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**UNFOLDING OF COMPOSITE SPECTRA
BY LINEAR REGRESSION**

by

W. MATTHES

1974



**Joint Nuclear Research Centre
Ispra Establishment - Italy**

Nuclear Study

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ABSTRACT

We consider the problem of resolving a measured pulse-height spectrum of a material mixture (e.g. Raman-spectrum, gamma-spectrum) into a weighted sum of the spectra of the individual components of the mixture. If the measured spectrum is contaminated with noise, the standard least-square method cannot be used to unfold the spectrum into its individual components for small signal-to-noise ratios. To improve the identification of the components in the mixture we constructed a "stepwise regression" method which gives very good results even for signal to noise ratios of the order of one. The new method is a combination of the least-square mechanism and repeated application of statistical tests.

KEYWORDS

SPECTRA
RESOLUTION
PULSES
LEAST SQUARE FIT

C O N T E N T S

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1) Introduction

We consider the problem of resolving a measured pulse-height spectrum of a material mixture (e.g. gamma-ray-spectrum, Raman-spectrum) into a weighted sum of the spectra of the individual constituents of the mixture [1,2,3,4]. The analytical formulation of this problem will be based on the following model:

- (1) the measured spectrum of the mixture is represented by the vector $\vec{N} = \{N_1, N_2, \dots, N_N\}$, where N_ν is the number of counts in channel ν
- (2) the vector $\vec{S}_i = \{S_{i1}, S_{i2}, \dots, S_{iN}\}$ represents the spectrum of constituent i ($i = 1, 2, \dots, L$) which might possibly be in the mixture
- (3) β_i is the magnitude of the contribution of constituent i to the measured spectrum \vec{N} (β_i is proportional to the concentration of constituent i in the mixture).

The number of counts observed in channel ν is then given by:

$$N_\nu = \sum_{i=1}^L \beta_i S_{i\nu} + D_\nu \quad (1)$$

where D_ν is an error-term for channel ν . For these error-terms we assume that they are statistically independent and have expectation value $E(D_\nu) = 0$. Each N_ν has a Poisson distribution with mean-value and variance equal to

$$\lambda_\nu = \sum_{i=1}^L \beta_i S_{i\nu} \quad (2)$$

The probability for the measured spectrum \vec{N} becomes

$$P(\vec{N}) = \prod_{\nu=1}^N \frac{e^{-\lambda_\nu} \lambda_\nu^{N_\nu}}{N_\nu!} \quad (3)$$

Now we apply the principle of Maximum Likelihood [5] to obtain estimated values for the unknown β_i . Due to this principle we have to perform the operations:

$$\frac{\partial}{\partial \beta_i} \ln P = 0 \quad (i = 1, 2, \dots, L) \quad (4)$$

on (3) and obtain:

$$\sum_{\nu=1}^N \left(1 - \frac{N_{\nu}}{\lambda_{\nu}}\right) \frac{\partial \lambda_{\nu}}{\partial \beta_i} = 0 \quad (5)$$

For practical applications we may assume that N_{ν} will deviate not too much from λ_{ν} so that we can put

$$\frac{N_{\nu}}{\lambda_{\nu}} \approx 1 + \frac{\delta N_{\nu}}{\lambda_{\nu}}, \quad \frac{\lambda_{\nu}}{N_{\nu}} \approx 1 - \frac{\delta N_{\nu}}{\lambda_{\nu}} \quad (6)$$

and therefore

$$1 - \frac{N_{\nu}}{\lambda_{\nu}} \approx - \left(1 - \frac{\lambda_{\nu}}{N_{\nu}}\right) \quad (7)$$

Using (7) and (2) in (5) we finally arrive at the system of linear equations for the β_k :

$$\sum_{k=1}^L \beta_k \left(\sum_{\nu=1}^N \frac{S_{i\nu} S_{k\nu}}{N_{\nu}} \right) = \sum_{\nu=1}^N S_{i\nu} \quad (i=1, 2, \dots, L) \quad (8)$$

This system (8) gives us point-estimators for the β 's but no information about their statistical distribution properties. The knowledge of these

distribution properties is important however for further statements about the accuracy of the solutions of (8), for the construction of confidence intervals and for testing assumptions about the β 's. To obtain this information about the statistical behaviour of the β 's we refer to the assumption made above, that N_ν will deviate not too much from λ_ν . Under this assumption N_ν (greater than about 20 counts) will behave approximately as a random variable with a normal distribution of mean value $E(N_\nu) = \lambda_\nu$ and variance $\sigma_\nu^2 = \lambda_\nu$.

The variance σ_ν^2 of N_ν in our case is a function of the β 's which are not known. On the other hand we have up to terms of higher order:

$$\frac{N_\nu - \lambda_\nu}{\sigma_\nu} \approx \frac{N_\nu - \lambda_\nu}{\sqrt{N_\nu}} \quad (9)$$

We assume therefore further σ_ν to be given and replace it for numerical calculations by $\sqrt{N_\nu}$.

We introduce the quantities:

$$\begin{aligned} Y_\nu &= N_\nu / \sqrt{N_\nu}; \quad \vec{Y} = \{Y_1, Y_2, \dots, Y_N\}; \quad \vec{\epsilon} = \{\epsilon_1, \epsilon_2, \dots, \epsilon_N\} \\ X_{i\nu} &= S_{i\nu} / \sqrt{N_\nu}; \quad \vec{X}_i = \{X_{i1}, X_{i2}, \dots, X_{iN}\}; \\ \zeta_\nu &= \sum_{i=1}^4 \beta_i X_{i\nu}; \quad \vec{\zeta} = \{\zeta_1, \zeta_2, \dots, \zeta_N\} \end{aligned} \quad (10)$$

which give equation (1) the form:

$$Y_\nu = \sum_{i=1}^4 \beta_i X_{i\nu} + \epsilon_\nu$$

or

$$\vec{Y} = \vec{\zeta} + \vec{\epsilon} \quad (11)$$

where ϵ_v is normally distributed with mean value 0 and variance 1.

Our problem can then be stated in the following way:

Given a vector \vec{y} in an N dimensional space V_N with components y_v ($v = 1, 2, \dots, N$). Determine that vector $\vec{z} = \sum_{i=1}^{N_L} b_i \vec{x}_i$ in the subspace V_L , spanned by the L linear independent vectors \vec{x}_i , which makes

$$S = |\vec{y} - \vec{z}|^2 = \sum_{v=1}^N (y_v - b_1 x_{1v} - b_2 x_{2v} \dots - b_L x_{Lv})^2 \quad (12)$$

to a minimum.

The corresponding b_i are found out of the system of equations (equivalent to (8)):

$$\sum_{k=1}^L (\vec{x}_i \cdot \vec{x}_k) b_k = (\vec{x}_i \cdot \vec{y}) \quad (13)$$

This means that the error vector

$$\vec{\epsilon} = \vec{y} - \sum_{i=1}^L b_i \vec{x}_i \quad (14)$$

is orthogonal to V_L and

$$|\vec{\epsilon}|^2 = S \quad (15)$$

has a χ^2_{N-L} distribution of (N-L) degrees of freedom.

The initial problem (1) to (8) is thus reduced to a multiple linear regression problem as formulated in (11) to (15). It is well known [6,7,8] that the solutions b_k of the system (13) are normally distributed with mean value $E(b_k) = \beta_k$ and variance $\sigma_{b_k}^2 = a^{kk}$ where a^{kk} is a diagonal element of the inverse matrix to $a_{ik} = (\vec{x}_i \cdot \vec{x}_k)$.

2) Stepwise linear regression

A straightforward solution of the system (13) for the b's could only be feasible if L is not too large and if the constituents-spectra \vec{x}_i were accurately known. These are just two points which are not fulfilled in our case. We shall have to deal with the problem that the number L of constituents which could be in the mixture under investigation is very large ($L \gg 50$ or more) but that the number of constituents which actually are in the mixture is very small ($\ll 10$). Further, the library spectra \vec{x}_i are in general found experimentally and as such are contaminated with a noise component. Thus taking into account all L components when solving (13) for the b_i the allowance for components which are not in the mixture ($\beta_i = 0$) will strongly influence the accuracy of those b_i for constituents which actually are in the mixture. To solve this difficulty we shall construct a stepwise regression procedure which selects a most probable combination of constituents from the set of library spectra by means of repeated application of statistical tests and the least square mechanism. The final result will give both the constituents contained in the mixture and their respective (relative) strength (contribution to the measured spectrum \vec{y}). To arrive at this stepwise regression procedure we employ the statistical test for the hypothesis that some of the β 's are identically zero. This test-situation can be described in the following terms [8,9] :

Let us assume that only the Q spectra out of the set $T_0 = \{\vec{x}_{i1}, \vec{x}_{i2} \dots \vec{x}_{iQ}\}$ contribute to the measured spectrum \vec{y} and no other spectrum out of the remaining set $T_1 = \{\vec{x}_{j1}, \vec{x}_{j2}, \dots, \vec{x}_{jL-Q}\}$ will give any "significant contribution".

In other words, if we would add an arbitrary spectrum $\vec{x}_{j\mu}$ of T_1 to T_0 and try to fit \vec{y} within the set $T_\mu^+ = \{\vec{x}_{i1}, \vec{x}_{i2} \dots \vec{x}_{iQ}; \vec{x}_{j\mu}\}$, the assumption says that no $\vec{x}_{j\mu}$ is necessary to fit \vec{y} or that the corresponding $b_{j\mu}$ are indentially zero for all μ . If our hypothesis is true, then (for each μ) the quantity

$$f(\mu) = \frac{S(T_0) - S(T_\mu^+)}{S(T_\mu^+)} \cdot (N - Q - 1) \quad (16)$$

has a $F_{(1, N-Q-1)}$ distribution with 1 and $(N-Q-1)$ degrees of freedom, where

$$S(T_0^{(\mu)}) = \text{MIN} \left| \vec{y} - \sum_{i \in T_0^{(\mu)}} b_i \vec{x}_i \right|^2 \quad (17)$$

Due to this fact we can find a number $F_{(1, N-Q-1)}^{P\%}$ such that

$$\text{Prob.} \left(f^{(\mu)} > F_{(1, N-Q-1)}^{P\%} \right) = P\% \quad (18)$$

choosing for P values of the order of 1%, 5% or 10% we can be (almost) sure that all $f^{(\mu)}$ will be smaller than $F_{(1, N-Q-1)}^{P\%}$ if our hypothesis is true (that no $\vec{x}_{j\mu}$ is necessary to fit \vec{y}).

The test of this hypothesis will therefore be performed in four steps:

- 1) Calculate $f^{(\mu)}$ for all $\vec{x}_{j\mu}$ out of T_1 .
- 2) If $f^{(\mu)} \leq F_{(1, N-Q-1)}^{P\%}$ for all μ go to step 4.
- 3) If for some μ we have $f^{(\mu)} > F_{(1, N-Q-1)}^{P\%}$, we choose the m largest of these $f^{(\mu)}$ (usually $m = 1$) and add the corresponding spectra to T_0 , remove then from T_1 and go back to step (1) to repeat this "forward selection procedure".
- 4) The hypothesis is accepted that the constituents of T_0 only make up the mixture and no component of T_1 will make any further "significant contribution" to \vec{y} .

It might still be possible that during this "forward selection procedure" we added to many components to T_0 and some of the $\vec{x}_{i\mu}$ (of the final T_0) have a negligible influence on y .

To test for this possibility we make the hypothesis that e.g. $\vec{x}_{i\mu}$ has no significant influence on y and to fit y it is enough to use the spectra of set T_{μ}^- only, where T_{μ}^- contains all spectra of T_0 except $\vec{x}_{i\mu}$. If this hypothesis is true, then the quantity

$$g^{(\mu)} = \frac{S(T_{i\mu}) - S(T_0)}{S(T_0)} \cdot (N-Q) \quad (19)$$

has a $F_{(1, N-Q)}$ distribution.

Now we apply the above "forward"-procedure in the reverse sense and perform the following four steps:

- 1) Calculate $g^{(\mu)}$ for all $\vec{x}_{i\mu}$ out of T_0 .
- 2) If $g^{(\mu)} > F_{(1, N-Q)}^{P\%}$ for all μ go to step 4.
- 3) If for some μ we have $g^{(\mu)} \leq F_{(1, N-Q)}^{P\%}$, we choose the n smallest of these $g^{(\mu)}$ (usually $n = 1$), remove the corresponding spectra from T_0 and go back to step (1) to repeat this "backward elimination procedure".
- 4) The hypothesis is accepted that no spectrum of T_0 may be neglected. This means that the experimental data (\vec{y}) are consistent with the assumption that all components of T_0 (and not more) are necessary to fit \vec{y} .

This stepwise regression method consists therefore of the two essential steps:

- a) the "forward selection procedure" selects by repeated application of the statistical tests out of the set of all L possible library spectra a subset T_0 of vectors which should all be included in the regression. All remaining vectors in T_1 are rejected as the final test supports the hypothesis that all B 's of the vectors in T_1 could be assumed to vanish.
- b) The "backward" elimination procedure" eliminates from T_0 found in step (a) further variables for which a test supports the hypothesis that their corresponding B 's can be assumed to vanish.

Only if we are sure that the complete T_0 is found (as a result of the applied tests in step (a)) we can start with the elimination procedure. This is the point where the method described above differs from published stepwise regression methods [7, 10] which perform the elimination procedure immediately after each forward selection step.

3) Application of the stepwise regression method

In order to test the efficiency of the stepwise regression method (SRM) we transformed the procedures described in chapter 2) in a computer programme. This programme was then used (to simulate a practical application) to unfold test-spectra which were obtained by mixing some spectra (out of a library of arbitrarily chosen spectra) and adding a noise component.

The L(=50) library spectra were constructed by a linear superposition of I (= 10) Gaussian curves with random amplitudes, random widths and random positions with an average distance of $\frac{M}{I}$ between the peak positions, where M (= 10 cm) is the range over which the spectra were chosen to extend (the range M is chosen to correspond to N (=100) channels). Figs. 1a,b,c show some typical library spectra. Some of these library spectra were now linearly superposed with different weighting factors to obtain a pure signal spectrum and to obtain measured spectra each signal spectrum was contaminated with (Gaussian random-) noise of mean zero and variance 1 and a constant background (=5.0) was added (see Fig. 2).

The contamination of the signal spectra with noise was done for different signal-to-noise-ratios (SNR) to simulate measured spectra for different experimental conditions. The signal-to-noise-ratio is defined by

$$SNR^2 = \frac{\sum_{i=1}^N (S(i) - \bar{S})^2}{\sum_{i=1}^N N(i)^2} \quad (20)$$

where: S(i) = Signal value in channel i

N(i) = Noise value in channel i

$$\bar{S} = \frac{1}{N} \sum_{i=1}^N S(i); \text{mean value of signal}$$

A typical sample of "measured" spectra for different SNR's is shown in Figs. 3a,b,c,d.

The results of the unfolding of the "measured" spectra of Fig. 3 using the least-square method and the step-wise regression method are shown in Figs. 4a-e.

Many calculations were made with varying numbers K of components in the signal spectrum (1 to 20 components out of 50) and different signal-to-noise ratios (0.5, 1, ..., 30).

The results of these calculations were all similar to the special case mentioned above and shown in Figs. 3 and 4.

4) Conclusion

The main conclusions out of these calculations can be summarized as follows:

The least-square method cannot be used to resolve a mixture of $K(\leq 10)$ out of $L(\geq 20)$ spectra in the range of $\text{SNR} \leq 5.0$.

The stepwise method in this range of the SNR identifies in general all components of the mixture and gives their (absolute) weights within 10%. It might happen that some components are not identified, but if the stepwise method identified a component it was in the mixture. For larger signal-to-noise ratios the ability of the least-square method to predict the component-spectrum of a mixture improves and for $\text{SNR} \geq 10.0$ both methods give equally good results (at least for the calculations performed, that means resolving mixtures of $K(\leq 10)$ out of $L(\leq 50)$ library-spectra).

Figure Captions

Fig. 1a,b,c: Some of the arbitrarily chosen library spectra.

Fig. 2 Superposition of a constant background and a random Gaussian noise component of mean zero and variance 1. The background is also considered as a library spectrum (no. 1) and can be chosen arbitrarily.

Fig. 3: a) Signal spectrum obtained from a linear superposition of the library spectra no. {3,6,9,13,19} with the weight factors {0.45, 0.15, 0.28, 0.37, 1.12}

b) Measured spectrum obtained from a linear superposition of the signal spectrum of Fig. 3a with the noise and background of Fig. 2 and arranged for a SNR of 0.5

c) same as Fig. 3b for a SNR = 1.0

d) same as Fig. 3b for a SNR = 5.0

Fig. 4: a) Component spectrum of the signal spectrum of Fig. 3a

b) Least-square solution for the component spectrum of Fig. 3c (SNR = 1.0)

c) Least square solution for the component spectrum of Fig. 3d (SNR = 5.0)

d) Stepwise solution for the component spectrum of Fig. 3c. Components 6 and 13 of Fig. 4a are not identified.

e) Stepwise solution for the component spectrum of Fig. 3d. Only component 6 of Fig. 4a is not identified.

Fig. 5: a) Component spectrum for a mixture of 10 components, each with the same weight-factor of 0.5.

b) Stepwise solution for the component spectrum of Fig. 5a for a SNR = 5.0.

Only component 39 has been wrongly identified.

c) Least-square solution for the component spectrum of Fig. 5a (SNR = 5.0).

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LIBR. SPECTR.

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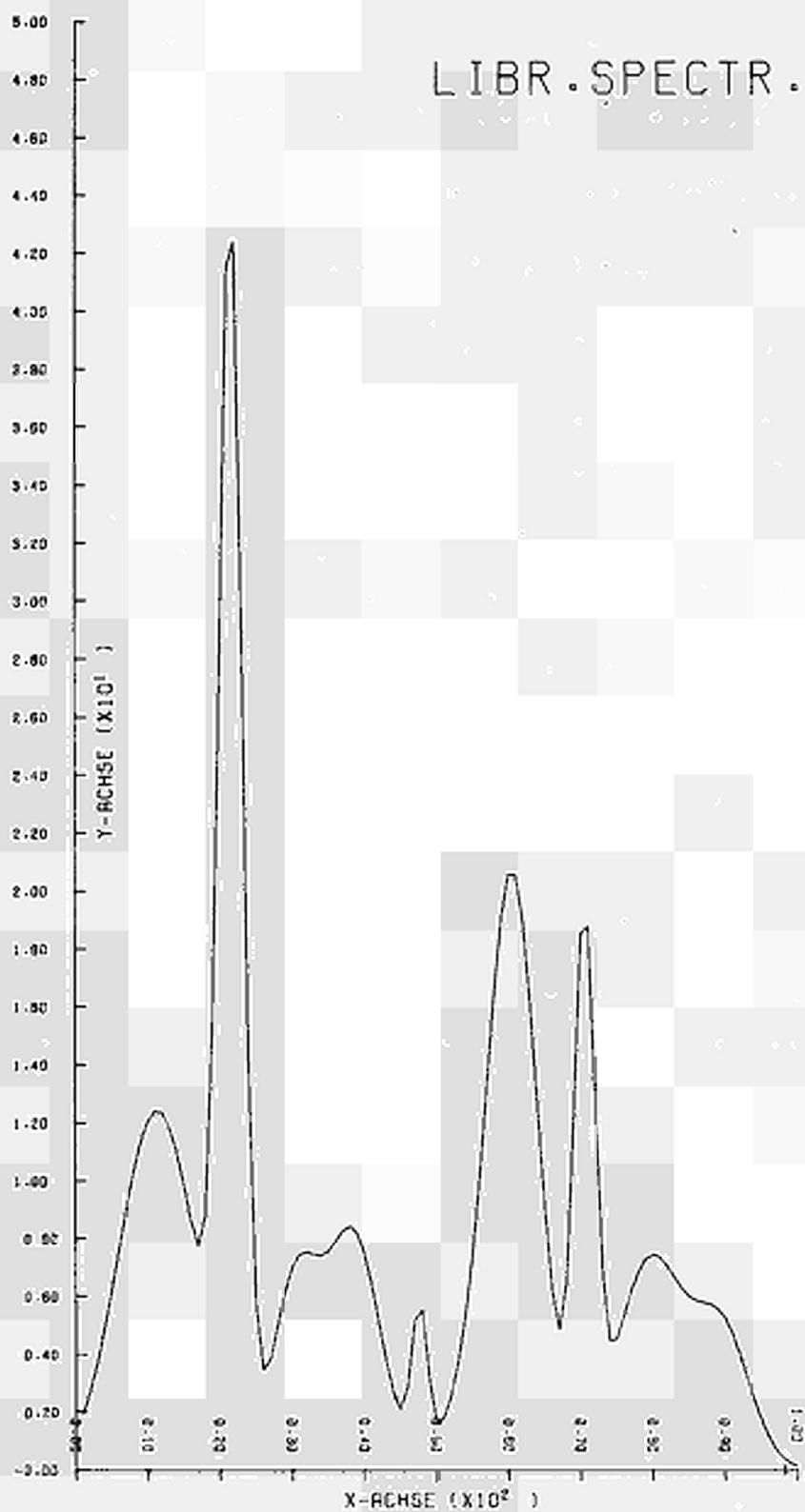


Fig. 1a

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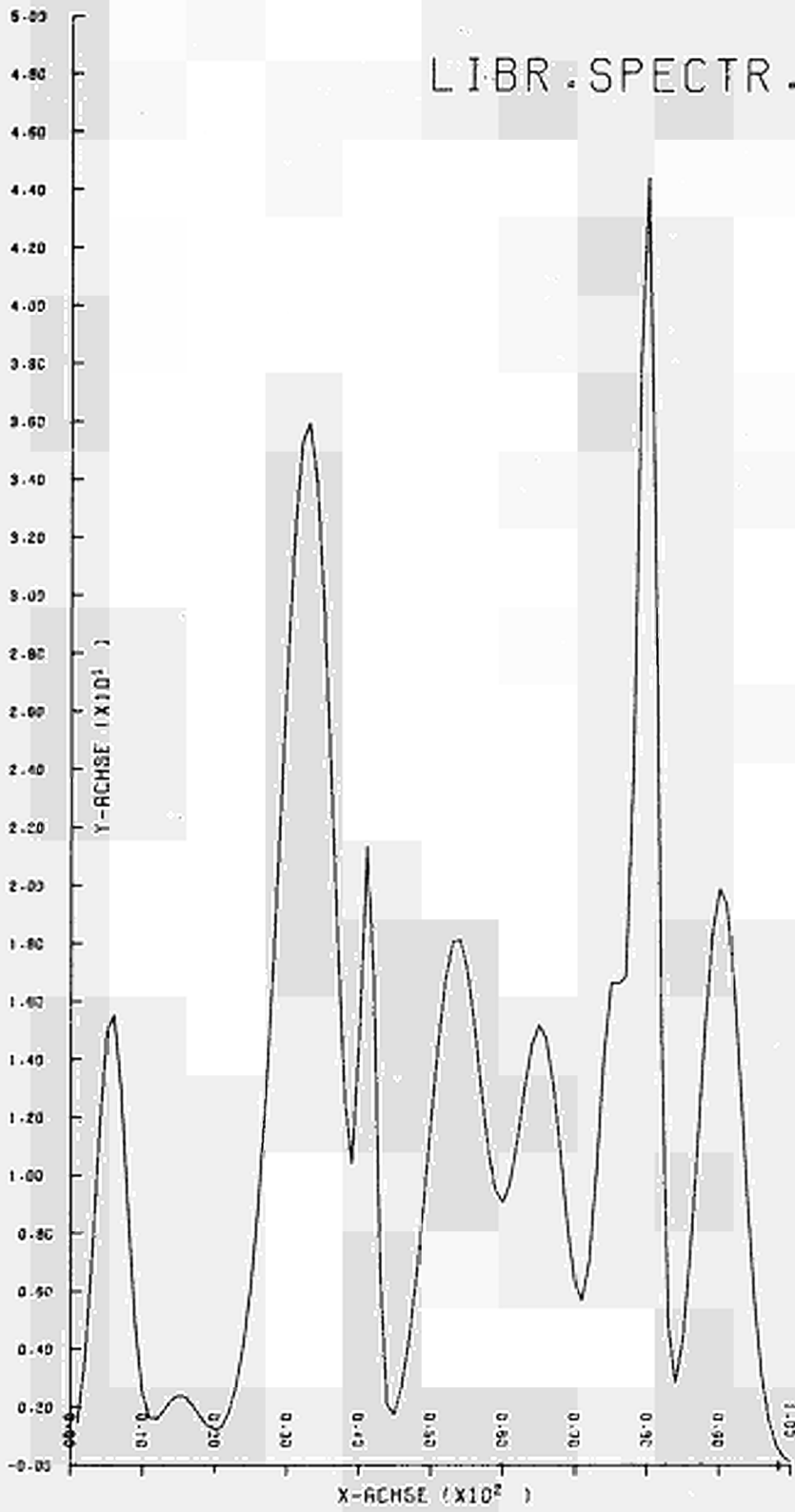


Fig. 1b

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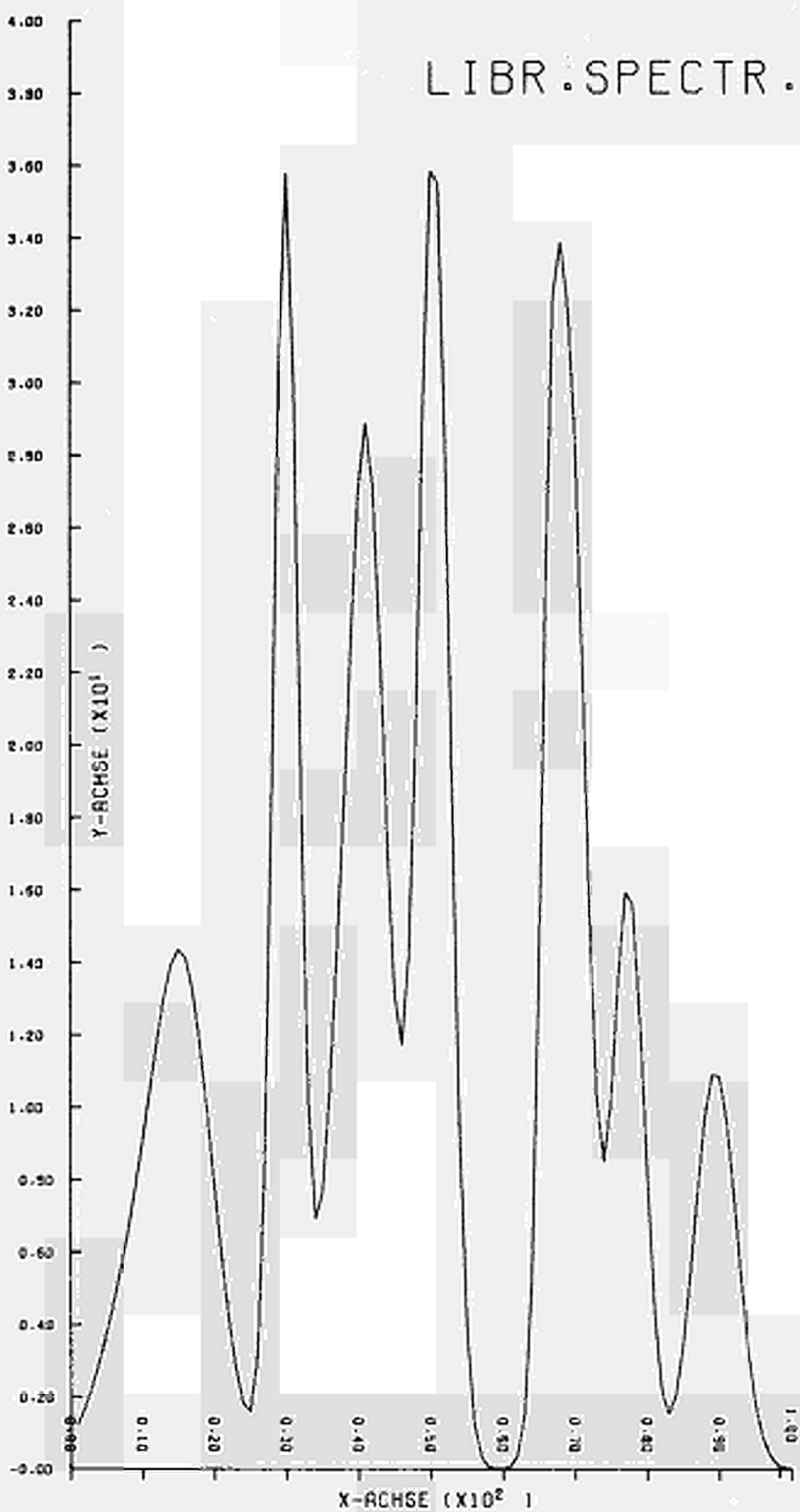


Fig. 1C

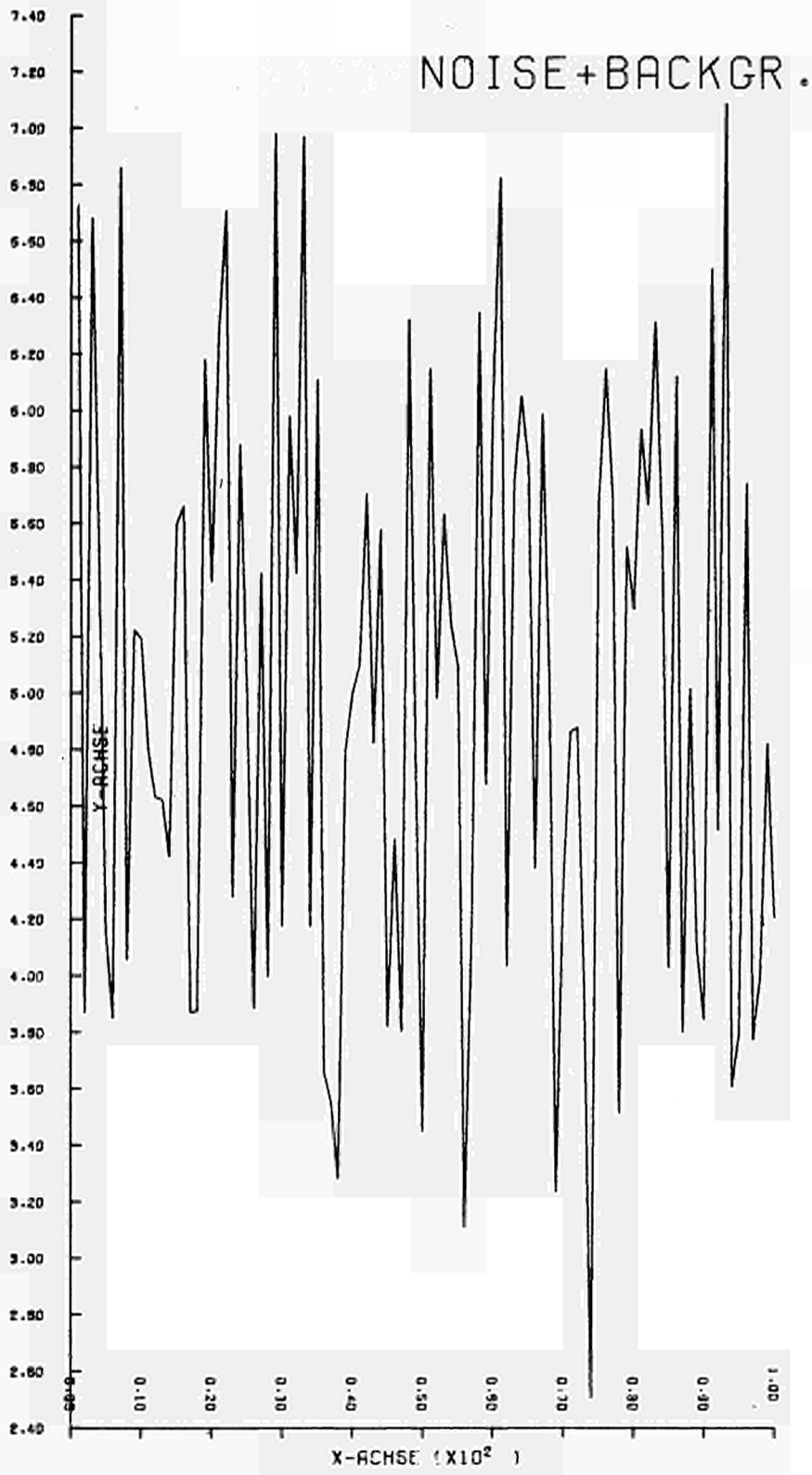


Fig. 2

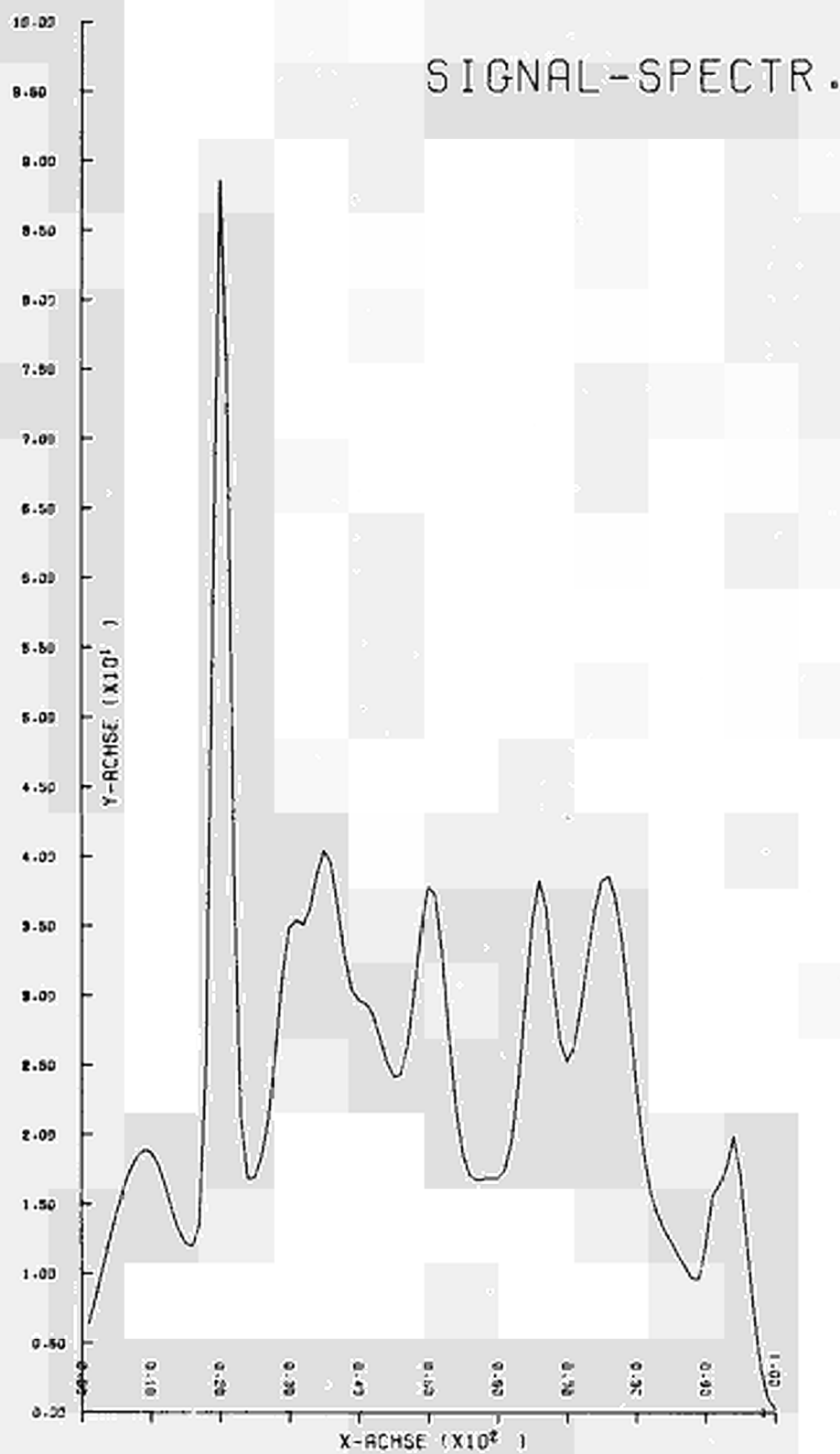


Fig. 3a

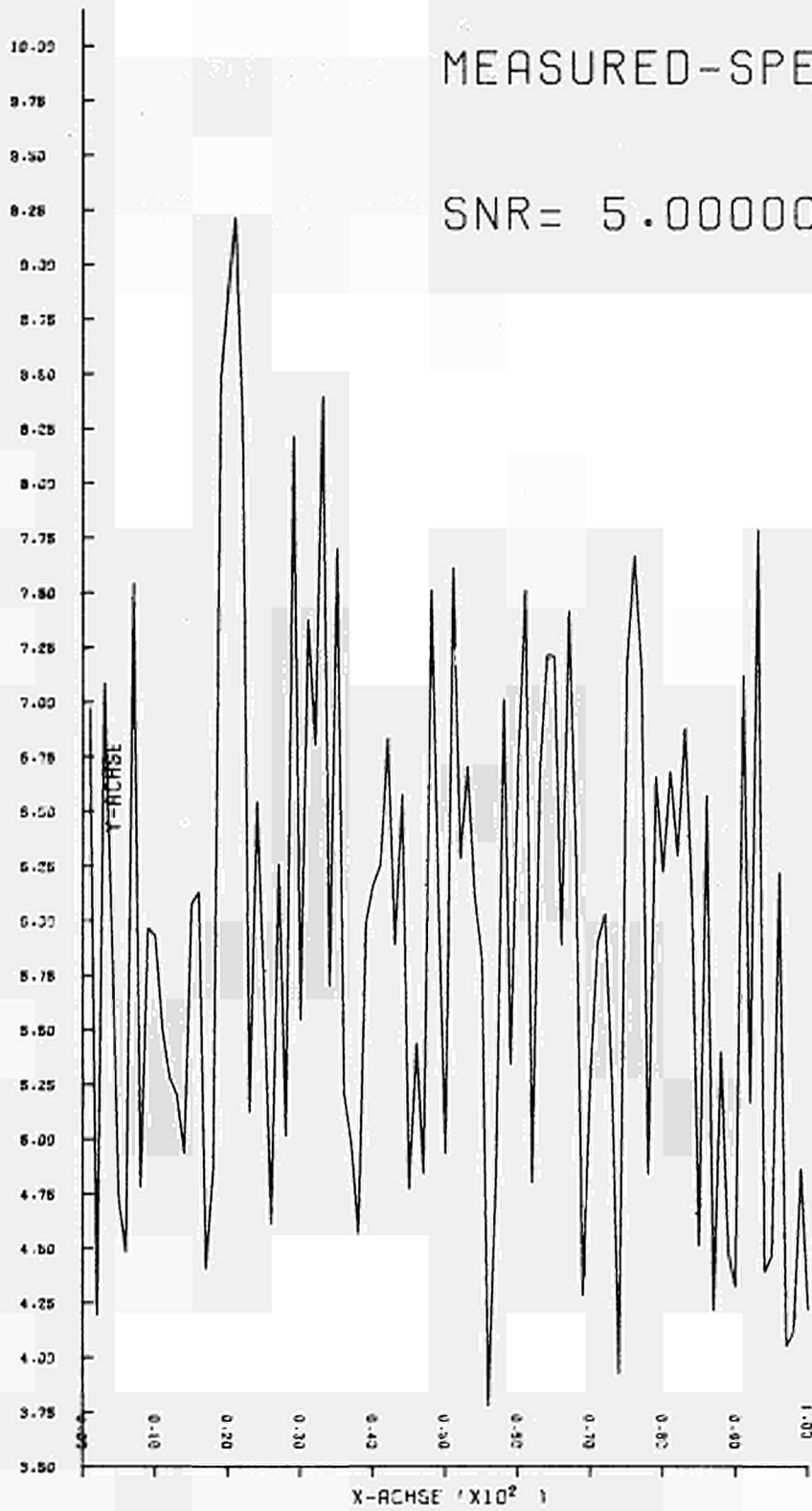


Fig. 3b

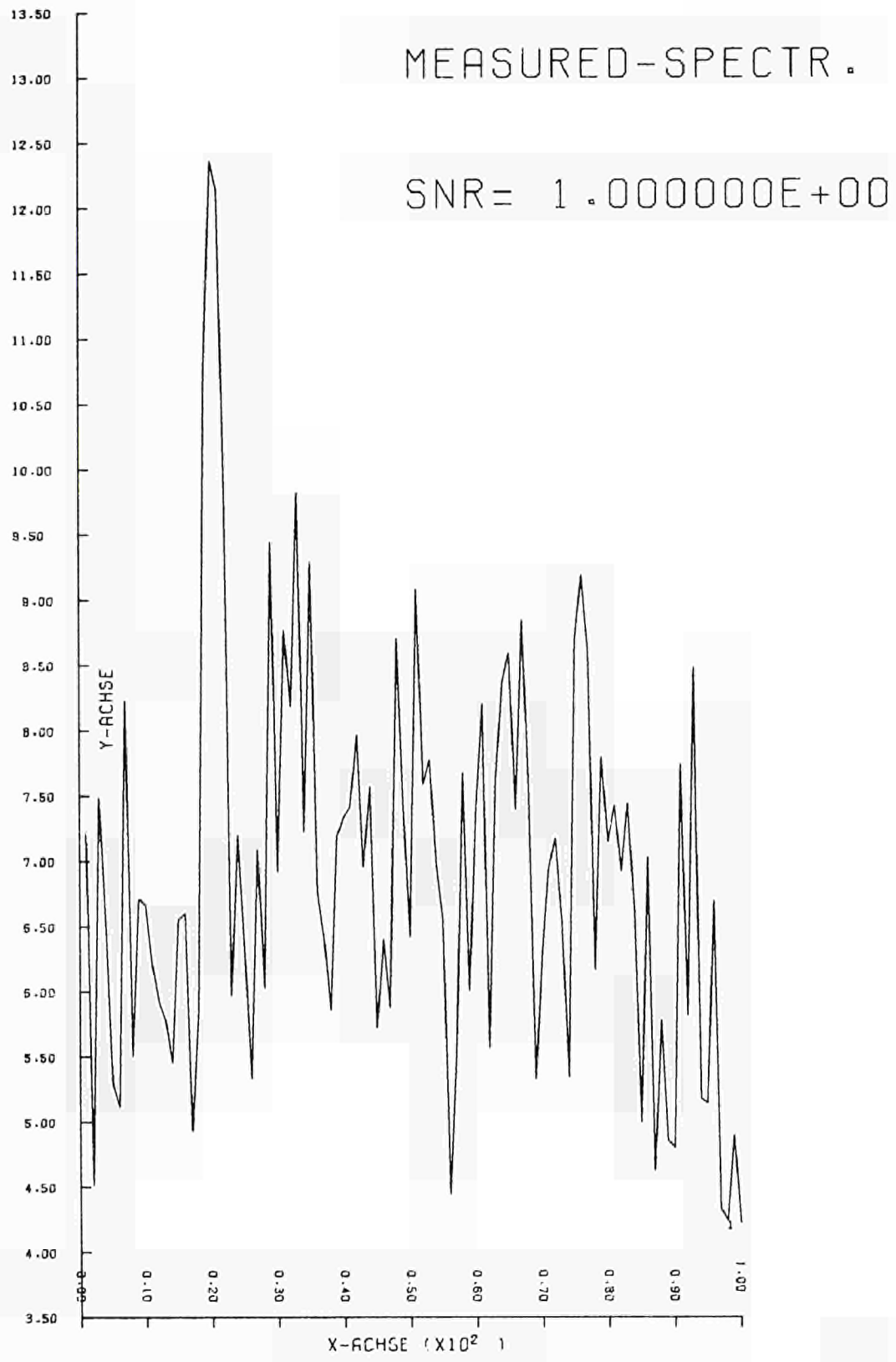


Fig 3c

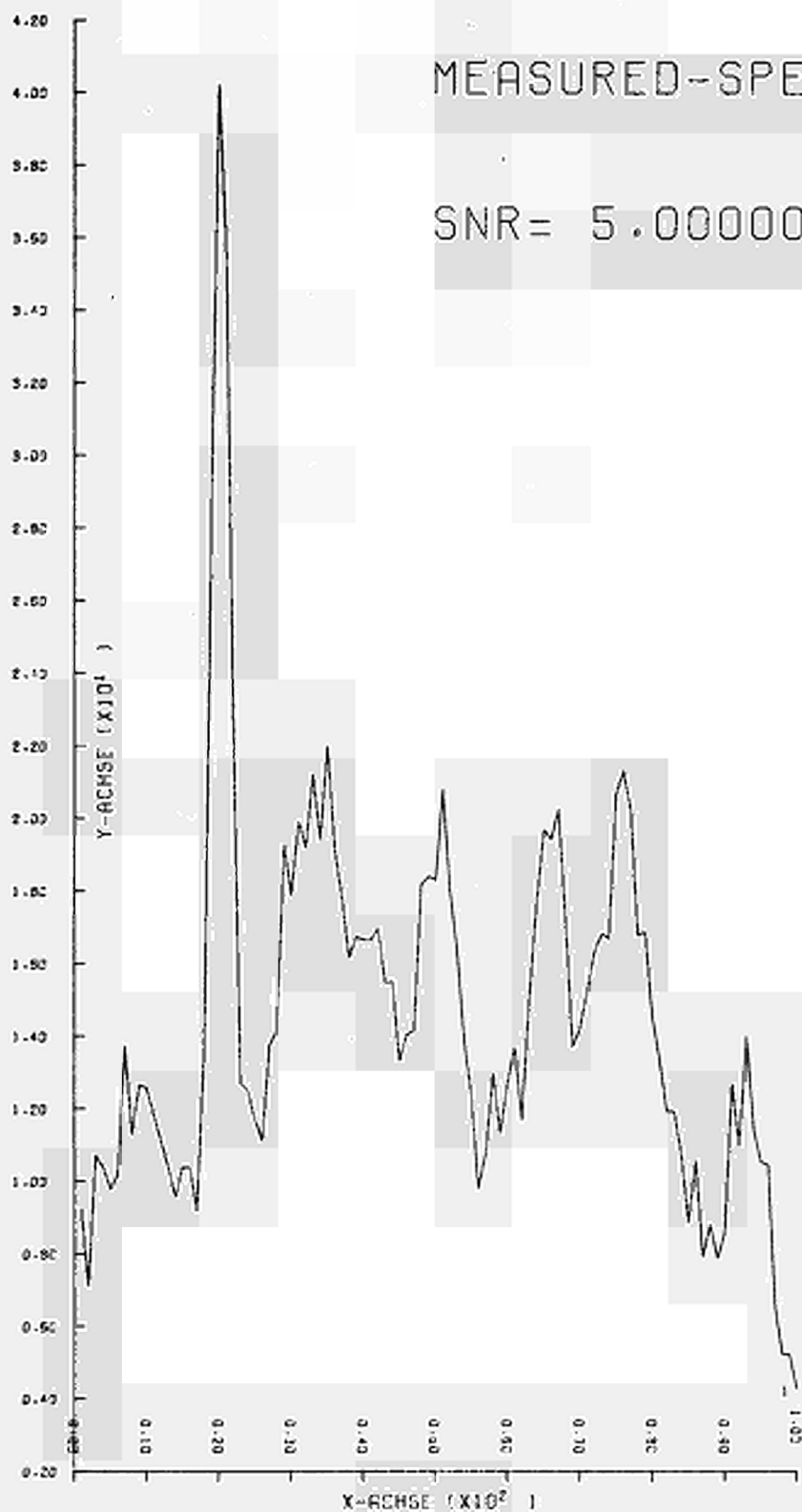


Fig. 3d

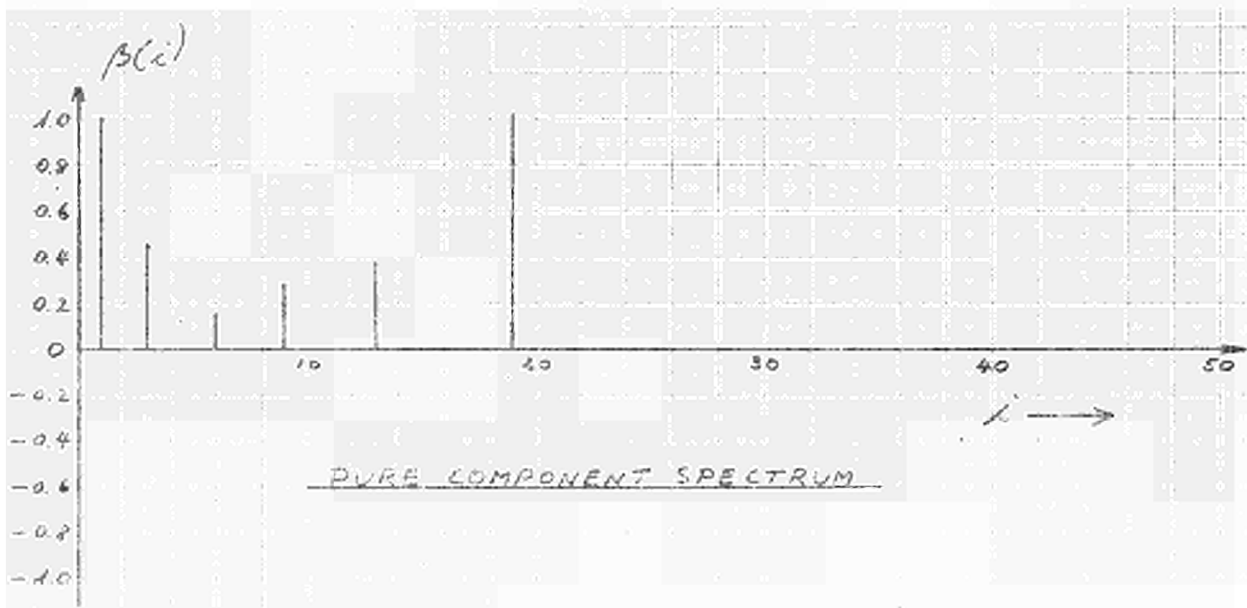


Fig. 4a

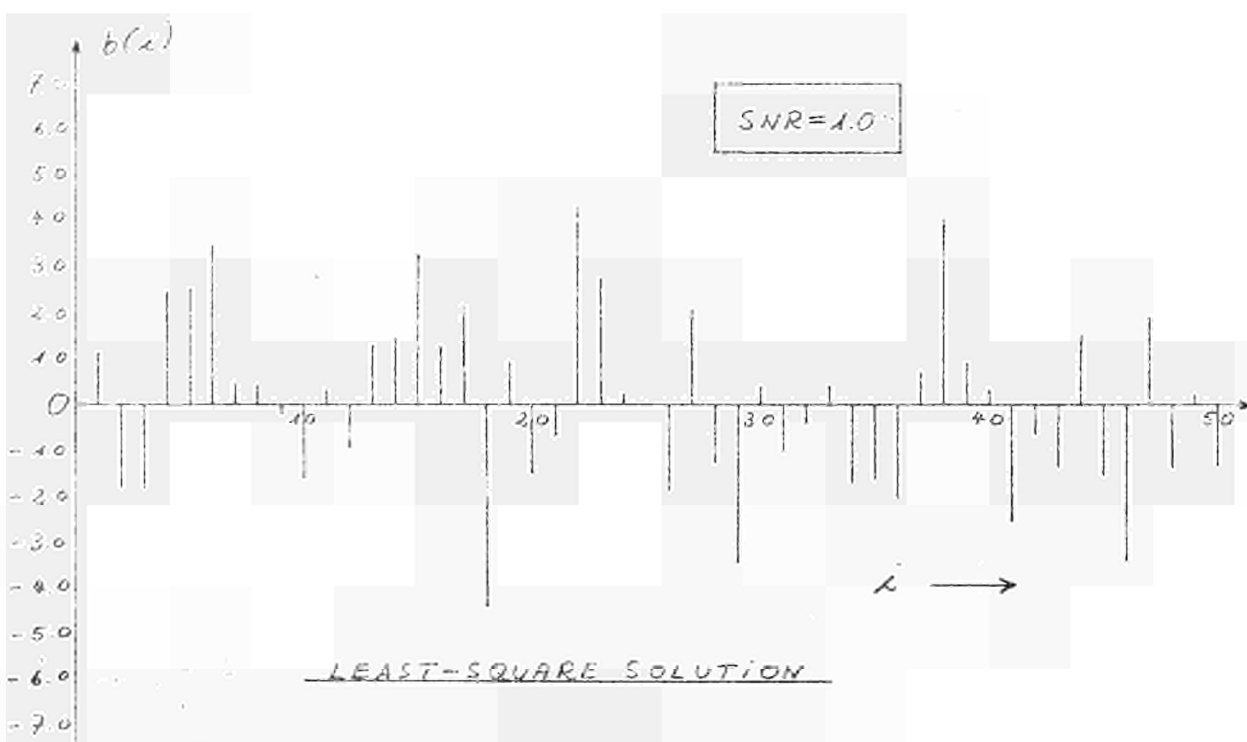


Fig. 4b

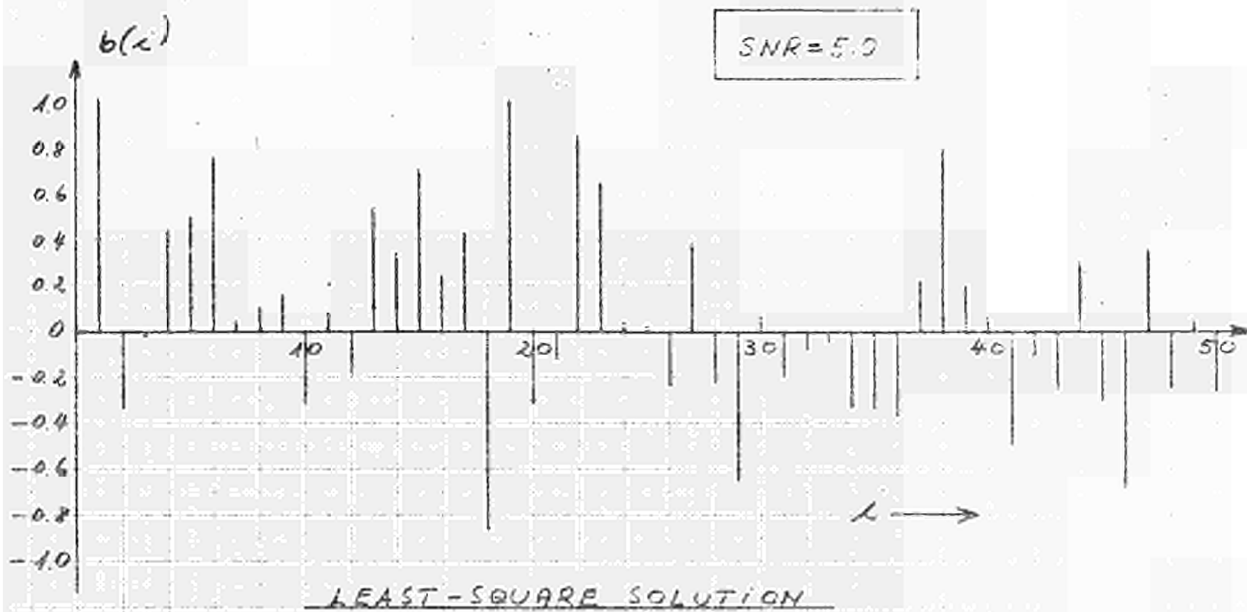


Fig. 4c

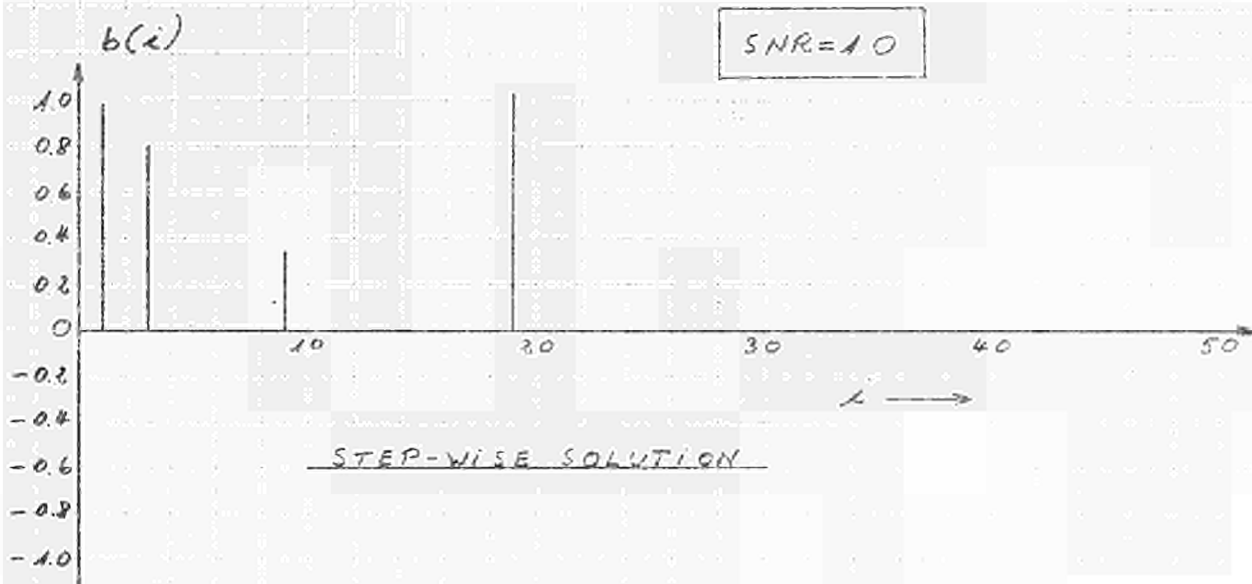


Fig. 4d

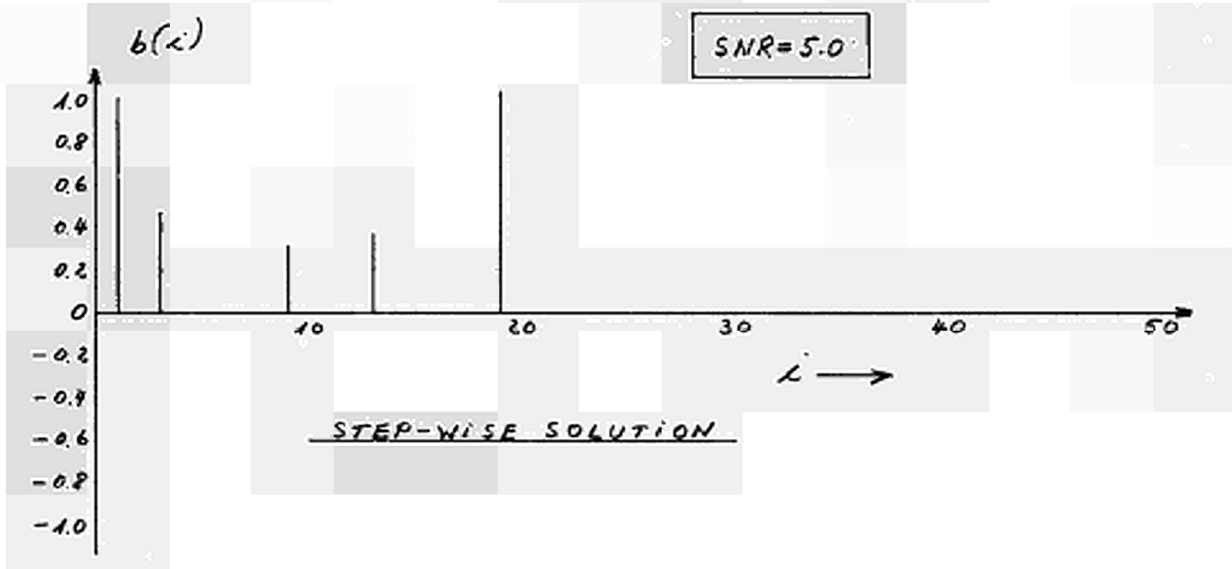
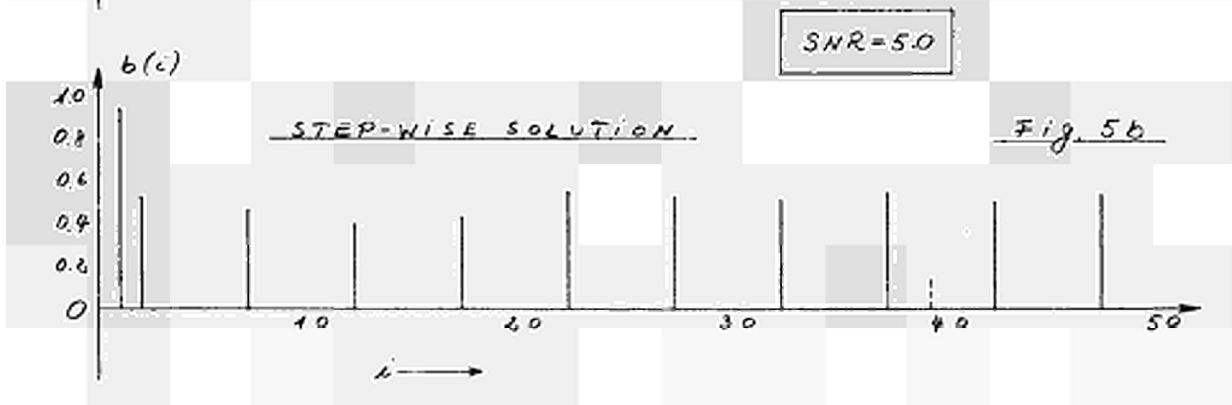
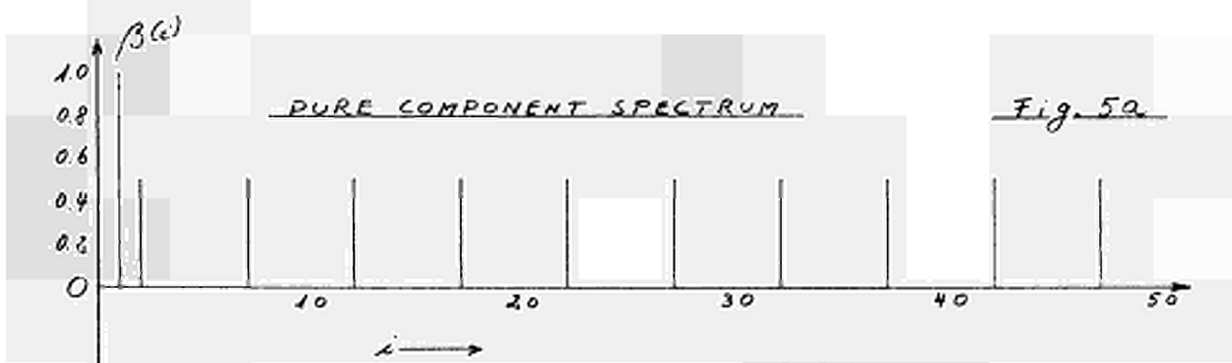
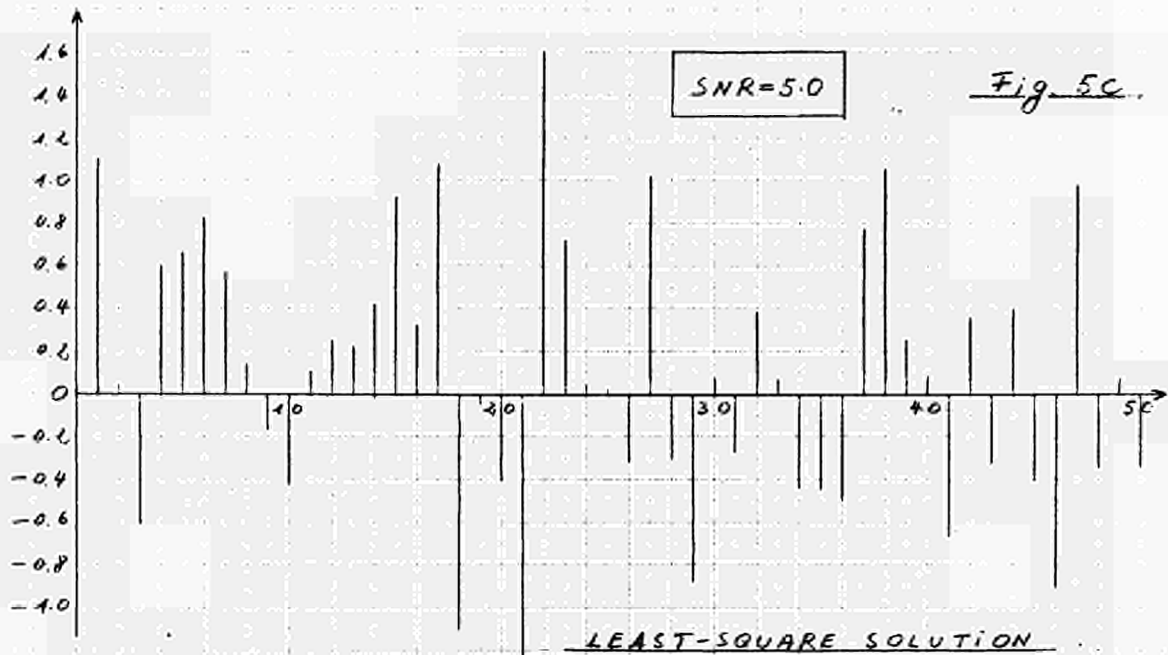


Fig. 4e

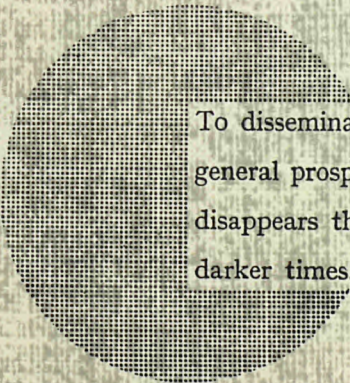




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