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Some Peculiarities of RTOF Method and Calibration of Diffractometer "Mini-Sfinks".

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ABSTRACT

The problems of correct definition of interplanar distances are discussed for RTOF diffractometer. The influence of absorption effect and phase error are described.

1. Introduction

Recently investigations of strain distribution using neutron diffraction attract much attention. Indeed, the neutron diffraction is a powerful tool for this purpose [1]. Such advantages of RTOF method [2] as a high level of utilization of neutron flux and observing a number of diffraction lines in one measurement make this method attractive for such investigation [1].

Some peculiarities of RTOF method introduce difficulties in correct extraction of interplanar distances from diffraction pattern. Main problem is so-cold "phase error" (the phase difference between modulation function and "Pick-up" signal) that can distort line shape and position [3]. Up to now all realizations of RTOF method work with phase unstability, because all of them use Pic-up signal obtained by analog method [4].

The phase problem is especially important for diffractometers with high (10^{-3}-10^{-4}) resolution.

The correct solution of this problem must include deconvolution of measured line shape to resolution function and scattering function of the sample. The resolution function can be obtained from measurement of sample with known scattering function (standard sample). This calibration measurement not so important for RTOF diffractometer with unstable phase because resolution function depends on phase error [3].

The other possibility to solve phase problem is introducing in fitting procedure varied parameters that describe influence of phase error. One possibility was described by Kudryashev et al. [3] for antisimmetric resolution function when asymmetry of line shape is proportional to phase error.

At present paper we shell show that using ordinary "looks like Gaussian" resolution function allows as to take in account the shift of line positions from phase error and other effects. It will be shown that with using some assumptions the problem can be practically reduced to the calibration of two diffractometer constants. This calibration must be done for every measurement when information about absolute values of line positions is extracted.

Keywords: RTOF, phase error, sample absorption

2. Structural studies. Results and problems.

"Mini- Sfinks" diffractometer has been used for investigation of structure of numerous compounds. There are a lot of results that are in agreement with results obtained by other techniques.

In case of $Zr_{0.98}Hf_{0.02}O_2$ (standard certificated sample) the excellent agreement of structure parameters obtained with tabulated data confirms the high resolution of diffractometer [5].

In case of hexaborides of rare-earth elements the mean-square displacements of atoms extracted by help of Rietveld method were confirmed by model calculations [6].

To get these results we used to allow variation of time shift (Delay) parameter, instead of traditional treatment of Delay as a diffractometer constant. The differences between initial and fitted values of Delay were higher than possible error of electronic equipment (0.1 μs) (See Table. 1).

Table. Fitted value of Delay for different experiment.
Rietveld analysis program "Mria" [7].

Compound	Fitted value of Delay channels, Δ·?μs
[illegible]	[illegible]
[illegible]	[illegible]
[illegible]	[illegible]
[illegible]	[illegible]
[illegible]	[illegible]
[illegible]	[illegible]
[illegible]	[illegible]
$Tm(DCOO)_3$	1000.11

In addition, in case of hexaborides (samples with high absorption) we had to vary Lsin(Θ) parameter using fixed lattice constant obtained from X-ray single crystal measurement.

For structural investigation these deviations of diffractometer constants from initial values are not so important if the correlations between these constants and physical meaningful parameters are negligible.

For correct extraction of line positions we have to use the correct values of diffractometer constants and understand why the differences appear.

3. Lsin(Θ) and absorption

Lattice constant of lanthanum hexaboride calculated using the positions of separate diffraction lines shows unphysical dependence on neutron wavelength λ.

This dependence can be described by help of absorption correction. The main idea of this correction is following (Fig. 1): absorption attenuation is different for different λ, so every point (x,y) of sample cross-section produces intensity on the detector with weight:

$$P(x,y) = \exp\left[-\mu(\lambda)\cdot\left(a(x,y)+b(x,y)\right)\right], \qquad (1)$$

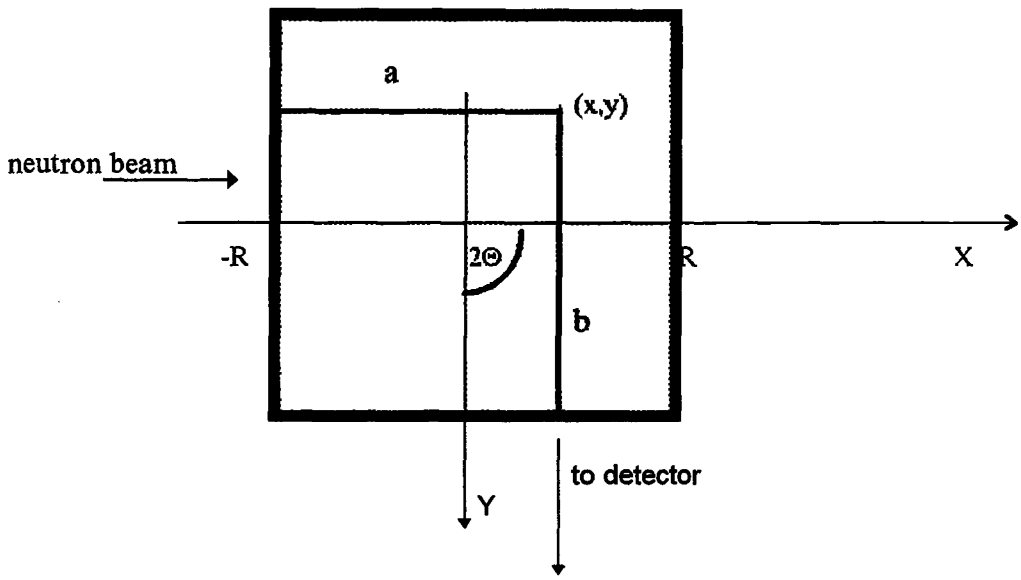

Fig. 1 Sample cross-section for 2Θ=90⁰ geometry.

The coordinates of center of gravity of this weight distribution are coordinates of effective scattering center (x_c,y_c). The position of scattering center depends on linear absorption coefficient μ and so on λ. The actual L and Θ can be calculated using x_c and y_c.

$$\left.\begin{array}{l} x_c = \frac{1}{M}\cdot \iint\limits_G \exp(-\mu(a(x,y)+b(x,y)))\,x\,dx\,dy \\ y_c = \frac{1}{M}\cdot \iint\limits_G \exp(-\mu(a(x,y)+b(x,y)))\,y\,dx\,dy \end{array}\right\} \qquad (2)$$

For cylindrical sample and backscattering geometry these integrals can be taken numerically.

Main assumptions we made are following:

- cinematic limit of diffraction,
- no divergence of primary beam,
- point detector assumption,
- uniform distribution of primary beam on width and height of sample.

Now corrected line positions can be calculated using formulas:

$$d_{hkl} = \frac{h\cdot\tau_{hkl}}{2m\cdot p_0\left(1-\frac{\delta p}{p_0}\right)}, \qquad (3)$$

$$\frac{\delta p}{p_0} = \frac{\delta L}{L_0} + \cot\Theta_0\cdot\delta\Theta, \qquad (4)$$

$$d_{hkl} = d^0_{hkl}\left(1+\frac{\delta p}{p_0}+\left(\frac{\delta p}{p_0}\right)^2+\cdots\right) \quad \text{where} \quad d^0_{hkl} = \frac{h\cdot\tau_{hkl}}{2m\cdot p_0} \quad \text{and } \delta p=\delta p(\lambda).$$

For LaB_6 data we can simplify this treatment using backscattering condition:

$\left(\cot \Theta \cdot \delta\Theta << \frac{\delta L}{L_0} \right)$ and the relation of lattice constant and hkl for cubic lattice:

$A = \sqrt{h^2 + k^2 + l^2} \cdot d_{hkl}$.

The result of calculation of this effect can be seen on Fig.2, where the experimental points and the results of modeling are in a good agreement.

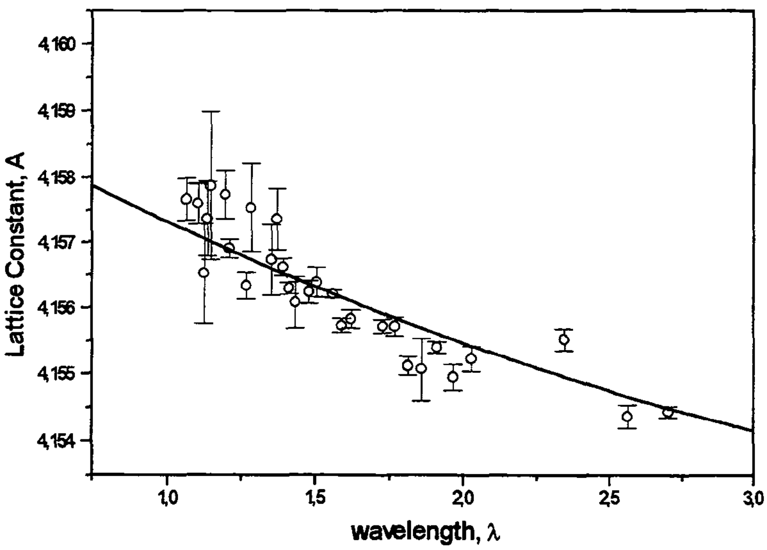

Fig. 2 Calculated from fitted line positions lattice constant of LaB_6.
Solid line is result of theoretical calculation.

It should be stressed that this effect can be visible for samples with high absorption in measurements made by using high resolution (10^{-3}-10^{-4}) machine.

4. Delay and phase error

The description of phase error influence on line positions is done by Kudryashev as well as the general method of fitting line positions when phase error is present [3]. For "Mini-Sfinks" fitting procedure this possibility in not realized, so phase error should influence on other parameters.

To describe the deviation of Delay from initial value we propose that the phase error can influence on line positions in the process of current fit.

We calculate the series of line shapes for different phase errors using as basic Gaussian line shape and method described in [3].

The results of fits of generated line shapes by Gaussian are shown at the Fig. 3.

It is clearly seen that for the small phase error there are not noticeable changes at line width and integral line intensity, but the line position shows strong linear dependence on phase error.

It means that all lines shift from these real positions with the same shift which depends on phase error. In process of fit we can see it in the deviation of fitted Delay value from initial Delay (see Table 1).

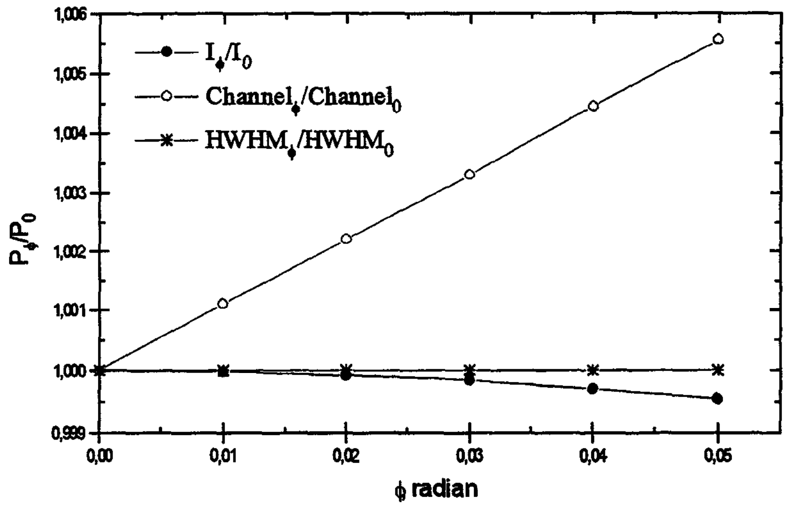

Fig. 3 Influence of phase error on integral intensity, HWHM and line position.

In case of measurement of investigated sample together with standard one the simple method should be proposed for calibration of Lsin(Θ) and Delay parameters. Using notation $\tau_{hkl} = \Delta \cdot \left(C_{hkl} + Delay\right)$ where Δ is channel width in μs, C_{hkl} is line position in channels, the formula (3) can be rewritten for cubic lattice in the following form:

$$C_{hkl} = \frac{h\Delta}{2m} \cdot A \cdot L \cdot \sin(\Theta) \cdot \frac{1}{\sqrt{h^2 + k^2 + l^2}} - Delay$$

Let us introduce following notations:

$$\alpha = \frac{h\Delta}{2m} \cdot A \cdot L \cdot \sin(\Theta)$$

$$P_{hkl} = \frac{1}{\sqrt{h^2 + k^2 + l^2}}$$

As one can see line positions C_{hkl} linearly depend on P_{hkl}. The slope α of straight line is proportional to the lattice constant and Lsin(Θ) parameter, the intercept is just (-Delay) value.

The results of such calibration measurements are shown at the Fig. 4 for simultaneous measurement of diamond powder and Mo-Cr alloy placed in one container. The fitted values of Delay are in a good agreement. The difference of slopes depends on the difference of lattice constants that allows to calculate the correct value of investigated alloy lattice constant instead of phase error.

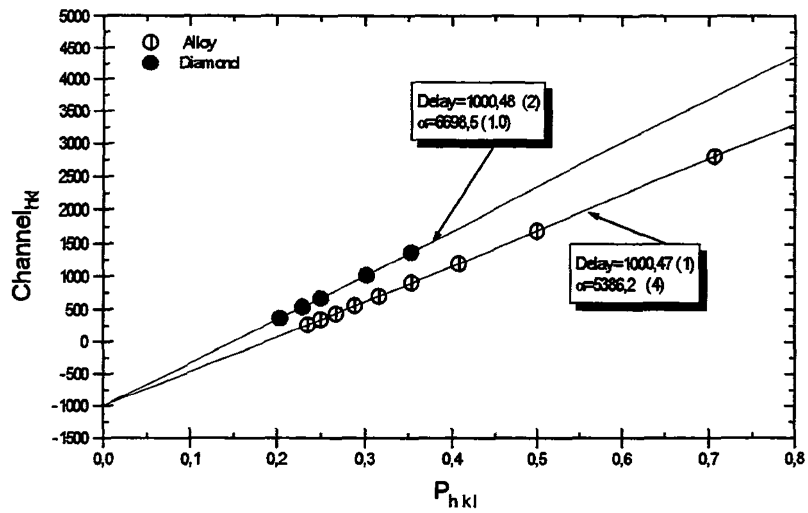

Fig.4 C_{hkl}-P_{hkl} plot for (diamond+Alloy) sample

5. Conclusion

For correct extraction of information from RTOF diffraction spectra it is needed to take in account the shifts of lines arising from Lsin(Θ)=f(λ) dependence and phase error.

There are few effects that could introduce the dependence of effective Lsin(Θ) parameter on neutron wavelength. Effective value of Lsin(Θ) can be calculated using coordinates of the center of weight distribution $P(x,y,z,\lambda)=\prod_i p_i(x,y,z,\lambda)$ where p_i describes influence of absorption, primary beam distribution on the exit of neutronguide, divergence effect et al.

Small phase errors mainly shift the line positions.

One can see these effects using "C_{hkl}-P_{hkl}" plots. The deviation of intercept from initial value of Delay must be proportional to phase error. Systematic deviations of experimental points from strait line will show on Lsin(Θ)=f(λ)dependence.

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