were oscillations on both wings of the peak on the level of 2% of amplitude of peak. Because Fig. 3 is in a logarithm scale the negative values are not displayed. Gold's deconvolution did not produce negative values and oscillations were very small ($< 0.01\%$).

3.2. The deconvolution of Gaussians with different FWHM

To determine the influence of the FWHM of the convoluted Gaussian on the result of deconvolution, we also made a convolution of the Gaussian with FWHM = 20 with RF. Then the difference in parameters of the resulting deconvoluted peak after 2000 iterations was compared with the difference for FWHM = 13.

The Van Cittert's deconvolution of convolution of the original Gaussian with a FWHM = 20 was in very good agreement with the original Gaussian, χ^2 from the fit was 0.04 and the FWHM was exactly the same.

The weighted difference of the fit of deconvoluted and original Gaussian was $\leqslant 10\%$ for counts $> 0.1\%$ of amplitude. The situation was worse for the convolution of the Gaussian with FWHM = 13. The FWHM of the deconvoluted peak was larger than about 1.8%. But the worst fact was that the χ^2 from the fit was

very large (Fig. 1) and the deconvolution was better described by two Gaussians ($\chi^2 = 1.8$). That means that the deconvolution changed the shape of the peak in such a way that the deconvoluted peak could be better described by two Gaussians though there should be only one. The weighted difference of the deconvolution fit and the original Gaussian was $\leqslant 10\%$ for counts $> 3\%$ of the amplitude. The parameters from the fit of deconvoluted peak were in excellent agreement with the original Gaussian. Problem was that for a Gaussian with FWHM = 13 there was a better χ^2 for two Gaussians as for one.

For the Gold deconvolution agreement with the original Gaussian was very good also for Gaussian with FWHM = 13 as we can see from Fig. 2. What was important was that there are no deformations in deconvoluted peak because the χ^2 for one Gaussian was very small (Fig. 2).

3.3. The deconvolution of the sum of two Gaussians with different intensity and position

To verify the possibility of distinguishing components of a doublet (sum of two peaks) by deconvolution, we made convolutions of the sum of two Gaussians with intensity ratios 1:1, 1:0.8, 1:0.5, 1:0.2 and mutual displacements of 1, 5, 10, 15, 20 channels. Both input Gaussians had FWHM = 13, the number of iteration steps N was 2000 and the deconvoluted peaks were fitted. We computed the weighted difference of these fitted parameters with the parameters of the original input Gaussians.

For the Van Cittert's deconvolution we got the following results:

If the peak displacement was 15 and 20 channels there was no problem to fit the deconvolution results. The variation from the parameters of the input Gaussians was small, $\leqslant 2\%$. For a displacement of about 10 channels, the fitting procedure for the ratio of intensities 1:0.2 failed. The variance was $\leqslant 5\%$ for ratios 1:1,1:0.8 and $< 19\%$ for 1:0.5. For a displacement of about 5 channels we were not able to get a fit for 1:0.2 and 1:0.5 ratios. The variances were $< 42\%$ (the largest for the amplitude, for the remaining parameters $< 10\%$). So for peak displacements larger than the FWHM of the peaks, the resolution was good also for the ratio of intensity of 20%. For a displacement smaller as the FWHM of the peak, the resolution was possible only for intensities close to each other. For a small displacement ($< 10\%$), the fit was impossible.

For the Gold deconvolution we also got a fit for a displacement of 10 channels and ratio 1:0.2 and a displacement of 5 channels and ratios 1:0.5, 1:0.2 with variances $< 27\%$. For a displacement of 1 channel the fitting procedure failed. So we can say that Gold's algorithm is much more able to distinguish a doublet and give us more precise results than the Van Cittert's algorithm.

In Fig. 4 convolutions of Gaussians and the results of the deconvolution procedure described above can be seen.

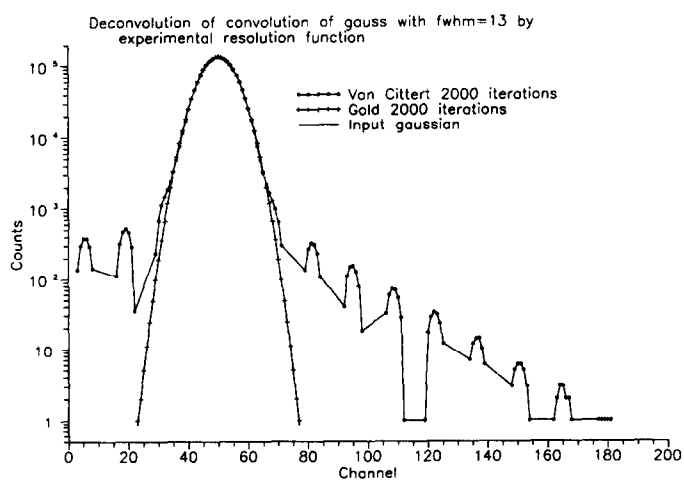

Fig. 3. Comparison of peak deconvoluted by Van Cittert and Gold method.

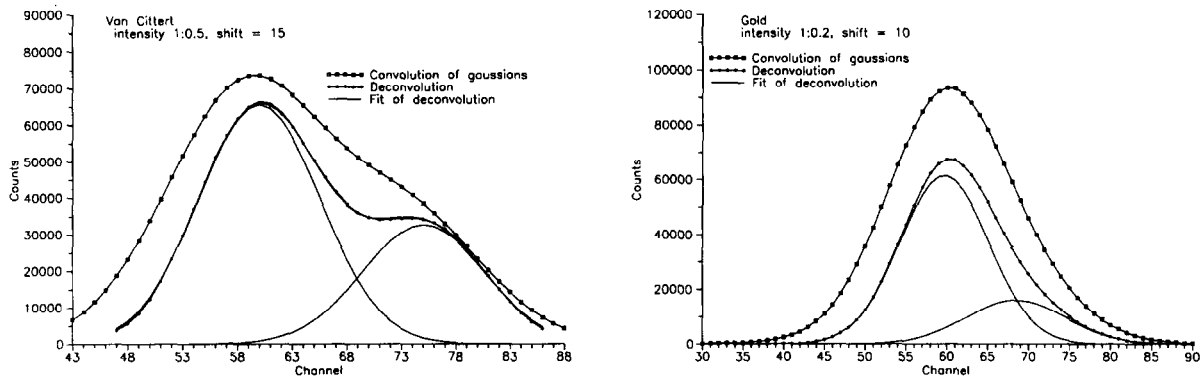

Fig. 4. Convolution and deconvolution of Gaussians.

4. Experimental spectra

All these studies of the convolution of Gaussians and following deconvolution were aimed at finding out the possibility and limitations of the deconvolution methods and their possible use in the deconvolution of experimental γ-ray spectra. Because the methods are quite reliable (especially Gold's method) we have used Gold's deconvolution method for our experimental spectra.

The effect of DBAL is quite smallscale and one needs a very stable measuring system for its measurement. We can get information about its stability from the 1274 keV peak, which is measured simultaneously with the 511 keV annihilation peak if we use ^{22}Na as a radioactive source.

In our experiment we used an HPGe detector with a resolution of 1.56 keV at 570 keV with a ^{207}Bi source. The energy calibration of the system was 163 eV/channel. The number of counts at the annihilation peak was ≈630 000 and the number of counts at the 1274 keV peak was ≈180 000. In Fig. 5 we can see the S-parameter with physical information about the momentum distribution of the electrons and the S-parameter of the 1274 keV peak, which carries information about the stability of the measuring system. If there was no instability during the measurement, it should be a constant. As we can see, the S-parameter of the 1274 keV peak was not constant, so the measuring system was not stable. So we do not know if the structure in the S-parameter of 511 keV peak was really a consequence of this instability or if it had a real physical meaning. Of course, there is a possibility to try to use the S-parameter of the 1274 keV peak as a correction on the instability, e.g. by calculating the ratio $R = S(511\text{ keV})/S(1274\text{ keV})$. The result of such procedure is shown in Fig. 5. As we can see, this simple procedure does not suppress the effect of instability. Because we are

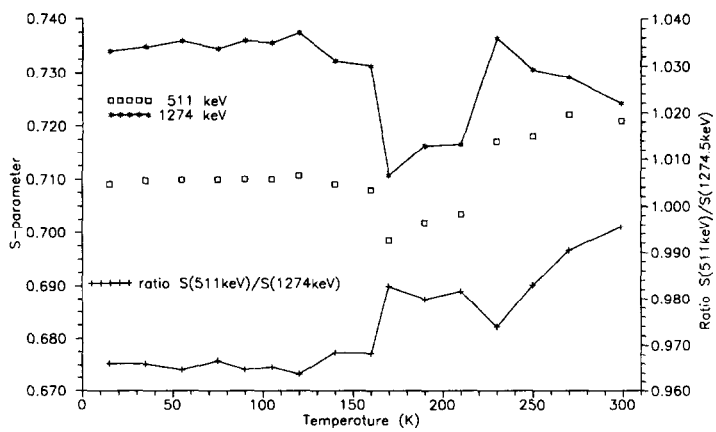

Fig. 5. S-parameters without the deconvolution procedure and the ratio R of the S-parameter at 511 keV to the S-parameter at 1274 keV.

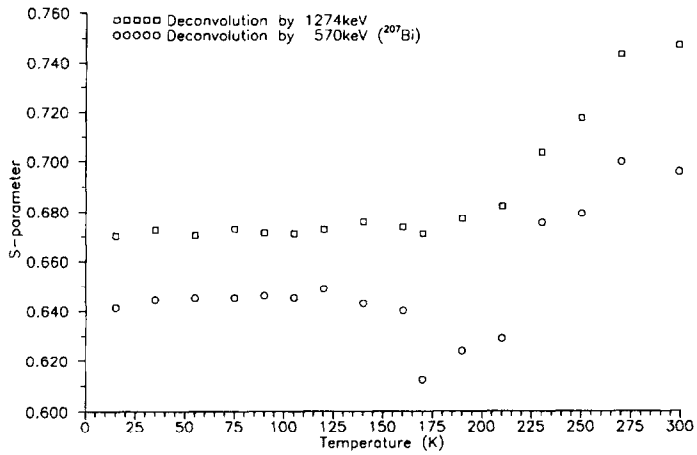

Fig. 6. *S*-parameters after deconvolution.

interested only in the relative changes of the *S*-parameter on temperature, we can make a deconvolution of the 511 keV peak with the 1274 keV peak and try to suppress the effect of instability.

Fig. 6 shows the result of these deconvolutions (the *S*-parameter after deconvolution) along with the *S*-parameter from deconvolutions of the 511 keV peak with the 570 keV peak (^{207}Bi), which was measured at the beginning of our experiment. So we can see that the structure at the temperature of 175 K was only a consequence of instability of the measuring system, because it disappeared after the deconvolution of the 511 keV peaks with the 1274 keV peaks. We can obtain physical information from the dependence of the *S*-parameter on the temperature by using a deconvolution procedure.

This procedure can also be used for a reliable estimate of the electron momentum distribution. In such case we need a better resolution function (e.g. 514 keV from the decay of ^{85}Sr) measured simultaneously with the annihilation line.

5. Conclusion

It turned out that the described deconvolution methods are in principle convenient for processing experimental spectra from DBAL measurements. They help to sup press influence of instability of the measuring system. Particularly the Gold deconvolution method works very well and it does not deform the deconvolution result. The reproducibility of the deconvolution results tested on simulated convolutions of Gaussians is very good. Van Cittert's algorithm gives the results with oscillations from positive to negative values on the wings of the deconvoluted peaks. Gold's deconvolution gives only small non-negative oscillations. Unambiguously we can say that Gold's algorithm is more reliable with regard to the precision of the deconvolution. The time needed for the deconvolution is practically the same for both algorithms.

So, the use of Gold deconvolution is a cheap and simple way to eliminate the electronic instabilities which blur physical effects.

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