New method for calculation of radial distribution function

A V Bondarev, I L Bataronov, Yu V Barmin and O A Kordin
Voronezh State Technical University, 14 Moskovski prospect, Voronezh 394026, Russia
E-mail: bondarev@vmail.ru

Abstract. A method for calculation of radial distribution function for amorphous alloys was developed. It is based on smoothing of each peak of the experimental structure factor (interference function) with the sets of basic functions in the form of wave packages with the subsequent Fourier transformation of the smoothed structure factor. The developed method enables one to analyze the influence of the separate components of the form of structure factor on peculiarities of the radial distribution function.

1. Introduction
In structural investigations of amorphous materials the analysis of the radial distribution function (RDF) plays the significant role. So the increasing of accuracy and reliability of the experimental data is an important problem [1, 2].

The reduced distribution function \( G(r) \) can be obtained as the sine Fourier transformation of the experimental structure factor (SF) \( S(k) \) [3]:

\[
G(r) = \frac{2}{\pi} \int_0^{\infty} [S(k) - 1] \sin(kr) dk. \tag{1}
\]

However, the direct application of this formula is sensitive to such experimental errors in the SF as statistical dispersion, measurements within the finite interval etc. To reduce the effect evoked by these errors the following algorithm was proposed [4, 5]: at first the experimental \( S(k) - 1 \) function is smoothed by its expansion in a set of basic functions, and then the Fourier transformation of the smoothed solution is performed according to formula (1). The odd orthonormalized Hermite functions were chosen as the basic ones. In [6] the initial region of the SF (before the first peak) was smoothed by the Hermite functions, and the rest of the interval – by the Laguerre functions.

As it was shown in [6], using the regularizing procedures based on the SF smoothing essentially increases the accuracy of calculation of the \( G(r) \) function and provides stability to the random experimental errors.

At the same time, as it is seen from the experience of smoothing with basic functions in the whole interval of the experimental SF, for obtaining the statistically significant accuracy of the expansion, the great number of basic functions (up to 160) is required. It is connected with the fact that the basic functions being used in the expansion not adequately describe the SF. In order to overcome this drawback, in this paper we propose to use the basic functions in the form of wave packages that allow us directly during the expansion to construct the set of basic functions that are adequate to the shape of the SF.

2. Computational technique
Let \( S(k) - 1 \) is a function determined from X-ray diffraction data. The smoothing procedure is conducted by expansion of the SF by the formula [6]:

\[
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where $a_n$ are coefficients of expansion, $\varphi_n(k)$ is set of linearly independent basic functions, $\xi(k)$ is weight function allowing us to take into consideration the greater role of any components of the SF. In this paper we used the weight function $\xi(k)=1$.

Substitute the expression (2) to the formula (1) and introduce the symbol

$$\Phi_n(r) = \frac{2}{\pi} \int_0^\infty k \sin(kr) \varphi_n(k) \frac{dk}{\xi(k)} ,$$

then we obtain:

$$G(r) = \sum a_n \Phi_n(r) .$$

The idea of use of the wave packages consists in that each peak of the SF is expanded into independent set of wave packages localized within the given peak. At first the SF must be transformed so that it will be represented by the separate positive peaks divided by the zero values of the function. In this paper such a transformation is carried out by adding to the SF a smooth function that is the lower envelope of the SF:

$$S'(k) = S(k) - 1 + e^{-bk} \sum_{m=0}^N \frac{(bk)^m}{m!} .$$

Here the parameters $b$ and $N$ are chosen directly by the SF, i.e. so that the $S'(k)$ function will be nonnegative in the whole region of its representation and the minimums of the $S'(k)$ function will touch the abscissa axis.

In the interval $[k_{\text{min}}; k_{\text{max}}]$ the structure factor usually have up to four peaks. Each peak of the experimental SF is approximated with a separate family of wave packages localized within the corresponding peak:

$$\varphi_n(k) = M k^\beta e^{-\alpha k} \cos(nkr) , \quad n = 0, 1, \ldots$$

where $M = \left( \frac{e\alpha}{\beta} \right)^{\beta}$ is normalizing factor; $\alpha$, $\beta$ and $R$ are parameters specifying the given family. The number of families is chosen during the smoothing by the number of peaks of the SF.

For determination of the parameters $\alpha$ and $\beta$ let us express them from other parameters $p$ and $q$:

$$p = \frac{\beta}{\alpha} , \quad q = \frac{\sqrt{\beta+1}}{\alpha} ,$$

which have the simple geometric sense: the value of $p$ determines position of the corresponding maximum of the $S'(k)$ function, and $2q$ determines its half-width. From (7) we can express $\alpha$ and $\beta$:

$$\alpha = \frac{p + \sqrt{p^2 + 4q^2}}{2q^2} , \quad \beta = \alpha p .$$

The parameter $R$ determines the number of half-waves in a package. Let us choose it so that the integer number of half-waves ($n$) will lie within the localization limits of the wave package:

$$R = \frac{\pi}{2p} \left( 1 + \frac{2p}{3q} \right) .$$

Fourier transformation of the functions (6) according to (3) has the form:
\[
\Phi_n(r) = \frac{M}{2} \frac{\Gamma(\beta + 2)}{\alpha^{\beta+2}} \left( \frac{\sin((\beta + 2)\omega_\pm)}{\rho_{\beta+2}^{\pm}} - \frac{\sin((\beta + 2)\omega_\pm)}{\rho_{\beta+2}^{\mp}} \right),
\]

(10)

where \( \rho_\pm = \left( 1 + \left( \frac{nR \pm r}{\alpha} \right)^2 \right)^{1/2} \), \( \omega_\pm = \arctan \frac{nR \pm r}{\alpha} \).

As an example, in figure 1 we show \( \phi_n(k) \) and \( \Phi_n(r) \) functions with \( p=2; q=0.5; n=0, 1, 2 \). As it is seen from figure 1, the \( \Phi_n(r) \) functions are also wave packages, but do not form a family of functions localized within one interval. From the formula (10) and figure 1 it follows that the \( p, q \) and \( R \) parameters also characterize the properties of the \( \Phi_n(r) \) function. Namely, the value of \( nR \) is a coordinate of the center of the \( \Phi_n(r) \) package, the value of \( 1/(2q) \) is a half-width and \( 2\pi/p \) is a half-period of oscillations. This correspondence allows us to perform the direct physical analysis of influence of separate components of the SF shape on the peculiarities of the RDF which characterizes the short-range atomic ordering in the amorphous systems.

**Figure 1.** Examples of wave packages (a) and their Fourier transformations (b).
Solid lines \(- n=0, \) dotted lines \(- n=1, \) dashed lines \(- n=3. \)

The final form of the RDF is obtained from (4) taking into account the transformation of (5):

\[
G(r) = \sum a_n \Phi_n(r) - \frac{2}{\pi} \sum_{m=0}^\infty \frac{\Gamma(m+2)\sin((m+2)\arctan \frac{r}{b})}{m!\left(b^2 + r^2\right)^{m+1/2}}.
\]

(11)

Then the coefficients \( a_n \) are found by the standard method [6]
3. Results and discussion

The proposed method of RDF calculation was approved on amorphous alloys of rhenium with transition metals of the sixth period (terbium, tantalum and hafnium) [7]. In figure 2 we present the SF of the Re$_{82}$Tb$_{18}$ amorphous alloy obtained from the X-ray diffraction experiment and the SF smoothed by the wave packages. The RDF calculated from it is shown in figure 3.

The advantage of the new method of the RDF calculation consists in that approximation of each SF peak with a separate set of wave packages allows us to control the smoothing process. In particular, the survey of intensity of scattered X-ray emission is often conducted in the “registration by points” regime with constant exposition time, and, therefore, the absolute value of random error on the experimental $S(k)$ curve increases with increasing $k$. Consequently, if a peak is situated at greater values of $k$, then the lesser number of basic functions is required for its approximation. In the adduced calculations for the Re$_{82}$Tb$_{18}$ alloy the SF was separated into four regions, which correspond to four peaks of the function. For approximation of the first peak on the SF we used 14 functions, for the second – 9, for the third – 5, and for the fourth – 3 functions. If we use the same set of basic functions on the whole interval of variation of $k$, then it will lead to that the smoothed SF will represent the random oscillations that have no physical sense and introduce the significant error into the resulting RDF.

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