Numerical Separation of Background in X-ray Diffraction Studies

S. A. Suevalov and I. G. Kaplan
Ural State University, Yekaterinburg, Russia
e-mail: sergey.suevalov@usu.ru
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Abstract—A method for the separation of X-ray background is suggested. The method is based on the integral digital filtration of experimental diffraction patterns and their “smoothening” and the subsequent combination of the smoothened and initial diffraction patterns. The combined diffraction pattern thus obtained is smoothened again, and the whole procedure is repeated anew. The criterion for concluding the iteration procedure (background criterion) is suggested to be the attainment of the situation where a certain percent of the points of the initial diffraction pattern would be below the background line determined. It is shown that, in some instances, the method yields more appropriate results than the traditional methods. The application of the new method is not limited by X-ray diffraction alone. © 2005 Pleiades Publishing, Inc.

INTRODUCTION

The importance of the background separation on X-ray diffraction patterns is tightly associated with the problems of determining phase composition and widespread use of the methods of determination of various crystal structures based on the graphical representation of experimental intensities of X-ray diffraction reflections. No matter how paradoxical this may seem, the problem of separation of background from X-ray diffraction reflections is complicated by the existence of many methods of its solution. It is well known that an experimental diffraction pattern consists of the informative part (signal) proper and the noninformative part (noise). Noise may be caused by various factors, e.g., inelastic scattering, electron noise, etc. The separation of the maxima of the coherent Bragg scattering from the remaining part of scattering may be considered, to a large extent, as the art of the experimenter. The traditional method of solution of the problem is the Fourier analysis of diffraction patterns. For example, the method of separation of X-ray background is implemented in the packages of programs of full-profile analysis GSAS [1] and FULLPROF [2]. The method of Fourier analysis seems to be quite reasonable and, because it is practically the only existing method for the solution of the problem, it has been never criticized. Moreover, Fourier analysis also allowed attainment of very good results in processing of other signals. One may make several remarks on Fourier analysis of X-ray diffraction patterns. The determination of the Fourier transform (Fourier integral) for a discrete limited set of points is insufficiently correct by definition. Once the Fourier transform is determined, we have to use a certain criterion in order to separate the low- and the high-frequency signals. The problem of the selection (formulation) of such a criterion should be considered as a certain type of art.

THEORY OF THE METHOD

Consider the iteration process of separation of X-ray background on the whole and use an arbitrary filter of the iteration type possessing the property

\[ I_{i}^{k+1} = P(I_{i}^{k}), \quad i \in [1, \ldots, N]. \]

Here, \( I_{i}^{k+1} \) is the current \((k+1)\)th iteration, \( I_{i}^{k} \) is the result attained at the previous iteration, \( N \) is the number of the experimental points, and \( P \) is a certain filter possessing certain properties which will be considered via the convenient and traditional representation in terms of the transformation into a Fourier integral. We believe that the conclusions made below do not depend on the type of the transformation used.

When expanding iterations into the Fourier integral, we may present the mechanism of the filter \( P \)'s action in the following way. There exists a certain limiting frequency \( \omega_{b}^{k} \) (subscript \( b \) indicates the background) at which the low-frequency component (background) \((l)\) is cut off from the high-frequency component (peaks) \((h)\) possessing the following properties (\( \omega_{b}^{k} = \text{const} \)):

\[ I_{(F)}^{k+1(h)} / I_{(F)}^{k+1(l)} = (1 - \alpha) (I_{(F)}^{k(h)} / I_{(F)}^{k(l)}), \]

where \( \alpha \) is an infinitesimal parameter of the filter action (\( 0 < \alpha < 1 \)). The lower index \((F)\) indicates that we deal with the Fourier transform of the X-ray diffraction pat-
tern. In other words, the fraction of the high-frequency component of the signal should decrease with each new iteration \( k \). If \( k \to \infty \), we have
\[
\lim_{k \to \infty} \left( \frac{k^{(h)}}{k^{(l)}} \right) = 0. \tag{3}
\]
In other words, the high-frequency component goes to zero. Determining the frequency \( \omega^k_b \) by some empirical method, and applying the inverse Fourier transformation of the low-frequency component at sufficiently large \( k \), we obtain a certain function which may be considered as the function of the X-ray background line. However, it is impossible to evaluate \textit{a priori} neither \( \omega^k_b \) nor \( k \) or \( \alpha \) for an arbitrary filter and an arbitrary diffraction pattern.

Now, we have to impose the following conditions
\[
\omega^{k+1}_b = (1 - \beta) \omega^k_b, \tag{4}
\]
where \( \beta \geq 0 \). The case \( \beta = 0 \) was considered above (see Eq. (3)), whereas at \( \beta > 0 \) and \( k \to \infty \), we have
\[
\lim_{k \to \infty} \omega^k_b = 0. \tag{5}
\]
In other words, the inverse Fourier transformation yields
\[
I_{b,i} = C, \quad i \in [1, \ldots, N]. \tag{6}
\]
This trivial background function is not very useful. Of course, we may also assume that
\[
\beta_i = f(I^k_i), \quad i \in [1, \ldots, N], \tag{7}
\]
i.e., to describe \( \beta_i \) as a certain function of the signal shape (e.g., by using a certain polynomial). In this case, at certain \( k \) values, \( \beta_i \) may be either greater or less than zero. At the appropriately selected function \( f \), it is possible to hope that \( \omega^k_b > 0 \) and
\[
\lim_{k \to \infty} \omega^k_b = \omega_b > 0. \tag{8}
\]
Now, the background function has a sufficiently appropriate form. The type of filtration used is not exceptional for processing diffraction patterns if criterion (2) is fulfilled. The mechanism of the filter action with respect to the parameters \( \alpha, \beta, k \), and \( f \) is empirical.

Thus, the problem of background separation is reduced to the selection of a simple and unique background criterion. This may be a certain conditional fraction of the points on a diffraction pattern with the intensities lower that the desirable background line. The evaluation of the fraction of the background points may be rather crude, because this does not affect the final result.

We suggest separating the X-ray diffraction background by the methods of digital integral filtration without opposing it to the method of the Fourier filtration. The method is rather simple and, in the majority of cases, efficient. Its algorithm is also rather simple: the initial diffraction pattern is smoothened by averaging the intensity of a certain central point with the intensities of some neighboring points. The next operation consists in the following: the intensities of the initial

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{diffraction_patterns.png}
\caption{Part of the diffraction patterns from magnetite FeFe_2O_4 and the separated background. The intensity of the highest peak is about 3500 pulses.}
\end{figure}