

The least-squares fitting programme POSITRONFIT: principles and formulas

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<p>Title and author(s)</p> <p>The Least-Squares Fitting Programme POSITRONFIT: Principles and Formulas by P. Kirkegaard and M. Eldrup</p>	<p>Date September 1971</p> <p>Department or group Reactor Physics Department</p> <p>Group's own registration number(s)</p>
<p>14 pages + 0 tables + 1 illustrations</p>	
<p>Abstract</p> <p>A computer programme is described that analyses positron lifetime spectra by a least-squares technique of the semi-linear Marquardt type. The model includes up to four lifetimes. Fixed values can be imposed on any number of lifetimes and relative intensities. Correction for the source annihilation can be made. The results include the variance-covariance matrix of the fitted parameters.</p>	<p>Copies to</p> <p>Standard distribution 125 copies</p>
<p>Available on request from the Library of the Danish Atomic Energy Commission (Atomenergikommissionens Bibliotek), Risø, Roskilde, Denmark. Telephone: (03) 35 51 01, ext. 334, telex: 5072.</p>	<p>Abstract to</p> <p>Standard distribution</p>

THE UNIVERSITY OF CHICAGO
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1. Introduction
2. Theoretical Framework
3. Methodology
4. Results
5. Discussion
6. Conclusion

The following table presents the results of the regression analysis. The dependent variable is the log of the number of votes received by the candidate. The independent variables are the candidate's name, the candidate's party, and the candidate's district. The results show that the candidate's name and party have a significant positive effect on the number of votes received, while the candidate's district has a significant negative effect.

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1. Introduction

In a measurement of positron lifetimes in an ordinary condensed material, a spectrum is obtained which is generally assumed to be composed of a number of exponential terms and a background. The computer programme POSITRONFIT was developed to perform the analysis necessary to extract lifetimes and their relative intensities from such spectra.

2. The Mathematical Model of the Spectrum

To obtain a mathematical representation of a measured spectrum we use the following two important assumptions:

- (a) In a measured spectrum the numbers in the channels fluctuate around a curve that is the sum of a constant background and a number of decaying exponential functions somewhat smeared on account of the finite time resolution. The smeared functions are determined by folding the exponentials with the resolution function of the measuring system. The fluctuations around this curve are distributed in accordance with Poisson statistics.
- (b) The resolution function of the system can be measured as the time spectrum for two γ -quanta emitted simultaneously (for instance from a Co^{60} -source). It is to a good approximation a Gaussian curve.

In addition, the following considerations were taken into account:

Since the spectra are normally recorded in the channels of a multi-channel analyser, they are not continuous curves, but the number in each channel is the average of the curve mentioned under (a) over the width of one channel.

Positrons annihilating in the source material contribute to a measured spectrum. A correction for this can be made by subtracting the lifetime spectrum of the source material from the measured spectrum, the area of the former being a proper fraction of that of the latter.

The ideal lifetime spectrum of a number of positrons annihilating at the rate $\lambda_j (= \tau_j^{-1}$, τ_j is the mean lifetime) is described by a decaying exponential function:

$$I_j(t) = I_{oj} \exp(-\lambda_j t) \quad t \geq 0$$

$$I_j(t) = 0, \quad t < 0$$

where t is the channel number (equivalent to time). I_{oj} is a constant.

If the measuring system has a time resolution function which is a Gaussian with the standard deviation σ and centred around channel No. T_o :

$$P(t) = \frac{1}{\sigma\sqrt{\pi}} \exp \left[-\frac{(t-T_o)^2}{\sigma^2} \right],$$

the ideal spectrum will be smeared as determined by the folding of I_j with P :

$$F_j(t) = \frac{I_{oj}}{\sigma\sqrt{\pi}} \int_0^{\infty} \exp(-\frac{(t-T_o-t')^2}{\sigma^2}) \exp(-\lambda_j t') dt'$$

Evaluating the convolution integral we have:

$$F_j(t) = \frac{1}{2} I_{oj} \left[\exp(-\lambda_j(t-T_o - (1/4)\lambda_j \sigma^2)) \right] \left[1 - \operatorname{erf}\left(\frac{1}{2} \lambda_j \sigma - \frac{t-T_o}{\sigma}\right) \right], \quad (1)$$

where $\operatorname{erf}(x)$ stands for the error function

$$\operatorname{erf}(x) = \frac{2}{\sqrt{\pi}} \int_0^x e^{-t^2} dt ;$$

σ is related to the Full Width at Half Maximum of the prompt curve by:

$$\text{FWHM} = 2 \sqrt{\ln 2} \sigma .$$

In order to find the number in channel No. i , $F_j(t)$ should, since the numbers recorded in the measured spectrum are averages over the width of one channel, be integrated over this width. The result is:

$$F_{j,i} = \frac{I_{oj}}{2\lambda_j} \left[Y_{j,i} - Y_{j,i+1} - \operatorname{erf}\left(\frac{t_i - T_o}{\sigma}\right) + \operatorname{erf}\left(\frac{t_{i+1} - T_o}{\sigma}\right) \right], \quad (2)$$

$$\text{with } Y_{j,i} = \exp \left[-\lambda_j(t_i - T_o) + (1/4)\lambda_j^2 \sigma^2 \right] \left[1 - \operatorname{erf}\left(\frac{1}{2} \lambda_j \sigma - \frac{t_i - T_o}{\sigma}\right) \right]$$

where t_i is the value of the variable t at the common limit of channels Nos. $i-1$ and i .

So the mathematical expression taken to describe a measured spectrum with k_o lifetimes and the background B is:

$$f_i = B + \sum_{j=1}^{k_0} F_{j,i} \quad (3)$$

The source correction can be made by subtracting from the measured spectrum a source correction spectrum given by:

$$N_i^s = C \sum_{j=1}^{k_s} F_{j,i}^s \quad (4)$$

This has k_s terms of the type (2) with $\frac{I_{oj}}{\lambda_j}$ replaced by $\frac{I_{oj}^s}{\lambda_j^s}$, where $(\lambda_j^s)^{-1}$ is the lifetime of the j 'th component of the lifetime spectrum of the source material, and $\frac{I_{oj}^s}{\lambda_j^s}$ its intensity. The normalization constant C in (4) can be written

$$C = \alpha \left[\sum_{j=1}^{k_0} \frac{I_{oj}}{\lambda_j} \right] \left[\sum_{j=1}^{k_s} \frac{I_{oj}^s}{\lambda_j^s} \right]^{-1}, \quad (5)$$

α being the fraction of source annihilations to the total number of annihilations.

The parameters contained in our model (3) are the annihilation rates, λ_j , channel number equivalent to time equal zero, T_0 , intensities, $\frac{I_{oj}}{\lambda_j}$, background, B , and the σ of the resolution function. It is natural to include all these, except perhaps σ and T_0 , in the list of fitting parameters. From a measured prompt curve can be determined its width, as given by σ , and the channel number at the position of its peak, T_0 (representing time equal zero). It is reasonable to consider σ as fixed. However, it was found that the result of the analysis was very sensitive to the value used for T_0 . Furthermore, an exact experimental determination of T_0 is very difficult, and in addition it is subject to changes caused by drift in the electronic equipment and changes in the position of the sample. Therefore it was included as one of the fitting parameters.

3. The Least-Squares Technique

We are now able to formulate the criterion for the fitting of our model to the measured spectrum.

Let the part of the spectrum that is used in the analysis contain n count rates y_i , i being the channel number.

According to the least-squares principle, we must find a parameter vector

$$b = (a_1, \dots, a_{k_0}, B, \lambda_1, \dots, \lambda_{k_0}, T_0)^T \quad (6)$$

(T stands for transpose, and $a_j = \frac{I_{0j}}{2\lambda_j}$ are the half-intensities) so that

$$\Phi = \sum_{i=1}^n w_i (y_i - f_i(b))^2 = \min \quad (7)$$

where $f_i(b)$ is the count rate in channel No. i predicted by the model given by (3) for the parameter vector b , and w_i are fixed weights of the data points.

The components of (6) may vary without any restrictions, or, alternatively, constraints of different types can be imposed on them. In this work permissible constraints will be of the following two types:

(a) A fixed value is assigned to one or more of the λ . (8)

(b) m linear combinations of the a are zero ($m < k_0$). (9)

Constraints on the relative intensities,

$$\frac{a_{j_1}}{a_1 + \dots + a_{k_0}} = q_1 = \text{constant} \quad (l = 1, \dots, m) \quad (10)$$

are seen to be special cases of (9).

Constraints of type (a) are realized simply by deleting the fixed λ from the parameter list.

Model (3) is linear in intensities and background, but nonlinear in λ_j and T_0 . The nonlinearity calls for an iterative method of finding the least-squares estimate of the parameters: In POSITRONFIT, Marquardt's technique is used¹⁾; it combines in an almost optimal fashion the method of Gauss - Newton and the method of steepest descent. Contrary to most other

fitting programmes, POSITRONFIT takes advantage of the fact that the model is only partially nonlinear. According to this, the iterations take place in the subspace of the nonlinear parameters, and conditional solutions for the linear parameters are calculated after each iteration. This method results as a rule in a considerable saving of iterations at the expense of a slight increase in the cost per iteration. The iterations start from a guessed initial set of λ_j and T_0 and are terminated when Φ has proved to be stationary. A detailed description of these ideas is given in an earlier report²⁾ that is the basis of the subsequent analysis.

First, we write down the decomposition of the parameter vector b in its linear and nonlinear components,

$$b = \begin{bmatrix} \alpha \\ \beta \end{bmatrix} \quad (11)$$

$$\text{with } \alpha = (\alpha_1, \dots, \alpha_{k_0}, B)^T \quad (12)$$

$$\text{and } \beta = (\lambda_1, \dots, \lambda_{k_V}, T_0)^T \quad (13)$$

so that the model $f_i(b)$ is linear in α and nonlinear in β . k_V in (13) is the number of free lifetimes ($k_V = k_0 - k_f$, where k_f is the number of fixed lifetimes according to type (a) constraints). The dimension of α is $k_\alpha = k_0 + 1$ and that of β is $k_\beta = k_V + 1$.

If we define $\alpha_{k_\alpha} = B$, $u_{i, k_\alpha} = 1$, and u_{ij} = the quantity in square braces in (2) for $j \leq k_0$, we have

$$f_i = \sum_{j=1}^{k_\alpha} \alpha_j u_{ij} \quad (14)$$

(cf. sec. 4 in ref. 2).

To set up eqs. (39) and (43) in ref. 2, we need the following list of matrices and vectors,

$C, K, \alpha, \mu, \gamma, \delta, A', M', E.$

C and γ are defined in (40), ref. 2. $\alpha = \alpha(\beta)$ is the solution of a conditioned on β ²⁾, i. e. the α -vector (cf. 12)) that for a fixed β (cf. (13)) minimizes Φ (cf. (7)) under the possible constraints (8), (9), (10). The matrix

K contains the k_0 coefficients of each of the m constraints of type (9). In the special case (10) the coefficients of K turn out to be

$$\left. \begin{aligned} k_{ij} &= \delta_{jj_1} - q_1, \quad j = 1, \dots, k_0 \\ k_{1k_a} &= 0 \end{aligned} \right\} \quad l = 1, \dots, m \quad (15)$$

(δ_{ij} stands for the Kronecker delta). The matrix δ is (both for (9) and (10)) the zero vector, $\delta = \theta$. μ is the vector of Lagrange multipliers²). A' and M' are given in (44) and (45), ref. 2. E is given in (46), ref. 2, and the use of (2) and (3) (this report) gives

$$\left. \begin{aligned} e_{jj'} &= \sum_i w_i \left[\delta_{jj'} v_{ij'} (y_i - f_i) - u_{ij} \sum_{j_1=1}^{k_a} a_{j_1} \delta_{j_1 j'} v_{ij'} \right] \\ &\quad (j \leq k_a, j' \leq k_v) \\ e_{j, k_\beta} &= \sum_i w_i \left[q_{ij} (y_i - f_i) - u_{ij} \sum_{j_1=1}^{k_a} a_{j_1} q_{ij_1} \right] \quad (j \leq k_a) \end{aligned} \right\} \quad (16)$$

Here, $v_{ij} = \frac{\partial u_{ij}}{\partial \lambda_j}$ and $q_{ij} = \frac{\partial u_{ij}}{\partial T_0}$. We find

$$\begin{aligned} v_{ij} &= \left(\frac{1}{2} \sigma^2 \lambda_j - (t_i - T_0) \right) Y_{j,i} - \frac{\sigma}{\sqrt{\pi}} \exp \left(- \left(\frac{t_i - T_0}{\sigma} \right)^2 \right) \\ &\quad - \left(\frac{1}{2} \sigma^2 \lambda_j - (t_{i+1} - T_0) \right) Y_{j,i+1} + \frac{\sigma}{\sqrt{\pi}} \exp \left(- \left(\frac{t_{i+1} - T_0}{\sigma} \right)^2 \right) \end{aligned} \quad (j \leq k_0) \quad (17)$$

$$q_{ij} = \lambda_j (Y_{j,i} - Y_{j,i+1}) \quad (j \leq k_0) \quad (18)$$

In particular, (16) gives for $j' < k_\beta$ and $j' \neq j$

$$e_{jj'} = -a_{j'} \sum_i w_i u_{ij} v_{ij'} \quad (j \leq k_a, j' \leq k_v, j \neq j') \quad (19)$$

and for $j = j'$

$$e_{jj} = \sum_i w_i v_{ij} [y_i - f_i - a_j u_{ij}] \quad (j \leq k_v) \quad (20)$$

Now we consider the iterations in the space of $\beta = \begin{bmatrix} \lambda \\ T_o \end{bmatrix}$, where $\lambda = \begin{bmatrix} \lambda_1 \\ \vdots \\ \lambda_{k_v} \end{bmatrix}$,

as described in ref. 2, sec. 4. We need the derivative matrix with elements $\frac{\partial f_i}{\partial \beta_{j'}}$. These are given in (41), ref. 2; they contain $\frac{\partial u_{ij}}{\partial \beta_{j'}}$, and we find

$$\frac{\partial u_{ij}}{\partial \lambda_{j'}} = \delta_{jj'} v_{ij'} \quad (j \leq k_a, j' \leq k_v) \quad (21)$$

$$\frac{\partial u_{ij}}{\partial T_o} = \begin{cases} a_{ij} & (j \leq k_o) \\ 0 & (j = k_a) \end{cases} \quad (22)$$

The iteration procedure follows exactly the description in ref. 2; it has converged when we have obtained a stationary Φ and a stationary parameter vector

$$b = \begin{bmatrix} a \\ \beta \end{bmatrix} = \begin{bmatrix} a_1 \\ \vdots \\ a_{k_o} \\ B \\ \lambda_1 \\ \vdots \\ \lambda_{k_v} \\ T_o \end{bmatrix} \quad (23)$$

which is the solution of our problem.

The procedure followed for analysis of a spectrum with source correction is as follows: First the recorded spectrum is fitted with the desired number of exponential terms and constraints, and the parameters for this spectrum are determined. With T_o from this procedure, the source cor-

rection spectrum is generated, formula (4) being used, and is subtracted from the measured spectrum. The corrected spectrum is then fitted, the procedure starting from the parameters determined from the first iteration cycle.

The final results of the analysis are estimates for the corrected spectrum of lifetimes and relative intensities as well as background and channel number equivalent to time equal zero. Further, statistical estimates, including in particular estimated standard deviations of the parameters, are given according to the principles discussed in the following section.

4. Statistical Analysis

In ref. 2 is given a statistical analysis of the least-squares problem with constraints of the present type. This analysis assumes

- (i) an ideal model²⁾
- (ii) small fluctuations of the data ordinates (count numbers) around their means
- (iii) "statistical weighting".

(i) is often well justified if we have made a clever choice of the number of lifetime components, k_0 , and of the possible constraints.

(ii) is justified if the total number of counts in the spectrum is sufficiently large; this was hardly the case for many of the analysed spectra.

(iii) concerns the choice of the weights, w_i , of the data points in expression (7) of Φ . These have hitherto been regarded as arbitrary coefficients, but several advantages will be gained²⁾ if we choose the so-called statistical weighting,

$$w_i = \frac{1}{\sigma_i^2} \tag{24}$$

where σ_i^2 is the variance of the i 'th data ordinate y_i . As y_i has a Poisson distribution with mean and variance η_i , we should choose $w_i = \frac{1}{\eta_i}$. However, as η_i is not known in advance, we use instead

$$w_i = \frac{1}{y_i} . \tag{25}$$

In cases of spectra with source correction, (25) is used in the first iter-

ation cycle. but is replaced by $w_i = \frac{1}{f_i}$ (f_i = ordinate predicted by the model without source correction) in the second cycle. This procedure improves the estimate of the background B (it can be shown that the effect of using (25) is to underestimate η_i approximately by 1).

With the assumptions (i), (ii) and (iii) above it is shown²⁾ that the solution vector b (cf. (19)) is a statistical variable with the var-cov matrix Q that is the minor of the matrix H^{-1} in (15), ref. 2. To find Q we must set up the matrix of system (10), ref. 2. The matrices P , W , R are defined in (11), ref. 2 (note that P in this connection means the derivative matrix with respect to all $k = k_\alpha + k_\beta$ parameters, in contrast to the P for the β -iterations). A column in P has the form

$$\begin{bmatrix} \frac{\partial f_i}{\partial b_1} \\ \vdots \\ \frac{\partial f_i}{\partial b_k} \end{bmatrix} = \begin{bmatrix} u_{i1} \\ \vdots \\ u_{ik_\alpha} \\ a_1 v_{i1} \\ \vdots \\ a_{k_\beta} v_{ik_\beta} \\ \frac{\partial f_i}{\partial T_0} \end{bmatrix} \quad (26)$$

$$\text{with } \frac{\partial f_i}{\partial T_0} = \sum_{j_1=1}^{k_0} a_{j_1} a_{ij_1} + \sum_{j_1=1}^{k_\alpha} u_{ij_1} \frac{\partial a_{j_1}}{\partial T_0} \quad (27)$$

where $\frac{\partial a_{j_1}}{\partial T_0}$ form the last column in A' in (43), ref. 2.

R is an extension of K (cf. (14)):

$$R = \begin{bmatrix} K & \vdots & \theta \\ & & k_\beta \end{bmatrix} \quad m \quad (28)$$

Now, we want to state the statistical results in terms of the lifetimes, τ_j , rather than λ_j , and relative intensities, i_j , rather than a_j . The connection is given by

$$\tau_j = \frac{c}{\lambda_j} \quad (29)$$

$$\text{and } i_j = \frac{a_j}{a_1 + \dots + a_{k_0}} \quad (j \neq k_0) \quad (30)$$

(the scale factor c in (29) occurs because it is practical to express $\frac{1}{\lambda_j}$ in units of a channel width and τ_j in nsec). The new parameter vector

$$b_1 = \begin{bmatrix} i_1 \\ \cdot \\ \cdot \\ \cdot \\ i_{k_0} \\ B \\ \tau_1 \\ \cdot \\ \cdot \\ \cdot \\ \tau_{k_0} \\ T_0 \end{bmatrix} \quad (31)$$

has a var-cov matrix Q_1 given by ³⁾

$$Q_1 = J Q J^T \quad (32)$$

where J is the Jacobian of the functional transformation $b \rightarrow b_1$. We find

our model is no good representation of the measured spectrum.

5. Description of POSITRONFIT Input/Output

The POSITRONFIT code is written in FORTRAN for the Burroughs B6700 computer at Risø. As a detailed description of the code is published elsewhere⁴⁾, only a short summary of the standard input/output list will be given.

Apart from the spectrum to be analysed the list of input data is:

- (a) A figure to indicate whether the results of each iteration should be printed out, and whether a table of the fitted and experimental points together with a graphical representation of these points should be printed out.
- (b) The FWHM of the prompt curve in nsec.
- (c) The time equivalent to one MCA channel in nsec.
- (d) A guess of T_0 , the channel number corresponding to zero time.
- (e) The number of exponential terms to be used in the fit and the number of fixed lifetimes and relative intensities.
- (f) Guesses of the lifetimes to be determined (in nsec) together with the values of the fixed lifetimes (in nsec) and relative intensities (in per cent).
- (g) Finally the number of exponential terms (up to 4) in the source correction spectrum and their lifetimes (in nsec) and relative intensities (in per cent) together with the fraction (in per cent) of the area of the lifetime spectrum that the area of the correction spectrum shall amount to.

The output data are:

- (a) As determined by the input, the results of each iteration and tables and graphs of the measured and the fitted points that may be printed out.
- (b) The analysed spectrum and the guessed and fixed values of the

Certain special options exist⁴⁾ that are omitted in this list.

lifetimes and T_0 .

- (c) Identification number of the spectrum (in the version used, the number in the second channel of the spectrum, which can be determined manually by means of the multi-channel analyser).
- (d) The number of iterations used by the computer to make the iterations converge (i. e. find the minimum value of Φ).
- (e) The FWHM of the prompt curve, the time per channel, and the parameters determining the source correction as fixed by the input data.
- (f) The variance of the fit.
- (g) The fitted and fixed parameters (lifetimes, relative intensities, background, and T_0), all with standard deviations.
- (h) The areas of the fitted curve (calculated as the sum of the absolute intensities) and the measured one (calculated by summing the numbers in all channels), the extent of agreement between which, together with the variance of the fit, show the goodness of the fit.
- (i) The variance, the parameters and the areas for the fit before source correction printed out for comparison.
- (j) Finally a matrix showing the total correlations between the fitted parameters is printed out.

In fig. 1 is shown the most important part of a typical output with one intensity constraint.

6. References

- 1) D. W. Marquardt, An Algorithm for Least-Squares Estimation of Nonlinear Parameters, J. SIAM 11, No. 2 (1963) 431-441.
- 2) P. Kirkegaard, Some Aspects of the General Least-Squares Problem for Data Fitting, Risø-M-1399 (1971).
- 3) H. Cramer, Mathematical Methods of Statistics (Princeton, University Press, Princeton, N. J., 1946).
- 4) P. Kirkegaard and M. Eldrup, POSITRONFIT: A Versatile Program for Analysing Positron Lifetime Spectra. Submitted to Computer Physics Communications, 1971.

CONVERGENCE OBTAINED AFTER 8 + 5 ITERATIONS

PARAMETERS FOR SPECTRUM NO 1254

MONOCRYSTALLINE ICE =182 DEGREES C

TIME SCALE = 0.070 NSEC/CHANNEL FWHM = 0.425 NSEC
NUMBER OF TERMS = 3 NUMBER OF LIFETIME CONSTRAINTS = 0

NUMBER OF CONSTRAINTS FOR RELATIVE INTENSITIES = 1
TERM INDICES FOR INTENSITY CONSTRAINTS 3
FIXED RELATIVE INTENSITIES 52.00

SOURCE CORRECTION 7.0 PCT
SOURCE LIFETIMES IN NSEC 0.342 1.150
SOURCE INTENSITIES IN PCT 90.00 11.00

VARIANCE OF THE FIT = 0.987
ITS DISTRIBUTION SHOULD BE APPROXIMATELY NORMAL (1.0,072)
EXCESS PROBABILITY = 57.27 PCT

LIFETIMES IN NSEC	0.124	0.440	0.657
STANDARD DEVIATIONS	0.015	0.026	0.007
RELATIVE INTENSITIES IN PCT	20.36	27.64	52.00
STANDARD DEVIATIONS	1.80	1.80	0.00
BACKGROUND	70.36		
STANDARD DEVIATION	0.47		

TIME=ZERO CHANNEL NUMBER 127.628
STANDARD DEVIATION 0.037

AREA CHECK AREA FROM FIT = 0.34152E 06
AREA FROM TABLE = 0.34188E 06

MATRIX OF TOTAL CORRELATIONS
(RELATIVE INTENSITIES, BACKGROUND, FREE LIFETIMES, T0)

1.000	=1.000	=0.000	0.110	0.871	0.952	=0.746	=0.530
=1.000	1.000	=0.000	=0.110	=0.871	=0.952	0.746	0.530
0.000	=0.000	0.000	0.000	=0.000	=0.000	0.000	0.000
0.110	=0.110	0.000	1.000	0.075	0.141	=0.206	=0.034
0.871	=0.871	=0.000	0.075	1.000	0.825	=0.554	=0.851
0.952	=0.952	=0.000	0.141	0.825	1.000	=0.874	=0.546
=0.746	0.746	0.000	=0.206	=0.554	=0.874	1.000	0.298
=0.530	0.530	0.000	=0.034	=0.851	=0.546	0.298	1.000

EXECUTION TIME = 76.62 SEC.

Fig. 1. The most important part of a typical output from a fitting with three terms and one intensity constraint.