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Secondary Reflexions of Neutrons Diffracted by a Single-Crystal Bar Vibrating at High Frequency

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Diffraction of neutrons by a longitudinally vibrating quartz single-crystal bar was investigated for thicknesses of 3 and 13 mm. The 1st, 3rd and 5th harmonic frequencies were excited in the bar. The observed increase of the integrated intensity of diffracted neutrons as a function of the vibration amplitude of longitudinal vibrations of the quartz bar is compared with that calculated from an approximate theory considering the possibility of secondary reflexions of neutrons during their flight across the sample.

1. Introduction

The investigations of neutron diffraction by vibrating single crystals presented by Moyer & Parkinson (1967), Klein, Prager, Wagenfeld, Ellis & Sabine (1967), Chalupa, Michalec, Petržílka, Tichý & Zelenka (1968) and Michalec, Chalupa, Petržílka, Galociová, Zelenka & Tichý (1969) have shown a significant increase in the integrated intensity of neutrons diffracted as a function of the vibration amplitude of the single crystal.

The theoretical explanation of some observed phenomena is given by Michalec, Sedláková, Čech & Petržílka (1971), Buras, Giebultowicz. Minor & Rajca (1972), Buras & Giebultowicz (1972), Mikula, Michalec, Sedláková, Čech, Chalupa & Petržílka (1973) and Michalec, Chalupa, Petržílka, Sedláková, Čech & Mikula (1974).

In diffraction experiments, neutrons with a wavelength of $\lambda=1$ to 2 Å are conventionally used. These neutrons with velocities of 4×10^5 to 2×10^5 cm s^{-1} are also suitable for the investigation of dynamical effects associated with the displacement of crystallographic planes and its influence upon the process of neutron diffraction.

As the frequency of longitudinal vibrations of the piezoelectrically excited quartz single-crystal bar of length 70 mm is $f\simeq40\times10^3$ Hz, it is convenient to investigate the integrated intensity of neutrons diffracted for the vibration period τ either much higher or comparable with the time Δt which the neutrons spend in a vibrating single crystal. This condition can be fulfilled by exciting the higher orders of the fundamental frequency and by using different single-crystal bar thicknesses.

2. Theoretical considerations

Let us suppose a bar-shaped perfect single crystal, vibrating longitudinally in the Y-axis direction. If we consider longitudinal vibrations only, then the oscillation direction and propagation direction of vibrations both coincide with the direction of the Y axis. In such a case the displacement u_{yK} of the plane (hkl) for the Kth harmonic frequency can be described by a function in space and time

$$u_{yK}=u_{0K}\sin\frac{K\pi}{L}y\sin K\omega t \tag{1}$$

where u_{0K} is the maximum amplitude for the Kth harmonic frequency, L is the length of the bar, y is the coordinate, $f=\omega/2\pi$ is the fundamental resonance frequency and K is the mode order (see Fig. 1).

The deformation and the movement of the lattice plane in the direction of the Y axis with a velocity $V_p(t)$, bring about the change $\varphi=\theta-\theta_B$ of the Bragg angle θ_B in the case of a symmetric transmission according to the equation

$$\varphi(t)=-\frac{u_{0K}K\pi}{L}\left[\cos\frac{K\pi}{L}y\sin K\omega t + \frac{C_y}{|V_{ny}|}\sin\frac{K\pi}{L}y\cos K\omega t\right]\operatorname{tg}\theta_B \tag{2}$$

which conforms with equation (2) of the paper of Michalec *et al.* (1971). $|V_{ny}|=V_n\sin\theta_B$, C_y is the velocity of ultrasonic waves in the direction of the Y axis, and V_n is the neutrons velocity.

Let us suppose that a polychromatic neutron beam (see § 3) impinges on the lattice planes at the Bragg angle θ_B. During the time of flight $\Delta t=T\operatorname{tg}\theta_B/|V_{ny}|$ of neutrons across the sample with the thickness T, the deformation gradient and the acceleration of the moving planes bring about the change $\delta\varphi(t)$ (Michalec *et al.* 1974).

$$\delta\varphi(t)=\varphi_1(t)-\varphi_2(t)$$

$$\varphi_1(t)=-\frac{u_{0K}K\pi}{L}\left[\cos\frac{K\pi}{L}y_1\sin K\omega t\right.$$

$$+\frac{C_y}{|V_{ny}|}\sin\frac{K\pi}{L}y_1\cos K\omega t\Big]\,\mathrm{tg}\,\theta_B \qquad (3)$$

$$\varphi_2(t)=-\frac{u_{0K}K\pi}{L}\Big[\cos\frac{K\pi}{L}y_2\sin K\omega(t+\Delta t)$$

$$+\frac{C_y}{|V_{ny}|}\sin\frac{K\pi}{L}y_2\cos K\omega(t+\Delta t)\Big]\,\mathrm{tg}\,\theta_B$$

where y_1, t ($y_2=y_1-T\,\mathrm{tg}\,\theta_B, t+\Delta t$) are the coordinate and the time at which the neutrons enter the bar (leave the bar when the diffraction does not occur).

After a simple calculation and omission of the subscripts, we find with a good approximation for $y\simeq L/2K$, $C_y/2L=f$,

$$-\delta\varphi(t)=\frac{2u_{0K}K\omega\,\mathrm{tg}\,\theta_B}{|V_{ny}|}\sin\frac{K\omega T\,\mathrm{tg}\,\theta_B}{2|V_{ny}|}$$

$$\times\sin\frac{K\pi}{L}y\sin K\omega(t+\Delta t/2)\,. \qquad (4)$$

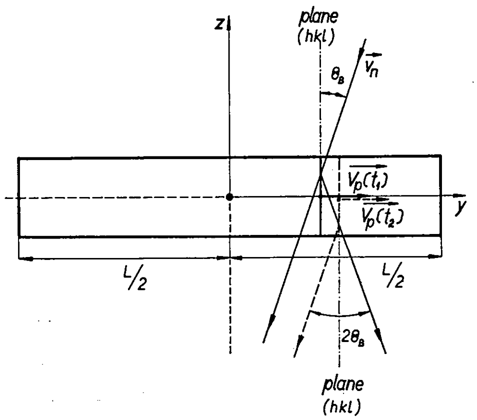

Fig. 1. Schematic arrangement for neutron diffraction by a vibrating single-crystal bar with respect to the crystallographic system of coordinates.

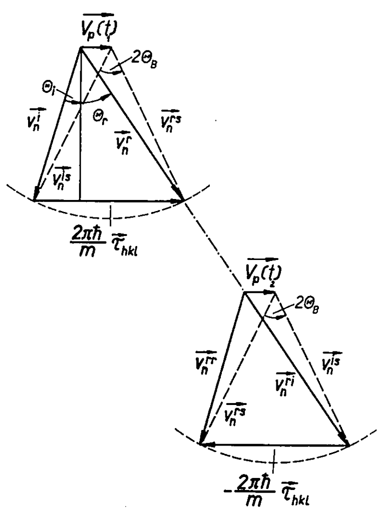

Fig. 2. Reciprocal-lattice construction of regular and secondary diffraction on two parallel planes of the same type, moving with the velocities $\mathbf{V}_p(t_1)=\mathbf{V}_p(t_2)$.

This relation holds if $T\,\mathrm{tg}\,\theta_B \ll y$.

Now we can write that the integrated intensity of a diffracted beam P, as in the paper of Antonini, Corchia, Nicotera & Rustichelli (1972) is proportional to $\overline{n(t)}$ of perfect 'crystalline layers' normal to the Y axis and given in our case of a vibrating single crystal bar by the expression

$$\overline{n(t)}=\frac{\overline{|\delta\varphi(t)|}}{2\cdot 66s}=\frac{4u_{0K}K\omega\,\mathrm{tg}\,\theta_B}{\pi|V_{ny}|2\cdot 66s}$$

$$\times\sin\frac{K\omega T\,\mathrm{tg}\,\theta_B}{2|V_{ny}|}\sin\frac{K\pi}{L}y \qquad (5)$$

where

$$2s=2N_c\lambda^2F/\pi\sin 2\theta_B \qquad (6)$$

is the angle interval of a total reflexion in a perfect non-vibrating single crystal. N_c is the number of elementary unit cells per unit volume. Other symbols of equation (6) have their usual meanings.

Since $\delta\varphi(t)$ is a sinusoidal function of t, it is evident that throughout the time interval when $n(t)\gtrsim 1$ the consideration mentioned above is not valid. It is necessary to make this time interval much shorter than the vibration period τ. This condition may be fulfilled by increasing the amplitude u_{0K}.

Hence, according to the equations (4), (5) and (6) the formula for the integrated intensity P^v of a beam diffracted by a vibrating single-crystal bar becomes

$$P^v=P_1\frac{\overline{n(t)}}{T}v \qquad (7)$$

where $v=S_0T$ is the irradiated crystal volume, S_0 the area of the face of the irradiated volume element and P_1 the integrated intensity of a beam diffracted by one 'crystalline layer' with a unit area of the face. As can be seen from the equations (4) and (5), increasing the thickness T for a constant amplitude u_{0K}, we can find $P^v_{\max}$ when

$$\sin\frac{K\omega T_{\rm eff}}{2V_n\cos\theta_B}=1\,. \qquad (8)$$

This condition determines the effective thickness $T_{\rm eff}$ for a given frequency.

The thickness saturation phenomenon may be explained by the presence of secondary reflexions, which mainly occur if

$$\frac{\pi}{2}<\frac{K\omega T}{2V_n\cos\theta_B}<\pi\,. \qquad (9)$$

The neutrons totally diffracted by the planes moving with a velocity $\mathbf{V}_p(t_1)$ at the instant t_1 can be again totally diffracted by the planes of the same type into the primary beam in the instant t_2 when

$$\mathbf{V}_p(t_2)=\mathbf{V}_p(t_1)\,. \qquad (10)$$

It is evident that throughout the time interval $t_2-t_1<\Delta t\simeq\tau$ the neutrons are passing through the bar.

The plane velocity of a longitudinally vibrating single crystal bar is given by the expression

$$V_p(t) = \frac{\partial u_{yK}}{\partial t} = u_{0K} K\omega \sin \frac{K\pi}{L} y \cos K\omega t \,. \quad (11)$$

The condition (10) for the presence of secondary reflexions can be expressed ($y \simeq L/2K$) in the form

$$\sin \frac{K\pi}{L} y_1 \cos K\omega t_1 = \sin \frac{K\pi}{L} \times [y_1 + V_n \sin \theta_B (t_2 - t_1)] \cos K\omega t_2 \quad (12)$$

for $t_2 - t_1 < \tau$.

Fig. 2 is a schematic diagram of a regular and of a secondary diffraction of neutrons by means of a reciprocal-lattice construction at the sample point $y \simeq L/2K$ without considering the lattice deformation; $\mathbf{V}_n^i$, $\theta_i(\mathbf{V}_n^r, \theta_r)$ are the velocity and the angle of the incident (reflected) neutron in a regular diffraction; $\mathbf{V}_n^{ri}(\mathbf{V}_n^{rr})$ is the velocity of the incident (reflected) neutron in the course of the secondary diffraction; $\mathbf{V}_p(t_1)$ $[\mathbf{V}_p(t_2)]$ the plane velocity at the instant $t_1(t_2)$ in the sample coordinate $y_1[y_1 + V_n \sin \theta_B (t_2 - t_1)]$ and τ_{hkl} the reciprocal-lattice vector. Other symbols marked by the subscript s correspond to the auxiliary reciprocal-lattice construction for a non-vibrating crystal. The values of the angles θ_i, θ_r and the velocities $\mathbf{V}_n^i, \mathbf{V}_n^r$ are calculated and confirmed by experimental results presented in paper of Mikula *et al.* (1973).

3. Experimental results

The measurements were carried out by means of the double-axis spectrometer (Michalec, Vavřín, Chalupa & Vávra, 1967). A beam of nearly monoenergetic neutrons with wavelength $\lambda = 1{\cdot}05$ Å impinging on the investigated piezoelectrically vibrating quartz bar was diffracted by the plane (01.0) in the position of symmetric Laue transmission and detected by a $^{10}BF_3$ detector.

The half width of the rocking curve was the same for both the vibrating and non-vibrating crystal, namely 12′. The high value of the mosaic spread of the monochromator relative to the perfection of the quartz single crystal enabled us to consider the neutron beam incident on the sample as a polychromatic one in the angle interval $\theta = \theta_B \pm \Delta\theta/2$ ($\Delta\theta \simeq 1'$). The width of the incident beam was 5 mm. Moving the specimen in the Y direction by means of a cross table, the single-crystal bar was scanned when not vibrating as well as when vibrating at resonance frequencies $f = 38{\cdot}948$, $115{\cdot}760$ and $198{\cdot}300$ kHz. Thus nodal lines and antinodes were determined. The dimensions of the bar-shaped quartz single crystal were: 3 mm in the X direction, 70 mm in the Y direction and 13 mm in the Z direction (Fig. 1).

Figs. 3 to 5 illustrate the dependence of the integrated intensity of neutrons diffracted by a vibrating single-crystal bar on the resonator current i, exciting the sample at the frequencies $f = 38{\cdot}948$ kHz (Fig. 3), $f = 115{\cdot}760$ kHz (Fig. 4) and $f = 198{\cdot}300$ kHz (Fig. 5) for two thicknesses $T_x = 3$ mm [curve (*a*)] and $T_z = 13$ mm [curve (*b*)]. The change of the thickness was made using simple rotation of the bar round the Y axis. All the effects were investigated in the antinodes ($y \simeq L/2K$).

The vibration amplitude u_{0K} was measured with a microscope for the fundamental frequency. A linear

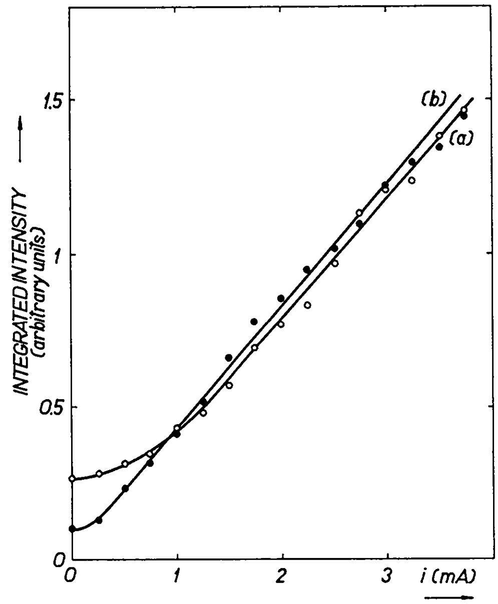

Fig. 3. The integrated intensity P^v as a function of high-frequency current i for the sample vibration at the fundamental frequency for two thicknesses $T_x = 3$ mm [curve (*a*)] and $T_z = 13$ mm [curve (*b*)].

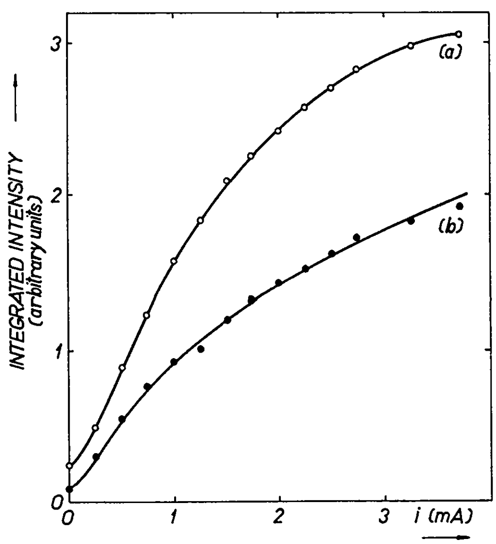

Fig. 4. The integrated intensity P^v as a function of high-frequency current i for the sample vibration at the third harmonic frequency for two thicknesses $T_x = 3$ mm [curve (*a*)] and $T_z = 13$ mm [curve (*b*)].

dependence of u_{01} on high-frequency current i flowing through the bar was observed. A vibration amplitude $u_{01}=4$ μm corresponds to the current $i=5$ mA. The dependence u_{0K} on i for $K=3$ and $K=5$ was not verified because the amplitudes u_{0K} were not measurable by the optical device used.

4. Discussion

In the case of $K=1$ equation (5) can be simplified for both thicknesses $T=3$ and 13 mm by means of the relation

$$2\sin\frac{\omega T\,\mathrm{tg}\,\theta_B}{2|V_{ny}|}\simeq\frac{\omega T\,\mathrm{tg}\,\theta_B}{|V_{ny}|} \tag{13}$$

and can be written in the form

$$\overline{n(t)}=\frac{2u_{01}\omega^2T\,\mathrm{tg}^2\,\theta_B}{\pi V_{ny}^2 2{\cdot}66s}\sin\frac{\pi}{L}y \tag{14}$$

which corresponds to the equation (5) of the paper of Michalec *et al.* (1974) where the calculated theoretical values of $(P^v/P_1)_c$ are compared to experimental results of $(P^v/P_1)_{\mathrm{exp}}$. Since $\overline{n(t)}/T$ is not a function of the bar dimensions, it follows from equation (7) that the integrated intensity P^v is roughly the same for both thicknesses.

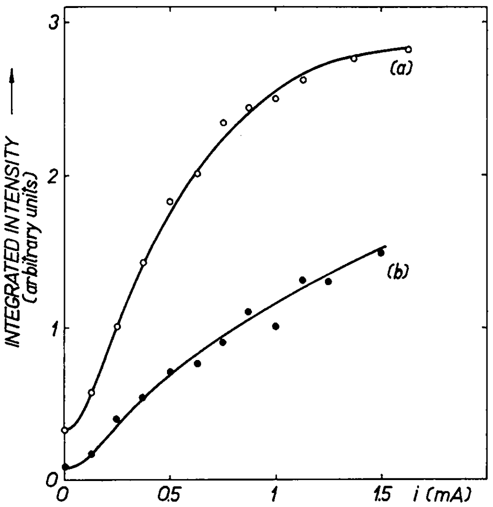

Fig. 5. The integrated intensity P^v as a function of high-frequency current i for the sample vibration at the fifth harmonic frequency for two thicknesses $T_x=3$ mm [curve (*a*)] and $T_z=13$ mm [curve (*b*)].

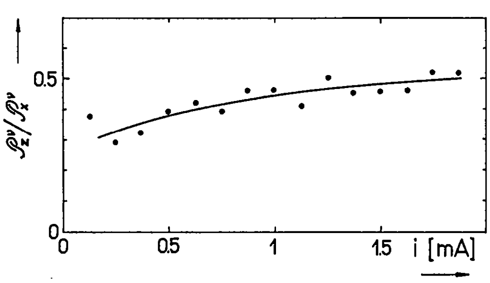

Fig. 6. The ratio $(P_z^v/P_x^v)_{\mathrm{exp}}$ as a function of high-frequency current i for the sample vibration at the fifth harmonic frequency.

Fig. 3 shows a very good agreement of the experimental results with the theoretical considerations mentioned above for $K=1$. The linear dependence P^v on the excitation current i proves the relations (5) and (7) in the case of $T=3$ (13) mm for $i>1$ (0·3) mA, considering u_{01} as a linear function of i.

As u_{0K} becomes still larger, P^v in equation (7) increases indefinitely. From the physical point of view, this is impossible, since there is an upper limit to the integrated intensity, which corresponds to the case of an 'ideally imperfect' crystal. For large u_{0K} the curve is expected to level off.

Another factor that should be considered is that for larger amplitudes u_{0K} is no longer a linear function of the crystal current i. Therefore the deviation from linearity in the intensity dependence for $K=3$ and 5 may be due to either one or both of the two reasons mentioned above.

The linear dependence P^v of the vibrating single-crystal bar on the irradiated volume v is analogous to the results of kinematical theory of diffraction on a small perfect single crystal. Similarly as in Zachariasen (1967), the kinematical relation (7) for integrated intensity P^v can be written in the form

$$P_{\mathrm{kin}}^v=P_0vA(\mu)Q'(u_{0K},\omega) \tag{15}$$

where P_0 is the incident intensity, $A(\mu)$ the transmission factor with linear absorption coefficient μ and $Q'(u_{0K},\omega)$ the integrated reflectivity of the crystal unit volume.

In the case of fundamental frequency ($K=1$)

$$Q'(u_{01},\omega)=\frac{P_1\overline{n(t)}}{P_0T}=\frac{P_1 2u_{01}\omega^2\,\mathrm{tg}^2\,\theta_B}{P_0\pi V_{ny}^2 2{\cdot}66\,s}\sin\frac{\pi}{L}y\,. \tag{16}$$

The transmission factor $A(\mu)$ can be considered to be unity for the quartz bar.

The analysis of the great intensity differences illustrated in Fig. 4 and 5 in cases of $K=3$ and 5 requires the following relations

$$P_x^v=P_1v\,\frac{2u_{0K}K^2\omega^2\,\mathrm{tg}^2\,\theta_B}{\pi V_{ny}^2 2{\cdot}66s}\sin\frac{K\pi}{L}y \tag{17}$$

for $T_x=3$ mm,

$$P_z^v=P_1v\,\frac{4u_{0K}K\omega\,\mathrm{tg}\,\theta_B}{\pi|V_{ny}|\,2{\cdot}66sT_z}\sin\frac{K\omega T_z\,\mathrm{tg}\,\theta_B}{2|V_{ny}|}\sin\frac{K\pi}{L} \tag{18}$$

for $T_z=13$ mm.

The presence of the term

$$\sin\frac{K\omega T_z\,\mathrm{tg}\,\theta_B}{2|V_{ny}|}$$

in equation (18) enables us to estimate the magnitude of the secondary reflexions presuming that each of the $\overline{n(t)}$ 'crystalline layers' diffracts totally as in the Bragg case.

Theoretical values of $(P_z^v/P_x^v)_c$ for $K=1$, 3 and 5 are 1·00, 0·76 and 0·41. The average experimental values of $(P_z^v/P_x^v)_{\rm exp}$ for $K=1$ and 3 are 1·03 (for $i\ge 0{\cdot}75$ mA) and 0·61 (for $1\ge 0{\cdot}25$ mA).

For $K=5$ the comparison of the calculated value with the average experimental one is not possible, because the experimental value of $(P_z^v/P_x^v)_{\rm exp}$ depends on the high-frequency exciting current i, which dependence was not observed for $K=1$ and 3. Thus in the case of $K=5$ it is only possible to compare individual experimental quantities at low values of i. The increase of the ratio $(P_z^v/P_x^v)_{\rm exp}$ (for $K=5$) *versus* i is shown in Fig. 6. The increase is brought about by relatively high acceleration of the moving planes, in which case the assumption that each of $\overline{n(t)}$ 'crystalline layers' diffracts totally is no longer justified. It is seen from Fig. 6 that for i ranging from 0 to 2 mA the change of $(P_z^v/P_x^v)_{\rm exp}$ is $\simeq 20\%$. The authors realize that the theory presented in this paper is applicable only to qualitative estimation of neutron diffraction by vibrating single crystals and to explanation of some phenomena observed.

Similarly, in case of $K=3$ and $K=5$ it is possible to introduce a factor y_k' analogous to the extinction factor $y_{\rm ext}$ (Zachariasen, 1967) and to express the integrated intensity P_k' in the form

$$P_k'=P_{\rm kin}^v y_k' \qquad (19)$$

where

$$y_k'\simeq \frac{\sin\dfrac{K\omega T_z\,{\rm tg}\,\theta_B}{2|V_{ny}|}}{\dfrac{K\omega T_z\,{\rm tg}\,\theta_B}{2|V_{ny}|}}\,. \qquad (20)$$

On the basis of our experimental data and from our approximate theory we can make the following statement: For $A(\mu)=1$, the integrated intensity difference between Bragg and Laue diffraction disappears when the displacement of the diffracting planes is accelerated in the direction of the reciprocal-lattice vector.

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Crystal Structure Determination by Simultaneous use of Cosine Invariant Computation and the Multisolution Method

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An algorithm is given for the rapid computation of the cosine invariants, $\cos(\varphi_{-H1}+\varphi_{H2}+\varphi_{H1-H2})$, and the results are compared with the actual values for three structures. A weighting scheme is derived which enables this information to be incorporated directly into the multisolution tangent method of phase determination. Details are given of the determination of four unknown structures by this method.

Introduction

Three different direct methods have been proposed for solving crystal structures.

(1) Methods based on the zero value of the mean sine invariant are all derived from use of the $\sum_2$ formula (Karle & Hauptman, 1953) and the tangent formula (Karle & Hauptman, 1958):

$${\rm tg}\,\varphi_{H1}=\frac{\sum\limits_{H2} A_{H1,H2}\sin(\varphi_{H2}+\varphi_{H1-H2})}{\sum\limits_{H2} A_{H1,H2}\cos(\varphi_{H2}+\varphi_{H1-H2})}\,,$$