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THE TREATMENT OF WEAK INTENSITIES OF REFLEXION

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Résumé - Bien que les intensités faibles posent des problèmes particuliers lors de la détermination des structures cristallines, elles portent au moins autant d'informations que les intensités fortes et ne doivent donc pas être ignorées /1,2,3,4,5/. Une réflection que est en vérité nulle ou faible peut être mesurée négative parce qu'une intensité est estimée par la différence entre les comptages dans le pic et les comptages dans le bruit de fond et parce que chaque comptage est sujet à des fluctuations statistiques (Poisson); dans le cas de la technique de comptage à temps constant la distribution de probabilité de cette différence est une fonction de Bessel modifiée /2,6/. Les méthodes Bayésiennes /3/ peuvent être utilisées, avec quelques approximations, pour obtenir une estimation vraisembable de la valeur positive correspondante. Toutefois les fluctuations statistiques introduisent un biais lorsqu'on prend la racine carrée de l'intensité pour obtenir le facteur de structure. La correction du biais est facile si l'intensité est modérée /7/ mais pose un problème si les intensités sont faibles /8,4/. On doit distinguer précision - la reproductibilité statistique de la détermination - et justesse - la capacité à approcher la vraie valeur de la quantité cherchée. Lorsque les erreurs ou défauts systématiques du modèle sont appréciables, la justesse de la détermination peut être bien plus faible que sa précision (/19/; comparer aussi /4/ et /10/); la légitimité statistique de la procédure classique d'"ajustement de la pondération" est douteuse.

Abstract - Though weak intensities present special problems in crystal-structure determination, they convey at least as much information as strong intensities, and should not be ignored /1,2,3,4,5/. Since an intensity is estimated as the difference between counts on peak and counts on background, and each count is subject to statistical (Poisson) fluctuations, a reflexion that is actually zero or small may be measured as negative; the probability distribution of the difference is given by a modified Bessel function /2,6/ for the fixed-time counting technique. With some approximation, Bayesian methods /3/ may be used to estimate the likely positive value in such cases. The statistical fluctuations, however, result in a bias when the square-root of the intensity is taken in order to obtain the structure factor. Correction of the bias is easy when the intensity is moderate /7/, but gives problems for weak intensities /8,4/. Precision, the statistical reproducibility of a determination, must be distinguished from accuracy, the extent to which a determination approaches the true value of the quantity sought. When systematic errors or defects in the structural model are appreciable, the accuracy of a determination may be much less than its precision (/9/; compare also /4/ and /10/); the common procedure of 'adjusting the weights' is of dubious statistical legitimacy.

I - INTRODUCTION

When I began to work in crystallography, in 1938, it was a common joke among research students, or rather only half a joke, that actual measurement of intensities was a waste of effort. If a structure had m parameters, then noting m accidental absences would give enough information to solve the structure and determine the parameters. I believe that one or two alloy structures, considered complicated at the time, were in fact solved by A.J. Bradley's group by the use of this method, but I have not been able to find a reference to it in the literature. Be that as it may, the weak reflexions convey at least as much information as an equal number of strong ones, and should not be ignored.

It is, or was, a common practice to omit from structure refinements all reflexions for which the observed intensity was less than two (or even three) times its standard deviation. There seems never to have been any formal theoretical justification for this. It is, of course, understandable that intensities measured as negative were omitted, since they indicated imaginary structure factors, but the rule of thumb of 2σ or 3σ eliminated many positive ones also. Various authors /1,2,3,4,5/ have argued [1] on various grounds for the inclusion of weak reflexions, and French and K. Wilson /3/ have proposed a method of dealing with those measured as negative.

The recommendation of Hirshfeld and Rabinovich /1/, to include in the refinement all intensities at their measured values, even if negative, is satisfactory for the determination of parameters by least squares, maximum likelihood, and similar methods. However, in many crystallographic studies, such as electron-density Fourier syntheses or difference Fourier syntheses, it is necessary to have estimates of the structure factors and their standard deviations, and this would appear to rule out the possibility of using measured-as-negative intensities. "Fortunately the problems are almost entirely due to poor statistical methodology. Instead of thanking the data for the information that certain structure factor moduli are small, we accuse them of assuming 'impossible' negative values. What we should do is combine our knowledge of the non-negativity of the true intensities with the information concerning their magnitude contained in the data /3/." The French and Wilson method will be discussed fully in section V below.

Measurements are normally subject to two types of error: random errors and systematic errors. The random errors (for example, statistical fluctuations in counting rates) affect the reproducibility of a measurement, technically called its precision. The systematic errors (for example, uncorrected extinction) introduce a displacement of the measured value from the true value, the amount of the displacement being the same for all repetitions of the measurement; the random errors then fluctuate about the displaced value, not the true value. The degree to which the displaced value approaches the true value is called the accuracy of the measurement. A measurement may thus be highly precise and simultaneously highly inaccurate. Crystal-structure determinations may also be subject to systematic errors in calculated quantities (for example, inadequate atomic scattering factors). A third type of error is bias, a systematic error in a derived quantity resulting from inappropriate or inexact treatment of the raw data.

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[1] A paper by Petit, Lenstra and Van Loock /11/ might, at a careless reading, be taken as advocating the opposite. It deals, however, with a means of economizing computer time before the structure is completely determined, and ends with an endorsement of the recommendations of Hirshfeld & Rabinovich /1/ for the final refinement.

The present paper reviews a series of topics that may at first sight seem unrelated. The connecting threads are (i) the problems are most acute for weak reflexions, though some of them affect strong reflexions also, and (ii) all are statistical. We consider in turn bias, origin of negative observed intensities, Bayes' theorem, the French and Wilson method, and precision versus accuracy.

II - BIAS IN STRUCTURE DETERMINATION

The origin of bias is mathematical rather than experimental; it is 'Unnoticed inappropriateness of mathematical techniques, whereby random errors, of mean value zero in the raw data, become a systematic bias in the derived quantities' /12/. Intensities measured by counting are in principle unbiased. This property of absence of bias is preserved by any linear transformation, such as (i) subtraction of background; (ii) division by counting time to obtain a counting rate; and (iii) division by trigonometrical factors; but is lost if any non-linear transformation is applied to the observed intensity. The two non-linear transformations commonly used in crystallography are (i) taking the square root of the intensity in order to obtain the modulus of the corresponding structure factor /7/; and (ii) using weights that depend on the intensity or its square root /12,4/.

The 'observed' structure factor can be written

$$\underline{F}_o = \underline{I}^{1/2} = [\underline{J} + \underline{e}]^{1/2} , \qquad (1)$$

where $\underline{I}$ is the observed intensity, $\underline{J}$ is the 'true' intensity, and $\underline{e}$ is the statistical fluctuation. If the fluctuation is small in comparison with the intensity, the square root can be expanded as a power series:

$$\underline{F}_o = \underline{J}^{1/2} + (1/2)\underline{J}^{-1/2}\underline{e} - (1/8)\underline{J}^{-3/2}\underline{e}^2 + \ldots ; \qquad (2)$$

the mean value of $\underline{e}$ in the second term is zero, but the mean value of $\underline{e}^2$ is the variance of $\underline{I}$. A better estimate of $\underline{F}$ is therefore

$$\underline{F}_{corr} = \underline{I}^{1/2} + (1/8)\underline{I}^{-3/2}\sigma^2(\underline{I}) + \ldots . \qquad (3)$$

Unfortunately this correction fails for small observed intensities, for which the methods of Rees /8/ and French and Wilson /3/ may be applicable. This bias, if uncorrected, would be expected to lead to some loss of definition in electron-density maps, somewhat low values of scaling factors, and somewhat high values of thermal parameters. A fuller discussion and many references will be found in /4/.

The bias in parameters introduced by the use of weights depending on the observed and/or calculated intensity may be seen in the following way. In least-squares refinement the parameters are determined by minimizing the sum

$$\underline{S} = \sum \underline{w}|\underline{I}(\underline{hkl}) - \underline{G}(\underline{hkl})|^2 , \qquad (4)$$

where $\underline{G}$ is the calculated intensity and $\underline{w}$ is a weight, by differentiating $\underline{S}$ with respect to each of the parameters in turn and equating the results to zero. Let $\underline{x}$ be the amount by which some desired parameter differs from its unbiased value. The sum $\underline{S}$ may be expanded in powers of $\underline{x}$:

$$\underline{S}(\underline{x}) = \underline{S}(0) + \underline{S}_{\underline{x}}(0)\underline{x} + \underline{S}_{\underline{xx}}(0)\underline{x}^2/2! + \ldots , \qquad (5)$$

where the subscripts denote differentiation and the derivatives are evaluated at $\underline{x} = 0$. The value of $\underline{x}$ giving minimum $\underline{S}$ is not 0 but

$$\underline{x} = - \underline{S}_{\underline{x}}/\underline{S}_{\underline{xx}} + \ldots , \tag{6}$$

and the minimum value of $\underline{S}$ is not $\underline{S}(0)$ but

$$\underline{S}_{min} = \underline{S}(0) - \underline{S}_{\underline{x}}^2/2\underline{S}_{\underline{xx}} + \ldots . \tag{7}$$

The calculated intensity $\underline{G}$ is a function of $\underline{x}$, and in some programs $\underline{w}$ may depend on $\underline{G}$. Since $d\underline{w}/d\underline{x} = (d\underline{w}/d\underline{G})(d\underline{G}/d\underline{x})$, differentiation of (4) gives

$$\underline{S}_{\underline{x}} = \sum [\underline{w}_{\underline{G}}\underline{G}_{\underline{x}}(\underline{I}-\underline{G})^2 - 2\underline{w}(\underline{I}-\underline{G})\underline{G}_{\underline{x}}] , \tag{8}$$

$$\begin{aligned} \underline{S}_{\underline{xx}} = \sum [& \{\underline{w}_{\underline{GG}}\underline{G}_{\underline{x}}^2 + \underline{w}_{\underline{G}}\underline{G}_{\underline{xx}}\}(\underline{I}-\underline{G})^2 \\ & - \{4\underline{w}_{\underline{G}}\underline{G}_{\underline{x}}^2 + 2\underline{w}\underline{G}_{\underline{xx}}\}(\underline{I}-\underline{G}) \\ & + 2\underline{w}\underline{G}_{\underline{x}}^2] . \end{aligned} \tag{9}$$

The important term [2] in the expression for $\underline{S}_{\underline{xx}}$ is the final one, since the others vanish with $(\underline{I}-\underline{G})$, while it remains practically constant. For a well refined structure it is therefore sufficient to write

$$\underline{S}_{\underline{xx}} = 2 \sum \underline{w}\underline{G}_{\underline{x}}^2 . \tag{10}$$

The expression for $\underline{S}_{\underline{x}}$ needs closer attention. If $\underline{e}$ is the actual statistical fluctuation in a particular observation, and $\underline{d}$ is the actual amount by which the calculated intensity differs from the true value (because of defects in the model etc.), $\underline{I} - \underline{G} = \underline{e} - \underline{d}$, and Equation (8) becomes

$$\underline{S}_{\underline{x}} = \sum [\underline{w}_{\underline{G}}(\underline{e}^2 - 2\underline{e}\underline{d} + \underline{d}^2) - 2\underline{w}(\underline{e}-\underline{d})]\underline{G}_{\underline{x}} . \tag{11}$$

Depending on the refinement program used, the weights may involve the observed or the calculated intensities -- or both. Expanding $\underline{w}$ as a power series in $\underline{e}$ and $\underline{d}$, taking expected values and using Equation (6) ultimately gives for the expected value of the parameter

$$\langle \underline{x} \rangle = - [2 \sum \underline{w}\underline{G}_{\underline{x}}^2]^{-1} \sum [(\underline{w}_{\underline{G}} - 2\underline{w}_{\underline{I}})\langle \underline{e}^2 \rangle + 3\underline{w}_{\underline{G}}\underline{d}^2 + 2\underline{w}\underline{d} + \ldots]\underline{G}_{\underline{x}} . \tag{12}$$

The terms in $\underline{d}$ express the errors in the parameter resulting from defects in the model. That resulting from statistical fluctuations is proportional to

$$(\underline{w}_{\underline{G}} - 2\underline{w}_{\underline{I}})\langle \underline{e}^2 \rangle = (\underline{w}_{\underline{G}} - 2\underline{w}_{\underline{I}})\sigma_{\underline{I}}^2 ; \tag{13}$$

we see that the bias resulting from using calculated intensities in the weights is of the opposite sign to the bias resulting from using observed intensities. The bias can thus be removed, to the second order in $\underline{e}$, by using the weighted mean $(\underline{I} + 2\underline{G})/3$ instead of $\underline{I}$ or $\underline{G}$ in any expression for the weights /12/.

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[2] There is a misplaced superscript in the corresponding equation in reference /12/. This does not affect any of the subsequent equations.

Does omission of observations introduce bias ? Prince and Nicholson /5/ argue that 'Actually, omitting a weak reflection, or any [3] reflection, cannot bias the parameter estimates'. One can accept this immediately for reflexions omitted at random, but one wonders about systematic omissions, of discrepant reflexions in the context of robust/resistant methods /9/, or of weak reflexions in the present context. It seems unlikely, for example, that the systematic omission of weak reflexions is without effect on the thermal parameters.

III - ORIGIN OF NEGATIVE MEASURED INTENSITIES

In the absence of disturbing influences, the number of counts recorded during the counting interval in the fixed-time mode fluctuates in accordance with the Poisson probability distribution; the probability of observing N_o counts is

$$p(N_o) = \exp(-N)N^{N_o}/N_o! \ , \tag{14}$$

where N is the true number of counts to be expected. The measured intensity of a reflexion depends on the difference (say R) between the 'true' number of counts T expected when the diffractometer is set to receive the reflexion, and the 'true' number of counts B when the diffractometer is set to receive the immediate background:

$$R = T - B \ ; \tag{15}$$

for simplicity it is assumed that the counting times for reflexion and background are the same. The observed values T_o and B_o will fluctuate in accordance with Equation (14), so that the observed value R_o will sometimes be negative, especially when T is about the same size as B -- that is, for weak reflexions. It is easy to write the formal expression for the probability of any particular value of R_o:

$$p(R_o) = \sum p(T_o)p(B_o) \ , \tag{16}$$

the summation being over all values of B_o and T_o related by

$$R_o = T_o - B_o \ . \tag{17}$$

The summation has been carried out by Skellam /13/, and leads to an expression in terms of modified Bessel functions of the first kind:

$$p(R_o) = \exp[-(B+T)](T/B)^{R_o/2} I_{|R_o|}[2(BT)^{1/2}] \ . \tag{18}$$

If B and T are not too small this does not differ very greatly from a normal distribution with mean R and variance

$$\sigma^2(R) = T + B \ ; \tag{19}$$

it is, however, somewhat skew, has a positive 'excess', and large values of R_o, positive or negative, are rather more likely than for a normal distribution. If the counting times for reflexion and background are not equal, or if fixed-count rather than fixed-time methods are used, the distributions become more complicated /2,6/. The differences from normality seem to remain as described for equal-time counting.

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[3] Italicized in the original.

IV - BAYES' THEOREM [4]

The combination of information about the probability distribution of intensities with the information that a certain reflexion has been measured and found to have the value I is made by the use of Bayes' theorem. Bayes' theorem is of very general application, but for concreteness in the present context it will be expressed in terms of intensity distributions. It is probably best to state it in words before putting it in mathematical form. Before making a measurement we expect the probability density distribution of the true intensity J to have a certain form, for example the centric or the acentric distribution /16/, or the distribution appropriate for the space group and chemical constitution /17,18/; this is called the a-priori probability distribution of J. We make the measurement I by a method known to be subject to statistical fluctuations, the probability density distribution of the fluctuations being known as a function of the intensity, for example a normal distribution with mean J and known variance σ^2, or the distribution discussed in the preceding section -- or one even more complex /6/. The measurement having been made, the theorem asserts that the probability distribution of the true intensity is proportional to the product of (i) the probability of getting I when the true intensity is J, and (ii) the a-priori probability of the intensity J. The result is called the a-posteriori probability density distribution of J.

Since French and Wilson /3/ have been the main proponents of this procedure it is convenient to adopt their symbols as far as practicable. We shall represent probability distributions by $P_x(.)$, the subscript indicating the quantity that is the principal variable in that distribution; other quantities appearing are parameters of the distribution for the time being. A vertical bar followed by the symbol of a quantity x is read 'given that the variable x has the particular value x'. We thus have, for the a-priori distribution of the true intensity J:

$$P_J(J)dJ \ , \tag{20}$$

for the distribution of the measured intensity I, given J:

$$P_I(I|J)dI \ , \tag{21}$$

and for the a-posteriori probability distribution of the true intensity J:

$$P_J(J|I)dJ = KP_I(I|J)P_J(J)dJ \ , \tag{22}$$

the proportionality constant K being determined by the fact that the total a-posteriori probability is unity:

$$K \int_0^\infty P_I(I|J)P_J(J)dJ = 1 \ . \tag{23}$$

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[4] Bayes' theorem is a simple application of the multiplication rule for probabilities [for a derivation see, for example, /14/, pp. 212-218], and is disputed by no one. Bayes' postulate, the Principle of the Equidistribution of Ignorance, makes many statisticians see red. Both are traced back to an early paper by Bayes /15/, called An essay towards solving a Problem in the Doctrine of Chances. It is rather difficult to extract them from the mass of verbiage in the original.

V - THE FRENCH AND WILSON PROCEDURE

French and Wilson /3/ take as the intensity to be used in the structure determination the expected value of $\underline{J}$ from Equation (3):

$$\langle \underline{J} \rangle = \underline{E}_{\underline{J}}(\underline{J}|\underline{I}) = \int_0^\infty \underline{J} \underline{P}_{\underline{J}}(\underline{J}|\underline{I}) d\underline{J} , \tag{24}$$

with variance

$$\sigma^2(\underline{J}|\underline{I}) = \int_0^\infty [\underline{J} - \langle \underline{J} \rangle]^2 \underline{P}_{\underline{J}}(\underline{J}|\underline{I}) d\underline{J} . \tag{25}$$

In their procedure they make three assumptions: (i) the data-reduction procedures used are such that $\underline{I}$ is an unbiased estimate [5] ; (ii) that the distribution of $\underline{I}$ is normal with mean $\underline{J}$; and (iii) that the variance of $\underline{I}$ has been properly assessed, with allowance not only for counting statistics, but also for instrumental instability etc. /19,20/. There remains the choice of a-priori distributions for $\underline{P}_{\underline{J}}(\underline{J})$. French and Wilson considered three, all zero for negative values of $\underline{J}$ and having the following forms for $\underline{J}$ positive:

(i) the improper [6] distribution

$$\underline{P}_{\underline{J}}(\underline{J}) = 1 ; \tag{26}$$

(ii) the acentric distribution

$$\underline{P}_{\underline{J}}(\underline{J}) = \Sigma^{-1} \exp(-\underline{J}/\Sigma) ; \tag{27}$$

and (iii) the centric distribution

$$\underline{P}_{\underline{J}}(\underline{J}) = (2\pi\Sigma\underline{J})^{-1/2} \exp(-\underline{J}/2\Sigma) , \tag{28}$$

where Σ is the local average intensity. Both the acentric and the centric distributions, but especially the latter, give a high a-priori probability to weak reflexions.

Obviously computer programs are necessary for carrying out the integrations in Equations (24) and (25); some are described in /3/. With more complex programs these integrations could be performed with specimen-specific a-priori distributions of intensity /18,19/ and non-normal fluctuation distributions /6/.

French and Wilson propose

$$\langle \underline{F} \rangle = \langle \underline{J}^{1/2} \rangle = \underline{E}_{\underline{J}}(\underline{J}^{1/2}|\underline{I}) = \int_0^\infty \underline{J}^{1/2} \underline{P}_{\underline{J}}(\underline{J}|\underline{I}) d\underline{J} , \tag{29}$$

as an almost unbiased estimate of the true structure factor, and

$$\sigma^2(\underline{F}|\underline{I}) = \int_0^\infty [\underline{J}^{1/2} - \langle \underline{J}^{1/2} \rangle]^2 \underline{P}_{\underline{J}}(\underline{J}|\underline{I}) d\underline{J} . \tag{30}$$

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[5] The conditions under which assumption (i) is valid were discussed elsewhere by Tickle /21/ and French /22/.

[6] See the Principle of the Equidistribution of Ignorance in footnote [4].

as an estimate of its variance. French (private communication) believes that there may be a small positive bias in the estimate of the structure factor, but it is clearly a better estimate than that given by Equation (3) when $\underline{F}$ is small.

VI - PRECISION versus ACCURACY

In spite of the intuitively attractive interpretation of the results of unweighted least squares -- the best least-squares fit between the observed and the calculated electron densities (Patterson functions) /23/ -- , the method is not greatly used in structure determination, for the following reason. The intensity measurements are of varying precision; their variances depend in a more or less complicated fashion on the number of counts involved in determining them /2,6,19,20/. It is then statistical practice to weight each term in Equation (4) by a factor inversely proportional to the variance of that term; it can be shown that if this is done the variance of each parameter is less than the variance that it would have if any other method of refinement were adopted [7] . This reduction of the estimated standard deviations of the derived parameters is the first advantage of correctly weighted over unweighted least squares; the second will be discussed in the next paragraph. The disadvantages are (i) the estimation of correct weights is not easy, and (ii) the function fitted by least squares is not the actual Patterson function or the electron density, but a fictitious electron density (Patterson function) /4/ in which each coefficient of the Fourier series is multiplied by the square root of the weight. If the model is perfect, in the sense that it has the same functional form as the true function, and differs from it only in that the parameters have to be adjusted, this distortion of the function will have little effect on the parameter estimates. If the model is defective, however, the estimates will be biased in the direction of fitting the fictitious density, distorted by the weights, rather than being unbiased estimates of the true parameters. Bias introduced by an explicit dependence of the weights on $\underline{I}$ or $\underline{G}$ /12/ has been discussed in section II above.

The second advantage of correctly weighted least squares -- shared with maximum likelihood -- is that of checking whether there are significant remanent defects in the model (remanent systematic errors). If statistical fluctuations are the only source of difference between $\underline{I}$ and $\underline{G}$, the mean value of $|\underline{I} - \underline{G}|^2$ will be close to the variance of $\underline{I}$, and the weighted sum,

$$\underline{S} = \sum_{\underline{hkl}} |\underline{I}(\underline{hkl}) - \underline{G}(\underline{hkl})|^2/\sigma^2(\underline{I}) \qquad (31)$$

should be close to the number of terms summed -- actually a little less, since refinement makes $\underline{G}$ as close as possible to the $\underline{I}$ actually observed, and not as close as possible to the true value. The expected value of $\underline{S}$, then, is not $\underline{n}$, the number of terms in the sum, but

$$\langle \underline{S} \rangle = \underline{n} - \underline{m} , \qquad (32)$$

where $\underline{m}$ is the number of parameters determined; for discussion and references see /10/. The standard deviation of the sum $\underline{S}$ is expected to be

$$\sigma_{\underline{S}} = [2(\underline{n} - \underline{m})]^{1/2} \qquad (33)$$

approximately /10/, so that if the actual value of $\underline{S}$ exceeds

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[7] For the necessary qualifications of this broad statement see Prince /9, Chapter 6/. Such parameters are 'best linear unbiased estimates'.

$$\langle S \rangle + k\sigma_S = n - m + k\sigma_S = n - m + k[2(n-m)]^{1/2} \quad (34)$$

(where k = 2 or 3, according to taste), one of two conclusions must follow: either (i) there are serious defects in the model (remanent systematic errors); or (ii) the statistical fluctuations have been seriously underestimated. The first interpretation has been generally accepted by crystallographers interested in the accuracy of the determination of lattice parameters /24,25,26,27,28,29/, and has in fact led them to improve certain corrections for systematic error /30,31,32/. Crystallographers interested in structural parameters have, on the whole, preferred the second interpretation, and have adjusted the weights by a factor designed to reduce S to an acceptable value. This practice may have arisen from a confusion between precision (statistical reproducibility) and accuracy (closeness to the true value), but it is more comfortable to postulate larger random errors than to attempt to improve the model or to reduce systematic errors. It would seem that adjusting the weights is legitimate only when there are objective reasons -- not mere discrepancy between observation and calculation -- for believing that the variances have been underestimated. For a discussion, see Prince /33/ and Rollett /34/.

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