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DETERMINISTIC REGULARIZATION METHOD FOR THE PROBLEM OF BACKGROUND LINE IDENTIFICATION IN EXPERIMENTAL DATA ANALYSIS

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Tikhonov's regularization method is applied to develop an efficient algorithm and the corresponding programs for identifying a smooth background line. The algorithm can also be used to construct the filtering function. Some applications are examined.

The problem of background line identification often arises in primary analysis of physical experimental data, when the background component has to be extracted from the raw observations. This is primarily associated with the analysis of diffraction spectra in neutronography, Auger spectroscopy, gas electronography, and with the analysis of Debye powder diagrams.

Random factors are generally responsible for the creation of the background line, and it is not always known what physical factors are actually responsible for its appearance. The key issue in background line identification is therefore how to choose the modeling functions. The main properties of these functions are smoothness and boundedness of the curvature function. The curvature function is defined as

$$k(u(x)) = \frac{\left|u''(x)\right|}{\left(1+(u'(x))^2\right)^{3/2}}.$$

From physical considerations we seek the background line B so as to minimize the norm of the curvature function. An obvious relationship holds for any norm: $\|k(u)\| \le \|u''\|$. Thus, by minimizing the norm $\|u''\|$ on some special set of functions we also minimize the norm $\|k(u)\|$.

Let us formalize our problem. Consider the space of functions square-integrable on $[a, b]$ with the metric

$$\rho_{U_2}(u,v) = \left\{ \int_a^b (u(x)-v(x))^2 dx \right\}^{1/2}$$

(in what follows we denote this metric simply by $\rho(u,v)$). This is the metric space $U_2[a,b]$. For $u, v \in U_2[a,b]$:

$$(u,v) = \int_a^b uv dx, \qquad \|u\|_{U_2}^2 = (u,u)$$

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(in what follows $\| \ \|$ is the norm $\| \ \|_{U_2}$). On the set $D = \left\{ u \in C^4 \left[a, b \right];\ u(a) = u'(a) = u(b) = u'(b) = 0 \right\}$ we consider the linear differentiation operators

$$L_1 u = \frac{du}{dx}, \qquad L_2 u = \frac{d^2 u}{dx^2}.$$

Assume that we have some $\delta > 0$ such that the given experimental function $u_\delta(x) \in C\left[a, b \right]$ satisfies $\rho(u_\delta, B) \le \delta$. Also assume that we have some number $R > 0$ such that $\| L_2 B \| \le R$. In practice, the values of δ, R are easy to find from experimental data. Introduce the set

$$D_{\delta,R} = \left\{ u \in C^4 \left[a, b \right]; \rho(u_\delta, u) \le \delta,\ \| L_2 u \| \le R \right\}.$$

Without loss of generality we may assume that $D_{\delta,R} \subseteq D$ (this can be achieved by a linear change of variables).

The background line identification problem is posed as the search for an element of the set $D_{\delta,R}$ with a minimum squared norm of the second derivative, i.e.,

$$\| L_2 u \|^2 \to \min, \qquad u \in D_{\delta,R}. \tag{1}$$

The solution of this problem exists, is unique ([1], pp. 133–139), and reduces to finding a Tikhonov regularized solution. The prior information about δ, R enables us to avoid determining the regularization parameter α. By the deterministic regularization method [2, p. 136] the solution of (1) is sought by minimizing a quadratic functional on the set $D_{\delta,R}$:

$$\min \Psi\left[u \right] = \| u - u_\delta \|^2 + \frac{\delta^2}{R^2} \| L_2 u \|^2, \qquad u \in D_{\delta,R}. \tag{2}$$

Let u_α $(\alpha = \delta^2 / R^2)$ be the solution of (2). Then it necessarily satisfies the Euler identity

$$(u_\alpha - u_\delta, v) + \alpha(L_2 u_\alpha, L_2 v) \equiv 0 \qquad \forall v \in D_{\delta,R}.$$

Noting that the operator $L_2 = L_2^*$ is self-conjugate for the set D, we obtain the Euler equation

$$\alpha u_\alpha^{(4)} + u_\alpha = u_\delta, \qquad u_\alpha(a) = u'_\alpha(a) = u_\alpha(b) = u'_\alpha(b) = 0. \tag{3}$$

The solution of (3) exists and is unique for any continuous functions u_δ [3, p. 117].

We consider four solution methods for problem (2), (3):

(A) by discretizing (2) on a nonuniform grid;

(B) by discretizing (2) on a uniform grid;

(C) by discretizing (3) on a uniform grid;

(D) by finding the solution of (3) at every point [a, b].

Method A

The functional (2) is discretized with a nonuniform grid in the argument $x : x_0, x_1, ..., x_n (x_0 = a, x_n = b)$, $u_{\delta,i} = u_\delta(x_i)$ and the sought values $u_i = u(x_i)$. The functional (2) reduces to a quadratic form in the variables $u_0, u_1, ..., u_n$:

$$\Phi(\bar{u}) = \sum_{i=0}^{n} h_i (u_i - u_{\delta,i})^2 + \alpha \sum_{i=1}^{n-1} h_i ((u_{i-1} - 2u_i + u_{i+1}) / h_i^2)^2 \to \min .$$

The (unique) minimum of this quadratic form is determined as the solution of the linear algebraic system $A\bar{u} = \bar{f}$, where $\bar{u} = (u_0, u_1, ..., u_n)^T$, $f = (h_0 u_{\delta,0}, h_1 u_{\delta,1}, ..., h_n u_{\delta,n})^T$, A is a symmetrical positive-definite matrix. Its upper triangular part has the form

$$A = \begin{pmatrix} h_0 + \alpha h_1^{-3} & -2\alpha h_1^{-3} & \alpha h_1^{-3} & 0 & \dots & 0 \\ \dots & \dots & \dots & \dots & \dots & \dots \\ & h_j + \alpha(h_{j-1}^{-3} + 4h_j^{-3} + h_{j+1}^{-3}) & -2\alpha(h_j^{-3} + h_{j+1}^{-3}) & \alpha h_{j+1}^{-3} & & 0 \\ \dots & \dots & \dots & \dots & \dots & \dots \\ & & & & h_{n-1} + \alpha(h_{n-2}^{-3} + 4h_{n-1}^{-3}) & -2\alpha h_{n-1}^{-3} \\ & & & & & h_n + \alpha h_{n-1}^{-3} \end{pmatrix}.$$

The linear algebraic system $A\bar{u} = f$ is solved by monotone sweeping [4, p. 98], which requires a total of $8n - 5$ additions and subtractions, $8n - 5$ multiplications, and $3n$ divisions. Sufficient conditions for stability of this sweeping method for the matrix A can be written as

$$\alpha \le \min\left\{\frac{1}{2} h_0 h_1^3, \frac{1}{3} h_1 h_2^3, \frac{1}{2} h_n h_{n-1}^3, \frac{1}{3} h_{n-1} h_{n-2}^3, \frac{1}{2} \sigma_{\min}\right\},$$

where

$$\sigma_{\min} = \min_{2 \le i \le n-2} \frac{h_i}{h_{i-1}^{-3} + h_{i+1}^{-3}}$$

[4, p. 100].

Method B

The functional (2) is discretized with a uniform grid in the argument $x: x_i = x_0 + ih,\ i = 0,...,n$. In this case also the functional (2) reduces to a quadratic form in the variables $u_0, u_1, ..., u_n$:

$$\Phi(\overline{u}) = \sum_{i=0}^{n} (u_i - u_{\delta,i})^2 + \alpha h^{-3} \sum_{i=1}^{n-1} (u_{i-1} - 2u_i + u_{i+1})^2 \rightarrow \min.$$

The minimum is obtained by solving the linear algebraic system $Au = u_\delta$ with the symmetrical positive definite matrix $A = E + (\alpha / h^3)D$, where the singular matrix $D = D^T$ is

$$D = \begin{pmatrix} 1 & -2 & 1 & 0 & 0 & \dots & 0 \\ -2 & 5 & -4 & 1 & 0 & \dots & 0 \\ 1 & -4 & 6 & -4 & 1 & \dots & \\ \dots & \dots & \dots & \dots & \dots & \dots & \dots \\ & & & 1 & -4 & 5 & -2 \\ & & & 0 & 1 & -2 & 1 \end{pmatrix}.$$

For the given matrix A we have an efficient inversion method (by sweeping) which requires an order of $9n$ multiplications and n divisions.

Method C

The Euler equation (3) is discretized on a uniform grid. The fourth-order differentiation operator is replaced with the corresponding fourth-order finite difference operator:

$$u_i^{(4)} \approx \frac{1}{h^2}(u_{i-2} - 4u_{i-1} + 6u_i - 4u_{i+1} + u_{i+2}), \qquad i = 0,...,n-1.$$

Noting the zero boundary conditions, we may treat our function as periodic (with period $T = b - a$) and take $u_{-1} = u_{n-1}$, $u_{-2} = u_{n-2}$, $u_{n+1} = u_1$, $u_{n+2} = u_2$. Then

$$u_0^{(4)} \approx \frac{1}{h^4}(u_{n-2} - 4u_{n-1} + 6u_0 - 4u_1 + u_2),$$

$$u_1^{(4)} \approx \frac{1}{h^4}(u_{n-1} - 4u_0 + 6u_1 - 4u_2 + u_3),$$

$$u_{n-2}^{(4)} \approx \frac{1}{h^4}(u_{n-4} - 4u_{n-3} + 6u_{n-2} - 4u_{n-1} + u_0),$$

$$u_{n-1}^{(4)} \approx \frac{1}{h^4}(u_{n-3} - 4u_{n-2} + 6u_{n-1} - 4u_0 + u_1).$$

We thus obtain the linear algebraic system $(E + pC)u = u_\delta$, $p = \alpha / h^4$ where $(E + pC)$ is a symmetrical circulant:

$$E + pC = \begin{pmatrix} 1+6p & -4p & p & 0 & 0 & \ldots & 0 & p & -4p \\ -4p & 1+6p & -4p & p & 0 & \ldots & \ldots & 0 & p \\ p & -4p & 1+6p & -4p & p & \ldots & \ldots & \ldots & 0 \\ \ldots & \ldots & \ldots & \ldots & \ldots & \ldots & \ldots & \ldots & \ldots \\ 0 & \ldots & \ldots & 0 & p & -4p & 1+6p & -4p & p \\ p & 0 & \ldots & \ldots & 0 & p & -4p & 1+6p & -4p \\ -4p & p & \ldots & \ldots & \ldots & 0 & p & -4p & 1+6p \end{pmatrix}.$$

Thus, the linear algebraic system can be solved without inverting the matrix and the solution can be obtained in explicit form (if the matrix is non-singular), because we know the real eigenvalues $y_k = 1 + p(6 - 4r_k + r_k^2 + r_k^{n-2} - 4r_k^{n-1})$ and the eigenvectors $x_k = (1, r_k, r_k^2, ..., r_k^{n-1})^T$, where $r_k = \cos\frac{2\pi k}{n} + i\sin\frac{2\pi k}{n}$ (the kth root of 1). To find y_k we evaluate

$$6 - 4(r_k + r_k^{n-1}) + r_k^2 + r_k^{n-2} = 6 - 8\cos\alpha_k + 2\cos 2\alpha_k = 16\sin^4\frac{\alpha_k}{2}, \quad \alpha_k = \frac{2\pi k}{n}.$$

Thus, $y_k = 1 + 16p\sin^4\frac{\pi k}{n} > 0$ (hence the matrix $E + pC$ is positive definite) and the solution is obtained as $u = \frac{1}{n}F^*\Lambda^{-1}Fu_\delta$, where $\Lambda^{-1} = \mathrm{diag}\left\{y_0^{-1}, y_0^{-2}, ..., y_{n-1}^{-1}\right\}$, F is the discrete Fourier transform matrix whose columns are the eigenvectors x_k.

We can also consider the following fourth-order finite difference operator [6, p. 234]:

$$u_i^{(2)} \approx \frac{1}{h^2}(\alpha_2 u_{i-2} + \alpha_1 u_{i-1} + \alpha_0 u_i + \alpha_1 u_{i+1} + \alpha_2 u_{i+2}),$$

$$u_i^{(4)} \approx \frac{1}{h^2}(\alpha_2 u_{i-2}^{(2)} + \alpha_1 u_{i-1}^{(2)} + \alpha_0 u_i^{(2)} + \alpha_1 u_{i+1}^{(2)} + \alpha_2 u_{i+2}^{(2)})$$

$$= \frac{1}{h^4}(\beta_4 u_{i-4} + \beta_3 u_{i-3} + \beta_2 u_{i-2} + \beta_1 u_{i-1} + \beta_0 u_i + \beta_1 u_{i+1} + \beta_2 u_{i+2} + \beta_3 u_{i+3} + \beta_4 u_{i+4}),$$

where $\alpha_0 = -60$, $\alpha_1 = 32$, $\alpha_2 = -2$. The values of β_k are expressed in terms of α_k as

$$\beta_0 = 2(\alpha_0^2 + \alpha_1^2 + \alpha_2^2), \quad \beta_1 = 2(\alpha_0\alpha_1 + \alpha_2\alpha_1), \quad \beta_2 = \alpha_1^2 + 2\alpha_2\alpha_0, \quad \beta_3 = 2\alpha_2\alpha_1, \quad \beta_4 = \alpha_2^2.$$

Taking $u_0 = u_n$, $u_{-1} = u_{n-1}$, $u_{-2} = u_{n-2}$, $u_{-3} = u_{n-3}$, $u_{-4} = u_{n-4}$, $u_{n+1} = u_1$, $u_{n+2} = u_2$, $u_{n+3} = u_3$, $u_{n+4} = u_4$, we obtain the eigenvalues

$$y_k = 1 + p(\beta_0 + \beta_1 r_k + \beta_2 r_k^2 + \beta_3 r_k^3 + \beta_4 r_k^4 + \beta_4 r_k^{n-4} + \beta_3 r_k^{n-3} + \beta_2 r_k^{n-2} + \beta_1 r_k^{n-1})$$

or

$$y_k = 1 + p(\beta_0 + 2(\beta_1 \cos\alpha_k + \beta_2 \cos 2\alpha_k + \beta_3 \cos 3\alpha_k + \beta_4 \cos 4\alpha_k)).$$

The solution is also written as $u = \frac{1}{n} F^* \Lambda^{-1} F u_\delta$. Only $\Lambda^{-1} = \mathrm{diag}\left\{ y_0^{-1}, y_0^{-2}, \ldots, y_{n-1}^{-1} \right\}$ changes. As we know, the eigenvectors $x_k = (1, r_k, r_k^2, \ldots, r_k^{n-1})^T$ are common for all circulants [7, p. 44].

Method D

Let us express the solution of (3) in explicit form. First we reduce the equation to

$$u^{(4)} + 4\lambda^4 u = 4\lambda^4 u_\delta, \qquad u(a) = u'(a) = u(b) = u'(b) = 0,$$

where $\lambda = (R/(2\delta))^{1/2}$. The solution of this equation is the function

$$u(x) = \sum_{k=1}^{4} c_k u_k(x) + 4\lambda^4 \int_a^x u_4(x-t) u_\delta(t)\,dt,$$

where

$$u_1(x) = \cosh\lambda x \cdot \cos\lambda x, \qquad u_2(x) = \frac{1}{2}\left(\cosh\lambda x \cdot \sin\lambda x + \sinh\lambda x \cdot \cos\lambda x\right),$$

$$u_3(x) = \frac{1}{2}\sinh\lambda x \cdot \sin\lambda x, \qquad u_4(x) = \frac{1}{4}\left(\cosh\lambda x \cdot \sin\lambda x - \sinh\lambda x \cdot \cos\lambda x\right).$$

The coefficients c_k are determined from the boundary conditions:

$$u_1(a)c_1 + u_2(a)c_2 + u_3(a)c_3 + u_4(a)c_4 = 0,$$

$$u_1(b)c_1 + u_2(b)c_2 + u_3(b)c_3 + u_4(b)c_4 = b_3,$$

$$-4u_4(a)c_1 + u_1(a)c_2 + u_2(a)c_3 + u_3(a)c_4 = 0,$$

$$-4u_4(b)c_1 + u_1(b)c_2 + u_2(b)c_3 + u_3(b)c_4 = b_4,$$

$$b_3 = -4\lambda^4 \int_a^b u_4(b-t)u_\delta(t)dt,$$

$$b_4 = -4\lambda^3 \left(\frac{d}{dx} \int_a^x u_4(x-t)u_\delta(t)dt \right)\Bigg|_{x=b}.$$

The proof is in [5].

Remark 1. The algorithm parameters δ and R are easily estimated by the moving mean method: let S_m be the m-point moving mean operator. Then we can take $\delta = \| S_m(u_\delta) - u_\delta \|$, $R = \| L_2 S_m(u_\delta) \|$.

Remark 2. If the sought function does not satisfy the zero boundary conditions (3), i.e., we have $u(a) = y_1$, $u'(a) = d_1$, $u(b) = y_2$, $u'(b) = d_2$ so that $y_1^2 + y_2^2 + d_1^2 + d_2^2 \neq 0$, then we introduce a new function $z(x) = u(x) - P_3(x)$, where

$$P_3(x) = \frac{1}{b-a}\left[(b-x)y_1 + (x-a)y_2 \right]$$

$$+ \frac{(x-a)(x-b)}{(b-a)^3}\left[(d_1(b-a) + y_1 - y_2)(x-b) + (d_2(b-a) + y_1 - y_2)(x-a) \right].$$

Then obviously $z(a) = z'(a) = z(b) = z'(b) = 0$, the function $u_\delta(x)$ is replaced with $u_\delta(x) - P_3(x)$, the value δ remains unchanged, but R changes to

$$R_z = \left(R^2 - \left(P_3'' P_3' - P_3''' P_3 \right)\Big|_a^b \right)^{1/2}.$$

Remark 3. The algorithm can be applied to filter noise from experimental data. Figure 1 shows the extraction of the useful signal by this algorithm.

Remark 4. Standard background line algorithms have difficulties with constructing the background line so that it lies strictly below the spectrum. Our algorithm successfully copes with this task (Fig. 2).

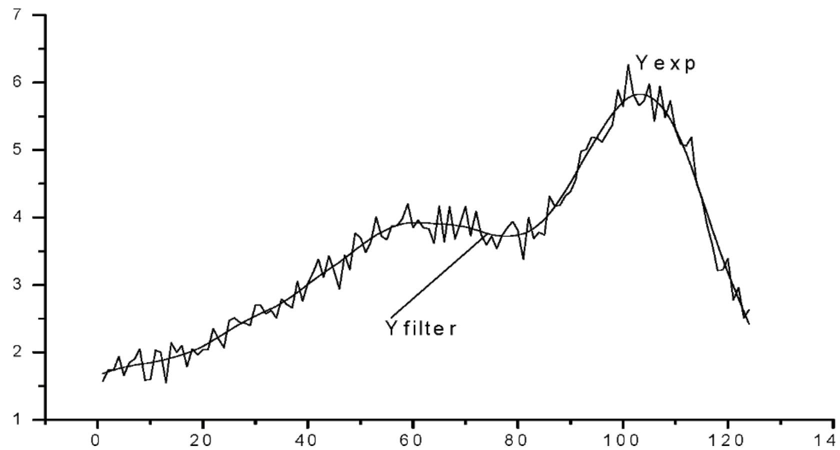

Fig. 1. Filtering noise from the experimental function.

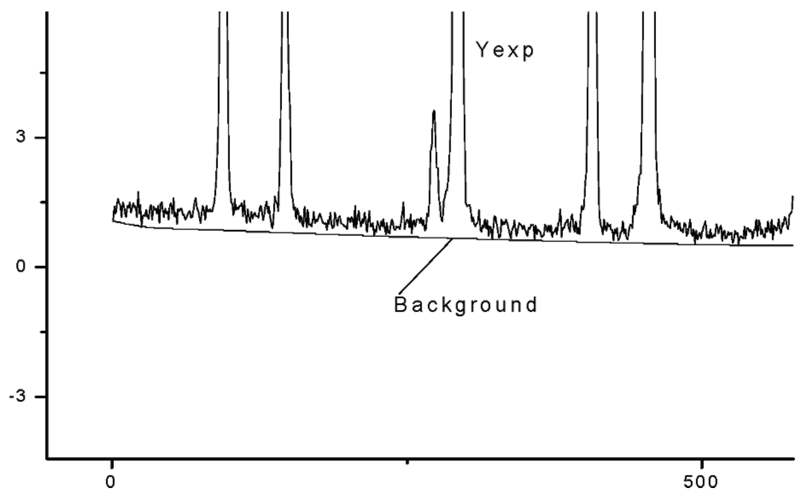

Fig. 2. The background line is constructed strictly below the spectrum.

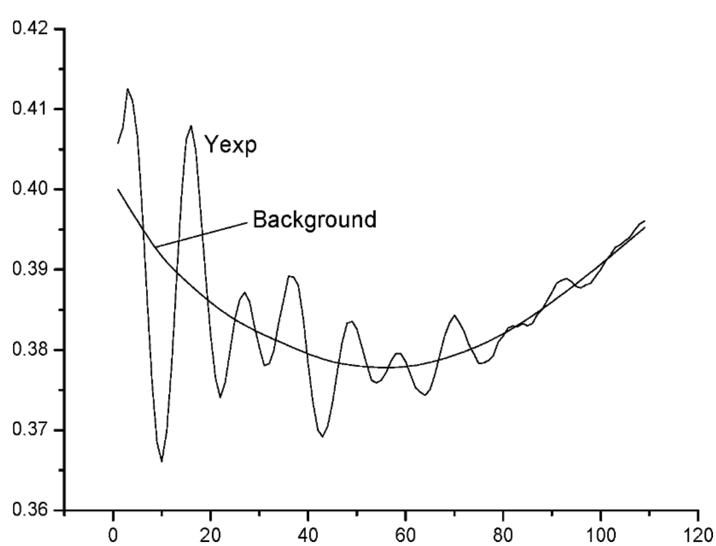

Fig. 3. Identifying the background line in gas electronography (Yexp is the electron scattering intensity by the gas molecule CHCl3).

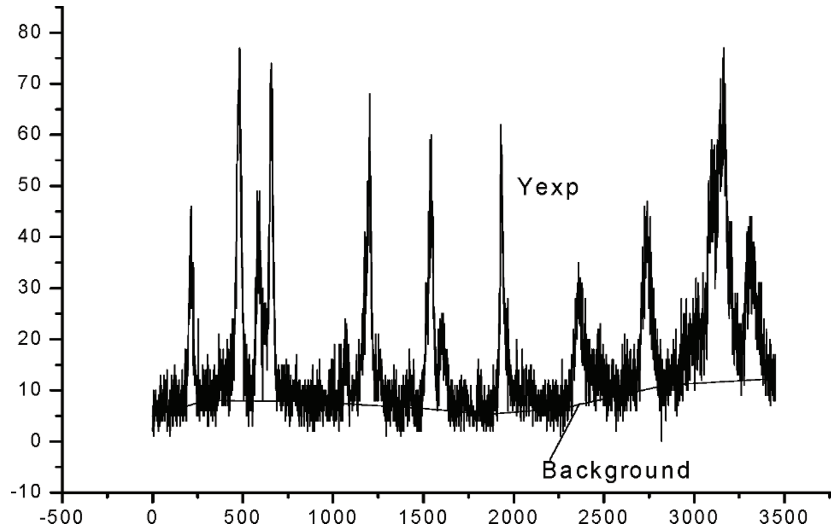

Fig. 4. Identifying the background line from an X-ray powder graph (3,500 points).

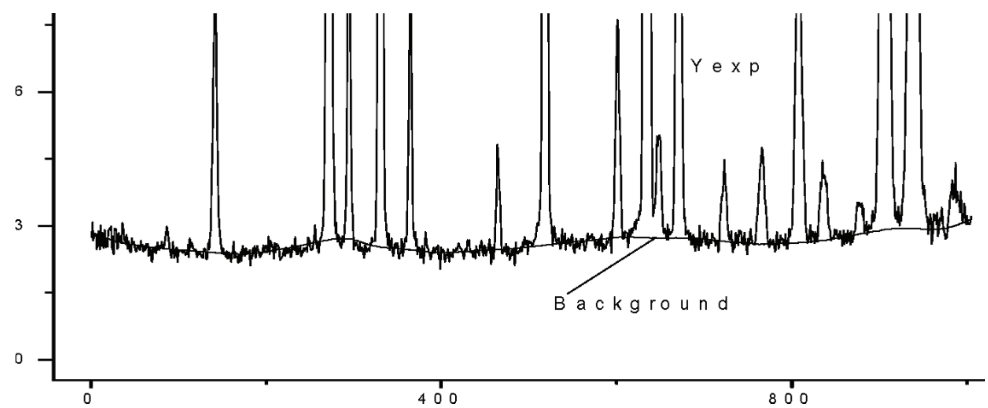

Fig. 5. Identifying the background line from a neutronogram.

Application

Our algorithm has been successfully applied to process the data of powder diffractograms, Auger spectroscopy, gas electronography, and neutronograms (Figs. 3–5).

With $N = 4,096$ spectral points the computation time is less than fractions of a second on a medium-performance PC. Tests have shown that the number of spectral points is restricted only by the PC memory. In this sense it is relevant to consider the following formulation of the background line problem: the spectrum is delivered by a continuously functioning instrument; it is required to output in real time the spectral values without the background line. Such an algorithm has already been developed, and it is currently being tested.

CONCLUSION

Tikhonov's regularization method has been applied to develop an efficient algorithm and appropriate software for the identification of a smooth background line from experimental data. The algorithm is not limited to background line identification: it also can be used to construct filtering functions.

REFERENCES

1. A. N. Tikhonov and V. Ya. Arsenin, *Methods of Solving Ill-Posed Problems* [in Russian], Nauka, Moscow (1979).
2. V. A. Morozov, *Regular Methods for Solving Ill-Posed Problems* [in Russian], Izd. MGU, Moscow (1974).
3. A. N. Tikhonov, A. B. Vasil'eva, and A. G. Sveshnikov, *Differential Equations* [in Russian], Nauka, Moscow (1980).
4. A. A. Samarskii and E. S. Nikolaev, *Solution Methods for Grid Equations* [in Russian], Nauka, Moscow (1978).
5. R. Z. Deyanov and B. M. Shchedrin, "Reconstructing a smooth function by deterministic regularization method," *Vychisl. Metody Programmirovanie,* MGU, Moscow, No. 39, 55–61 (1983).
6. I. S. Berezin and N. P. Zhidkov, *Numerical Methods* [in Russian], vol. I, Nauka, Moscow (1966).
7. V. V. Voevodin and E. E. Tartyshnikov, *Computations with Toeplitz Matrices* [in Russian], Nauka, Moscow (1987).