American Mineralogist, Volume 60, pages 257–264, 1975

Statistical Limitations of Mössbauer Spectral Fitting

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Abstract

The statistical quality, Ξ, of any peak in a given Mössbauer spectrum can be defined in terms of the observed spectral parameters: background counts, velocity increment, and the width and dip of the peak. For isolated peaks the statistical uncertainty with which each peak parameter may be determined, from a given spectrum, is an inverse linear function of the peak Ξ value.

For spectra with partially overlapped peaks, parameter uncertainty further depends on component peak separation. Below a critical separation where separate minima are visually discernible, probable parameter error rises very rapidly to infinity. The limits of recovering the parameters of strongly overlapped peaks and the (large) uncertainties involved are graphically presented.

Introduction

It is not uncommon to find, in the recent literature, examples of Mössbauer spectra which show a limited number of well-defined minima, but which have been fitted with a substantially larger number of "incompletely resolved component peaks" by the investigator. The validity of this procedure and the probable errors involved depend upon the uncertainty of the spectral measurements and upon the fitted model. Under optimum conditions with no systematic errors present in the data and *an appropriate model,* the fitting errors are those due purely to the statistical uncertainty of the spectral data. It is this statistical limitation to spectral fitting that is the subject of this paper and is considered herein for the specific case of Mössbauer spectra measured in the usual constant-acceleration mode. The conclusions, however, are adaptable to other types of spectroscopy.

An empirical approach using computer-simulated Mössbauer spectra is employed, and the effects of peak quality and peak overlap on probable peak-parameter errors are graphically presented. It is assumed that the spectroscopist is already aware of the statistical limitations of the data. This paper is rather aimed at the users, or potential users, of Mössbauer data who wish to know, "to what degree does this particular data set support this particular model?"

Statistical Quality of Mössbauer Spectral Peaks

Most routine Mössbauer spectra are measured using a so-called constant acceleration spectrometer which accumulates the spectrum in a multichannel analyzer (see, *e.g.,* Greenwood and Gibbs, 1971, chap. 2). In this experimental configuration the gamma-ray source (or absorber) is repeatedly swept at constant acceleration through a range of velocities, typically on the order of -5 mm/sec to $+5$ mm/sec, for Fe-57 studies. Each channel of the multichannel analyzer (MCA) contains the counts recorded while the source was moving in a particular velocity interval. For instance, velocities of 5.00 to 4.98 mm/sec may be associated with channel 1, velocities of 4.98 to 4.96 to channel 2, *etc.* In this example the velocity increment of 0.02 mm/sec/channel would require 500 channels to cover a -5 to a $+5$ mm/sec range.

An example of such a spectrum is shown in Figure 1, and is seen to consist of a plot of the total number of counts received in each of the MCA channels. (The relation of velocity to channel number, or calibration of the spectrometer, is accomplished through electromechanical or electrooptical techniques, or through the use of standards.) Note that the counts at high negative velocities (low channel numbers) and again at high positive velocities (high channel numbers) are nearly constant, differing only by statistical noise. This background count, which could be more precisely defined as the average count per channel at velocities well removed from any spectral peaks, is termed the off-resonance count. Nearer the center of the spectrum some of the channels show a significant decrease in the number of counts accumulated. This loss of counts is due to the resonant absorption and re-emission of gamma photons in the absorber sample being studied.

Ideally the shape of the Mössbauer absorption

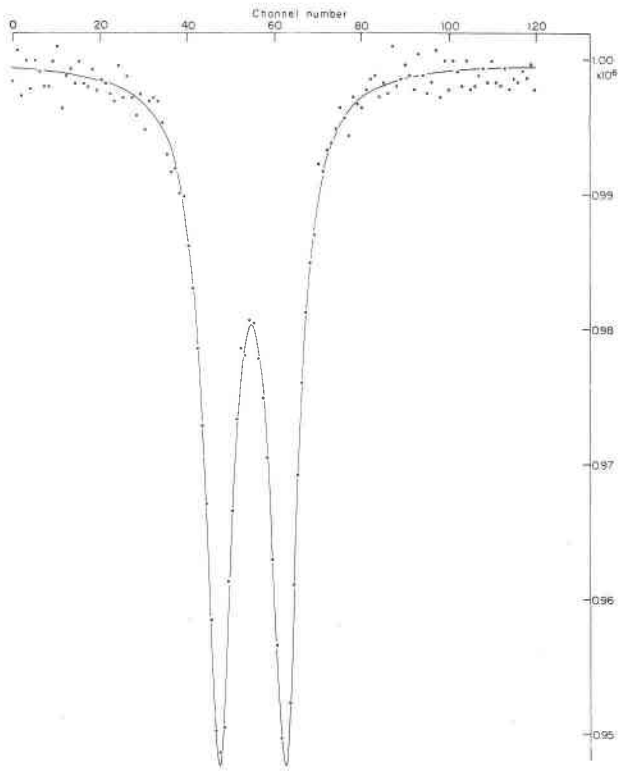

FIG. 1. Example of computer simulated spectrum. The data points simulate the number of counts accumulated in each channel of a multichannel analyzer. The solid curve represents the least-squares fitted envelope which is the sum of two Lorentzian peaks separated by 2.0 peak width.

peak conforms to a Lorentzian (also known as Cauchy) profile. In practice, it is found that most spectra closely approximate this form although relatively minor deviations do occur. In those cases where the deviations from Lorentzian form are real, it is often difficult to determine whether this is an inherent property of the material under study or merely an artifact caused by faulty sample preparation, imperfect spectrometer, *etc.* At any rate, these deviations are small and do not materially affect the statistical conclusions arrived at below.

The spectral parameters which characterize each peak in a spectrum are:

1. center location (in units of channel numbers)
2. area (in units of counts × channel numbers)
3. width (in units of channel numbers

The width is usually expressed as full width at half maximum. How well these parameters are defined by the observed spectrum varies widely due to such factors as the strength of the Mössbauer effect in the particular sample studied, the length of time over which the spectrum was accumulated, the amount of sample used and the amount of specific Mössbauer nuclide in the sample, the number and types of different sites within the sample over which this isotope is distributed, the activity of the gamma-ray source, the experimental configuration, *etc.* Despite these experimental variations, however, it is possible to estimate the statistical quality of a specific peak in a given spectrum.

How well defined the spectral parameters are depends basically on the peak-to-background ratio of the specific peak in question. This may be expressed as the depth of the peak below background in units of estimated standard deviations. In the following, the estimated standard deviations are taken directly from counting statistics, thereby assuming no systematic error is present in the spectrum. The probable errors arrived at therefore refer to the precision rather than the accuracy of the spectrum.

The peak depth can be given as the difference in the number of counts observed at the peak center and the off-resonance count. As the Mössbauer effect results in only a small percentage decrease in the counting rate at the peak, the standard deviation of the count in any channel is closely approximated by the square root of the off-resonance count. Thus the desired peak depth in units of counting standard deviation is given by the ratio,

$$R = (N_\infty - N_0)/\sqrt{N_\infty} \qquad (1)$$

where N_∞ refers to the off-resonance count and N_0 the count at the peak.

For spectra measured under constant experimental conditions, the above ratio, R, is in fact proportional to the statistical quality of a given peak. However, consider a spectrum measured under the conditions that the velocity increment per channel is very small and, therefore, a peak of a given width is recorded over a relatively large number of channels. Without appreciable loss of information, under these conditions, the spectrum could alternatively be represented by half as many data points where each point represents the sum of two consecutive channels. For the statistical quality of the peak to be the same in either data set, the ratio derived above must take into account both the peak width and the MCA velocity increment per channel, or specifically:

$$\Xi = (N_\infty - N_0)\sqrt{G/N_\infty} \qquad (2)$$

where Ξ is the herein defined statistical quality of the peak, and G is the peak width (FWHM) in units of

channel numbers. The peak width in units of mm/sec can be converted to units of channel numbers by dividing by the velocity increment per channel, or by rewriting the formula as

$$\Xi = (N_\infty - N_0)\sqrt{\Gamma/\nu N_\infty} \tag{3}$$

where Γ is the peak width in mm/sec and ν is the velocity increment per channel also in mm/sec.

The fractional peak dip, p, can be expressed as $p = (N_\infty - N_0)/N_\infty$ leading to another useful form of Ξ, *viz*

$$\Xi = p\sqrt{N_\infty G} \tag{4}$$

Finally, for pure Lorentzian peaks, the area of the peak is related to the dip and width parameters; $A = \pi G(N_\infty - N_0)/2$ which then rearranged and combined with Equation (2) above gives:

$$\Xi = \frac{2A}{\pi\sqrt{N_\infty G}} \tag{5}$$

Although experimental conditions employed in the measurement of spectra vary widely, it is possible to give typical ranges of the significant spectral parameters and thereby arrive at the range of expected peak Ξ values. Background or off-resonance count is largely determined by the length of time the spectral measurement is made and the activity of the source. Currently reported spectra have background counts on the order of 10^6 counts per channel which represents an order of magnitude increase over typical spectra reported five years ago. The fractional dip observed with mineral samples depends primarily upon the Fe concentration of the sample. The amount of sample used, however, is limited by other effects such as absorption, saturation, or thickness broadening. Typical Mössbauer-effect dips are on the order of 5 to 10 percent for the major peaks in a spectrum but may be less than 1 percent in the case of sites with small Fe occupancies. Mössbauer peakwidths reported for minerals show a narrow range of about 0.30 ± 0.05 mm/sec, although broadened peaks are occasionally encountered. The velocity increment employed depends on the size of the MCA and the necessity to record the entire spectrum simultaneously. As the resolution limit is fixed by the peak width, there is little or no advantage in using a velocity increment that is very small compared to the peakwidth. Literature-reported values are normally in the range of 0.01 to 0.05 mm/sec/channel, *i.e.*, peaks appear some ten to twenty channels wide.

Combining these typical parameter ranges leads to typical statistical peak qualities, Ξ, on the order of 100–200. Peaks with higher values are exceptionally well determined by the data whereas peaks of Ξ values significantly less than 100 are only indistinctly resolved in the given spectrum.

It remains to demonstrate that Ξ is in fact a measure of the statistical quality of a spectral peak. As mentioned above, the statistical quality is a measure of how well the peak parameters are defined by a given data set, *i.e.*, how small the errors are in the determination of the peak parameters from that data set. Thus the higher the statistical quality, the higher the precision with which the spectral parameters can be determined or conversely the smaller the probable error in the estimates of these parameters.

Therefore, it can be expected that

$$\Xi \propto 1/\epsilon_p \tag{6}$$

where ϵ_p is the relative error in the spectral parameter under consideration. To demonstrate that this proportionality holds, a series of Mössbauer spectra were numerically simulated as follows. The expression for the number of counts accumulated in the ith channel is

$$N_{(i)} = N_\infty - \frac{2A/\pi G}{1 + 4(N_{(i)} - x_0)^2/G^2} + N_\infty^{1/2} R(0, 1) \tag{7}$$

where x_0, A, and G are, respectively, the location, area, and width of the peak, and $R(0, 1)$ is a normally distributed random variable with mean of zero and variance of 1.0. The normally distributed random variables were computed by the method of Hamming (1962). Spectra were simulated with peak widths of from 2 to 60 channels and Ξ values from 6 to 750. These values span the range of most spectra normally encountered.

These single-peak, simulated spectra were then fit by the usual (Newton-Gauss) least-square methods which yield estimated standard errors, σ_p, for each of the fitted parameters. The corresponding relative standard errors, $\epsilon_x = \sigma_x/G$, and $\epsilon_G = \sigma_G/G$ are shown plotted in Figure 2 *versus* the inverse of the appropriate Ξ value for that spectral peak. (The data points for the relative error in the area $\epsilon_A = \sigma_A/A$ which outline a similar curve falling between those shown, were deleted in the interest of graphical clarity.) The plot shows that indeed the relative errors are inverse linear functions of the Ξ values, *i.e.*:

$$\epsilon_p \equiv (\sigma_p/P) = k_p/\Xi \tag{8}$$

where k_p is a constant. From Figure 2 the values of these proportionality constants can be evaluated; for the relative error in peak location, $k_x \approx 0.81$, for the relative error in peak area, $k_A \approx 1.63$, and for the relative error in the peak width, $k_G \approx 2.23$.

The Ξ values of Figure 2 were calculated from the ideal input spectral parameter values. The increased spread of points at small Ξ values is due in part to the small difference between the Ξ values obtained in this manner and the Ξ values obtained by using the least-squares fitted values of area, width, *etc*, and in part due to the imprecision of the error estimates of small Ξ value peaks.

Thus from Figure 2 or the algebraic expressions, the probable error in the determination of the various spectral parameters of *isolated* peaks can be determined from a knowledge of their Ξ values. The observed parameter esd's for a number of isolated peaks from actual spectra were checked against these relations and excellent agreement was found.

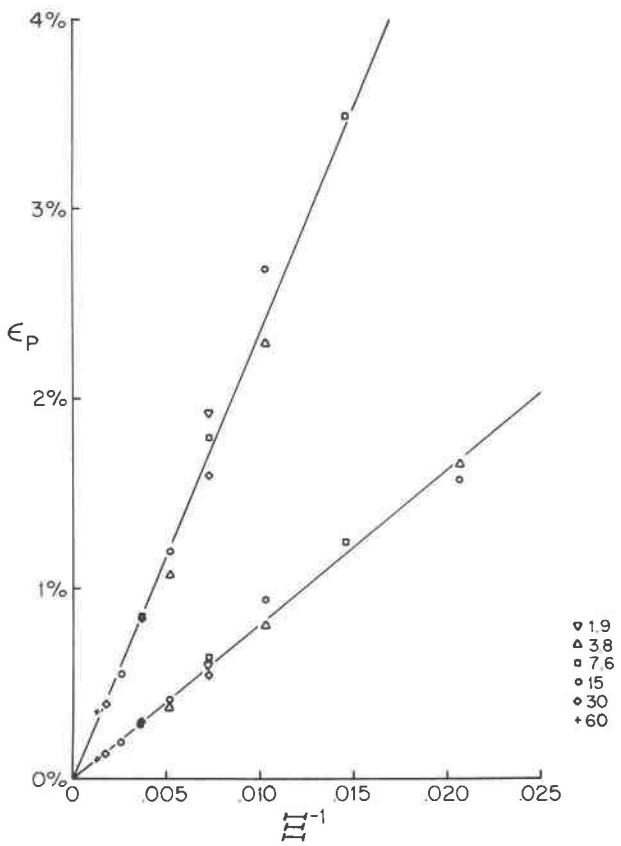

FIG. 2. Plot of relative error of peak parameters *versus* inverse of peak Ξ value. Symbols refer to various peak widths employed. Upper line and associated data points refer to relative error in peak width, lower line and associated data points refer to relative error in peak location. Relative error in peak area (not shown) falls in between.

Ruby (1973) has recently introduced several new parameters useful in characterizing Mössbauer spectra, including a parameter, S, which, "has the properties desired for a figure of merit or for a measure of the quality of a (spectral) measurement." This S value is a characteristic of the entire spectrum and therefore is closely related to the component peak Ξ values. Ruby defines S,

$$S = N_\infty \sum^{l} p(i)^2 \tag{9}$$

where $p(i)$ is the fractional dip observed in the ith channel, and the summation extends over the l channels comprising the spectrum. For a Lorentzian peak of maximum dip, p_0, the fractional dip observed in any channel is given by

$$p(i) = p_0/(1 + 4\delta^2/G^2) \tag{10}$$

where δ is the separation of the ith channel from the peak maximum and G is the peak width, both in units of channel-numbers. In a spectrum with peaks that are many channels wide, one of the channels will be sufficiently close to the center of the peak to be considered to show the maximum fractional dip of p_0, *i.e.*, $\delta \approx$ zero, for this channel. For successive neighboring channels, $\delta = \pm 1, \pm 2, \pm 3 \ldots$ Then:

$$S = N_\infty \sum^{l} (p_0/1 + 4\delta^2/G^2) \tag{11}$$

or

$$S = N_\infty p_0^2 \{1 + 2 \sum^{\infty} 1/(1 + 4n^2/G^2)^2\} \tag{12}$$

The value of the infinite series in curly braces can be numerically evaluated and is equal to $0.25\pi G$. Then:

$$S = 0.25\pi p^2{}_0 N_\infty G$$

or in terms of Ξ,

$$S = 0.25\pi\Xi^2 \tag{13}$$

for a single-peak spectrum. For a multipeak spectrum, two extreme cases must be isolated. For non-overlapping peaks the contribution to S of the ith channel is due to one or another of the component peaks (but not more than one) and therefore is included in one of the component peak Ξ values, thus:

$$S(\text{non-overlapped}) = 0.25\pi\Sigma\Xi(i)^2$$

However, for overlapped spectra, the dip observed in the ith channel is due to more than one component peak. In the extreme case of complete overlap of two peaks, $p_0 = p_a + p_{b'}$, and from above:

$$S(\text{completely overlapped}) = 0.25\pi[\Sigma\Xi(i)]^2$$

In general, then, S lies between these two extremes:

$$0.25\pi[\Sigma\Xi(i)]^2 \geq S \geq 0.25\pi\Sigma\Xi(i)^2$$

depending upon the degree of overlap of the component peaks in a particular spectrum.

Effect of Peak Overlap on Errors in Parameter Determination

In Figure 1, the separation of the peaks is large enough that their characteristic parameters can be determined essentially to the precision given by Equation (8). However, for spectra with successively smaller separations, the overlap of one distribution on the other decreases the precision with which all parameters of the two peaks can be determined. Consider the envelope formed from the sum of two (Lorentzian shaped) peaks which are moved closer and closer together. The separation of the envelope maxima is shown in Figure 3 as a function of the separation of two equal height, equal width peaks. At a peak separation of several peak widths (such as shown in Fig. 1) the envelope consists of two maxima with a local minimum at the midpoint between the peaks. The envelope maxima are located essentially at the component peak maxima and the envelope heights at the maxima are only slightly greater than the heights of the peaks. As the peaks are moved closer together, (1) the heights of the envelope maxima rise, but (2) the height of the midpoint minimum rises faster and (3) the location of the envelope maxima moves from directly over the peaks towards the peak midpoint. At a critical peak separation (equal to $1/\sqrt{3}$ peak widths for equal height peaks) the two envelope maxima have coalesced into a single maximum located at the component peak midpoint. Moving the peaks still closer merely increases the height and decreases the width of the single envelope maximum. Since the sum of two equal-width Lorentzian peaks of area A_1 and A_2 located at the same point is identical to a single Lorentzian peak of area $A_1 + A_2$, the single maximum looks more and more like a single peak as the component peak separation is decreased towards zero. It is in this region of peak separations (below $\sim 0.6\ \Gamma$) where the eye no longer discerns separate maxima, that there is a rapid decrease in the precision with which all peak parameters can be determined.

As above for the single-peak spectra, a larger number of two-peak spectra were simulated on an IBM 360-91 at UCLA and on a UNIVAC 1108 at the University of Wisconsin, Madison. For these spectra the normally distributed random numbers were generated using the Hamming (1962) method for the spectra simulated on the IBM computer and the Marsaglia, McLaren, and Bray (1967) procedure on the Univac computer. About 150 total spectra were generated. These spectra had Δ values (peak separations expressed in units of peak widths) of 0.0 to 2.0 and Ξ values from 8 to 200. Several different ratios between the areas of the two peaks were used as well as several different peak widths. In all cases the generating values of the peak widths of the two component peaks were kept equal. This is in keeping with real spectra where drastically different peak widths in the same spectra are the exception.

The simulated two-peak spectra were then fit (or attempted) by the usual (Newton-Gauss) full-matrix least-squares methods. Starting parameters were usually rough approximations of the ideal generating values but it was found that the number of steps to convergence and general least-squares refinement behavior was the same even if the exact input values were used as starting parameters. Refinement was done with two models, the first with no parameter constraints, *i.e.*, the location, area, and widths of both peaks were simultaneously refined, and in the second model the widths of both peaks were constrained to be equal.

The behavior of the refinement procedure depends

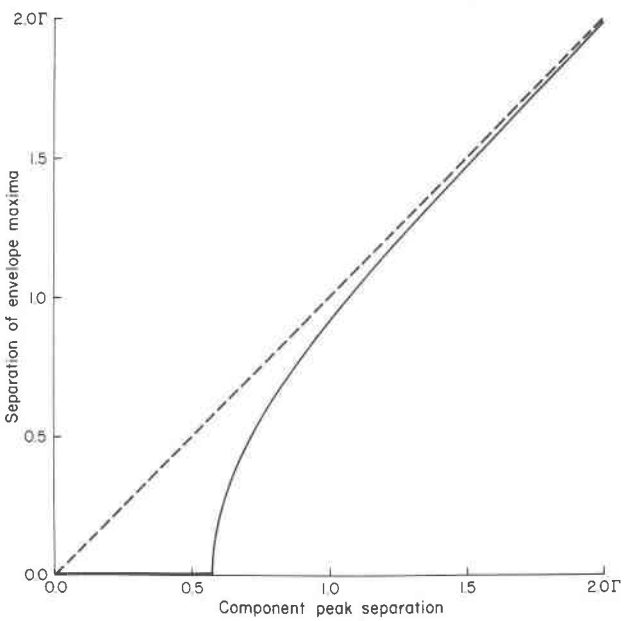

FIG. 3. Separation of maxima of the envelope formed by two partially coalesced equal-area, equal-width Lorentzian peaks, as a function of component peak separation. Separations are given in units of peak widths.

on the relative peak separation, Δ. For Δ values greater than 0.6 Γ, all simulated cases smoothly and rapidly converged. The convergence criterion used was that all parameter changes on the "final" cycle must be less than 2 percent of their respective estimated standard errors. This criterion is in fact much more severe than normally employed, but was deemed necessary in a few critical cases.

For Δ values below $\sim$ 0.6 Γ, convergence behavior depended upon the Ξ values as well. For a series of spectra with peaks having similar Ξ values but with steadily decreasing peak separations, the number of cycles to reach convergence increased. As the Δ values decrease, the parameter values tend to oscillate roughly about the input or ideal values, and for small enough Δ values these oscillations increase on successive least-squares cycles. That is, the fit between observed number of counts and calculated values becomes worse. As the magnitude of the standard errors of the spectral parameters becomes large compared to the parameters themselves, the probability of divergence increases rapidly. As the relative parameter errors depend on the peak Ξ values, then this oscillation and divergence behavior occurs at larger peak separations when dealing with smaller peaks. The exact cut-off point, beyond which a unique least-squares solution cannot be found, depends on Δ and Ξ values, the number of constraints, the particular statistical noise distribution, and the specific mathematical routines employed.

Figure 4 summarizes the relation of relative error, peak separation, Δ, and peak quality, Ξ. Holding for the moment the Ξ values of the peaks constant, Figure 4 may be interpreted as a simple plot of relative spectral parameter error *versus* peak separation. Note that on a semilog scale, as Δ decreases below $\sim$ 0.6 (peak widths), the probable error in determining the various peak parameters increases spectacularly.

In Figure 4 the solid points refer to cases where the least-squares solution converged. The open points refer to error estimates from least-squares solutions which diverged. Ordinarily, unless convergence occurs, *i.e.*, unless the parameter changes decrease to zero, the least-squares procedure cannot supply correct parameter error estimates. However, if the cor-

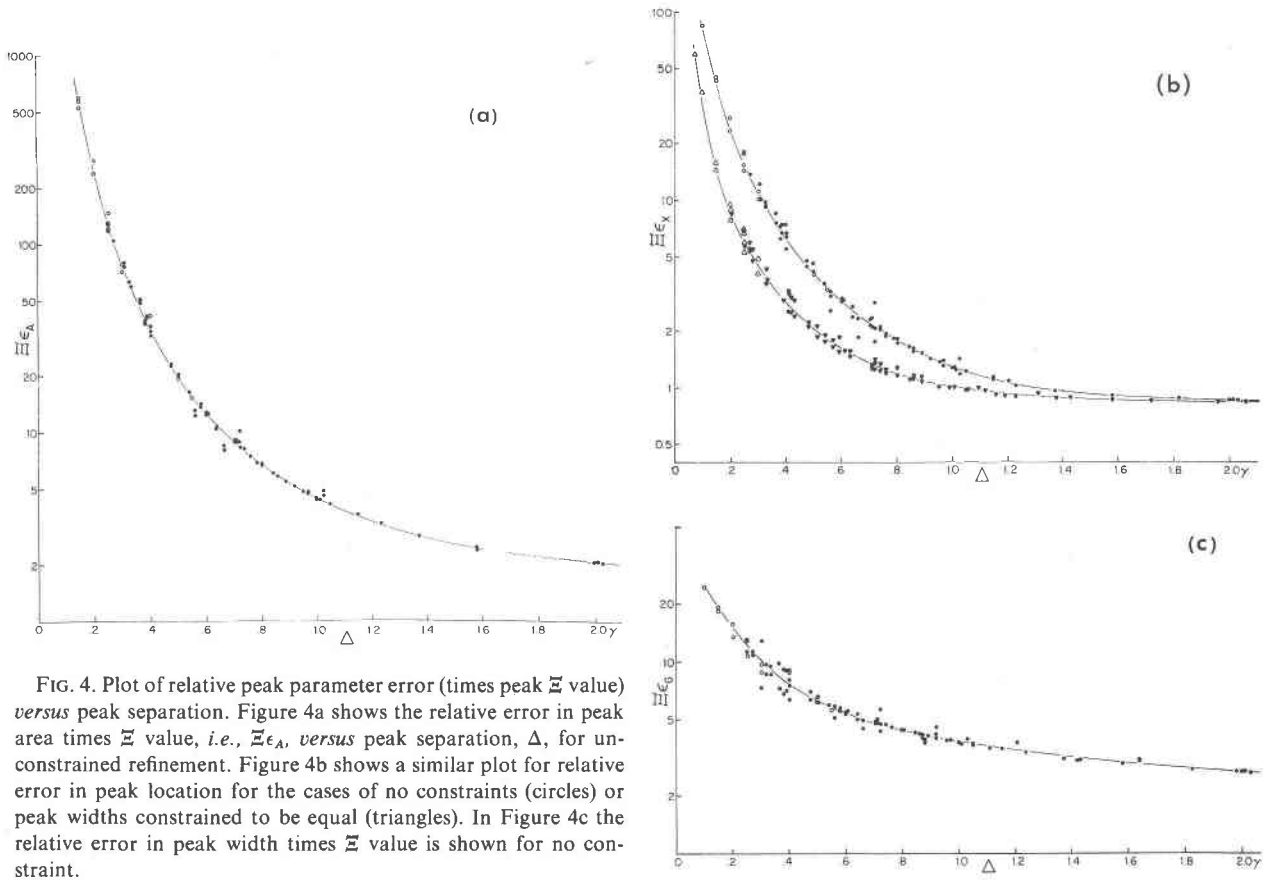

FIG. 4. Plot of relative peak parameter error (times peak Ξ value) *versus* peak separation. Figure 4a shows the relative error in peak area times Ξ value, *i.e.*, $\Xi\epsilon_A$, *versus* peak separation, Δ, for unconstrained refinement. Figure 4b shows a similar plot for relative error in peak location for the cases of no constraints (circles) or peak widths constrained to be equal (triangles). In Figure 4c the relative error in peak width times Ξ value is shown for no constraint.

rect parameter values are known (as is this case where the ideal generating values are specified), then the errors furnished by a least-squares cycle starting with the correct parameters are good approximations of the true errors. It can be seen (Fig. 4) that the open points plot along with the closed points and allow extrapolation to very small Δ values where no cases of convergence occurred.

In further explanation of the application of Figure 4, consider a peak, of $\Xi \simeq 50$, overlapped partially by a second peak. If it is desired to determine the relative area of this first peak to $\pm$ 10 percent, then the question arises how close may the overlap between the two peaks be? For this case $\epsilon_A = \sigma_A/A = 0.1$ and the product $\epsilon\ \Xi = 5$. Figure 4b shows that the two peaks must be separated by at least 0.9 Γ (if the individual Γ values are independently determined) in order for the relative error to be less than 10 percent. If the peak in question has a Ξ value of about 200, then $\epsilon\ \Xi = 20$, and separations of $\Delta = 0.5\ \Gamma$ or more will yield area estimates to better than 10 percent for no constraints.

With spectra displaying poorly defined peaks (low Ξ values) the estimates of the parameters of the weak peak are imprecise and, furthermore, the estimates of the probable *errors* in these parameters furnished by the least-squares method are then also imprecise. In Figure 4 the relative error estimates are plotted only for those simulated spectra with peaks having Ξ values of 20 or greater. Error estimates from weaker peaks are less precise and consequently would show a significantly greater spread in such a plot. Use of such imprecisely determined error estimates (rather than the true estimates which can be obtained from Fig. 4) may be misleading. As an example, a spectrum was generated from two equal area peaks each having Ξ values of 100 and a separation of $\Delta = 0.3\ \Gamma$. This simulated spectrum was least-squares fit with two peaks, without constraint. The fit converged with the two best-fit peaks having strongly dissimilar areas and widths. Because of the small Δ value the fit is highly imprecise, and one of the peaks had a best-fit Ξ value of only 14. The error estimates of the parameters of both peaks then were very imprecisely determined by the least-squares procedure and were in this case severely underestimated. In fact, by using the least-squares-furnished error estimates, it could be shown that the difference in peak areas between the two fitted peaks amounted to six sigma, *i.e.*, statistically highly significant. If the correct error estimates were taken from Figure 4, the difference between the fitted peak areas was found to be about one sigma, *i.e.*, statistically insignificant.

Regardless of overlap, the parameters of the envelope remain defined. Therefore, if it is possible to specify any relationship (constraint) between the parameters of the component peaks, the precision with which the remaining parameters are defined increases. The effect of constraints of the peak widths is illustrated in Figure 4b. The lower set of points (triangles) shows the (Ξ-scaled) expected errors in peak location *versus* peak separation for spectra with peak widths constrained to be equal. Note that the expected errors are in all cases smaller than for the unconstrained refinement or, put another way, that for the same size probable error, a more severe peak overlap may be tolerated. A similar "smaller error" curve for expected area errors, when peak widths are constrained, can be drawn. For constrained peak widths, the expected error in the peak width itself is essentially independent of overlap since the width of the envelope remains well defined.

Application to Actual Spectra

The foregoing treatment, including Figure 4, is applicable to real (as opposed to simulated) spectra. The actual errors in real spectra, however, are larger than those due to counting statistics alone. The additional sources of parameter imprecision include:

(a) systematic errors in the data due, *e.g.*, to spectrometer drift, thickness or cosine line-broadening, and others;
(b) errors in the model, including wrong number of peaks, deviations from Lorentzian form;
(c) overlap of more than two peaks;
(d) simultaneous refinement of one or more background parameters.

The magnitude of the errors derived above from simulated spectra then must be considered lower limits of the errors expected with real spectra. Nevertheless, with weak or closely overlapped peaks the error due to counting statistics is likely to be the dominant source of parameter imprecision.

Examples of real spectra fit with peaks having separation of less than $\sim 0.6\ \Gamma$ (*i.e.*, separate maxima not discernible to the eye) are abundant in mineralogical studies. A typical example is the fitting of two (ferrous-iron) doublets to the spectrum of Trelavour biotite by Hogg and Meads (1970). From their illustrated spectrum the following can be evaluated: $N_\infty = 2.2 \times 10^6$, $p \approx 3.9$ percent, $v \approx 1/28$ mm/sec, $\Gamma \approx 0.39$ mm/sec, and thus Ξ values are in the range of 100 to 150. Component peak separations are about 0.51 Γ and thus from Figure 4a the uncer-

tainty in the area of each peak is about 13 to 19 percent of its area. Consequently, site occupancy ratios computed from such peak areas have similarly high uncertainty levels. This example is not extreme in that it represents a higher off-resonance count and wider peak separation than many. In cases where the component peaks (either real or imagined) are more closely overlapped, least-squares convergence is often only obtained through the mechanism of constraining peak widths and/or areas, as discussed above. Obviously, whenever strongly overlapped peaks are fitted, both author and reader should proceed with caution as well as a generous uncertainty estimate.

Acknowledgments

The manuscript benefited from the comments of Mr. Gary I. Sherwood. The study was facilitated by the hospitality of Professor S. W. Bailey, University of Wisconsin, and was supported by Grant GA-32268 from the National Science Foundation.

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Manuscript received, December 26, 1973; accepted for publication, November 15, 1974.