Interference effects in dynamic neutron diffraction under conditions of ultrasonic excitation

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Results are reported of a theroetical and experimental study of a new type of Pendellösung beats produced in Bragg diffraction under conditions of ultrasonic excitation. The dependences of the thermal-neutron scattering intensity I on the sound amplitude w are measured in a silicon single crystal. Oscillations of $I(w)$ due to formation of additional gaps on the dispersion surface are observed, as are oscillations of the average slope of the $I(w)$ plots when the sample is rotated around the reciprocal-lattice vector. It is shown that near the threshold frequency the contrast of the oscillations and the average slope of $I(w)$ are determined mainly by the parameter $\cos(\delta q T)$, where δq is the shift of the neutron dispersion surface when the ultrasound is turned on and T is the crystal thickness. The experimental data agree with the theoretical calculations.

1. INTRODUCTION

Ultrasonic (US) oscillations influence substantially the intensity of x-ray and thermal-neutron scattering in perfect single crystals. Depending on the ultrasound frequency ν_s, a distinction can be made between two different mechanisms. At $\nu_s \ll \nu_{th}$ (ν_{th} is a certain threshold value connected with the extinction length τ)[1)] the US strains simply expand the Bragg-scattering angle interval, so that when the sound amplitude w is increased the diffraction intensity rises up to the kinematic limit.[1,2] A transition to higher frequencies alters the situation qualitatively. Ultrasound with $\nu_s > \nu_{th}$ mixes the states that correspond to different sheets of the dispersion surface, and this leads to a number of experimentally observed phenomena, e.g., resonant suppression of the Borrmann effect,[3] additional reflection in the Bragg geometry,[4] and others. Certain new aspects of the interaction between high-frequency ultrasound and radiation was recently observed.[5] It was shown in particular that in neutron diffraction (in contrast to that of x rays), an important role is played by exchange, with the US wave, of not only momentum but also a rather small amount of energy ($\sim 10^{-7}$ eV). It was found that the presence of an US lattice in the crystal leads to the onset of Pendellösung beats that depend on the US oscillation amplitude w. These beats were observed in measurements of neutron-diffraction intensity within the Borrmann delta at $\nu_s > \nu_{th}$ (Ref. 6) and in measurement of the integrated intensity of x-ray scattering ($\nu_s \approx \nu_{th}$).[7]

We report here the results of further theoretical and experimental investigation of US Pendellösung beats as applied to neutron diffraction.

2. THEORY

The general procedure of calculating the intensity of neutron diffraction in the presence of high-frequency ($\nu_s \approx \nu_{th}$) US oscillations is given in Ref. (5). We pay principal attention here to a more detailed analysis of the threshold situation ($\nu_s \approx \nu_{th}$).

When a neutron interacts with an US wave characterized by a wave vector $\mathbf{k}_s$, the dispersion surface of the neutron is shifted in the crystal by a vector $\delta\mathbf{q}_\pm$ (the $\pm$ signs corresponds to absorption and emission of a US phonon)[5]

$$\delta\mathbf{q}_\pm = \mathbf{k}_s \cos\varphi \pm \delta q \mathbf{e}, \qquad (1)$$

where

$$\delta q = 2\pi\nu_s / v_n \cos\theta_B,$$
$$\mathbf{e} = 2^{-1/2}(\mathbf{e}_0 + \mathbf{e}_H),$$

$\mathbf{k}_s \cos\varphi$ is the projection of $\mathbf{k}_s$ on the scattering plane, v_n is the neutron velocity, θ_B is the Bragg angle, and $\mathbf{e}_0$ and $\mathbf{e}_H$ are unit vectors in the propagation directions of the incident and refracted waves. (There is no second term in (1) in the case of x rays.)

If $\nu_s > \nu_{th}$, a displacement of the dispersion surface by $\delta\mathbf{q}_\pm$ causes crossing of certain modes (see Fig. 1). Interac-

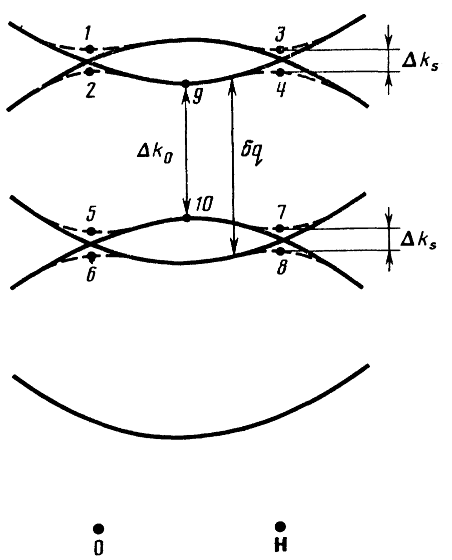

FIG. 1. Dispersion surface of a neutron in a crystal, modified by interaction with ultrasound.

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tion with ultrasound lifts the degeneracy of the states at the crossing points and leads to the appearance of additional gaps $\Delta k_s = \Delta k_0 |\mathbf{H}\cdot\mathbf{w}|$ on the dispersion surface (Δk_0 is the usual "soundless" gap and $\mathbf{H}$ is the reciprocal-lattice vector), and hence to Pendellösung beats governed by the ultrasound.

In the calculation of the diffraction intensity, we confine ourselves for the sake of argument to symmetric Laue reflection in a plane-parallel plate. We assume further that a transverse US wave with polarization directed along the vector $\mathbf{H}$ propagates perpendicular to the scattering plane. Note that in this geometry ultrasound does not influence x-ray diffraction at all (a pure "energy" situation in the terminology of Ref. 5). Using the wave function of the neutron in the crystal from Ref. 5, we obtain after some transformations the scattering-intensity increment ΔI connected with the ultrasound ($|\mathbf{H}\cdot\mathbf{w}| \ll 1$):

$$\Delta I/\overline{I(0)} = 1 + |\mathbf{H}\mathbf{w}\cos\gamma| \left\{ 2\int_0^z J_0(x)\,dx - \cos(\delta q T)\left[\int_0^z J_0(x)\,dx + 2\left.\frac{\partial J_0(x)}{\partial\tau}\right|_{x=z}\right]\right\}, \tag{2}$$

where

$$\sin\gamma = \Delta k_0/\delta q, \quad z = |\mathbf{H}\mathbf{w}|\Delta k_0 T, \quad \overline{I(0)} = {}^1/_4 \pi \Delta k_0 / Q \sin\theta_B,$$

$J_0(x)$ is a Bessel function, T is the plate thickness, $I(0)$ is the scattering intensity in the absence of ultrasound, $\overline{I(0)}$ is the value if $I(0)$ averaged over the usual extinction beats, and Q is the neutron momentum. Let us analyze expression (2) at $T \ll \tau_s = 2\pi/\Delta k_s = \tau|\mathbf{H}\cdot\mathbf{w}|^{-1}$, i.e., $z \ll 1$, we have

$$\Delta I/\overline{I(0)} \approx 2(\mathbf{H}\mathbf{w})^2 \Delta k_0 T |\cos\gamma| \tag{3}$$

and at $T \gg \tau_s$ ($z \gg 1$)

$$\Delta I/\overline{I(0)} \approx 2|\mathbf{H}\mathbf{w}\cos\gamma| \{1 - {}^1/_2 \operatorname{tg}^2\gamma\cos(\delta q T) - (2/z)^{1/2}[1 + {}^1/_2 \operatorname{tg}^2\gamma\cos(\delta q T)]\cos(z + \pi/4)\}. \tag{4}$$

Averaging over the sample thickness leads to vanishing of terms containing $\cos(\delta q T)$, and then expression (4) coincides with Eq. (22) of Ref. 5 for the "sound" increment to the diffraction intensity. Note that no account was taken in the calculations of Ref. 5 of the so-called interband interference of states (the points 1,2 and 5,6 and also 3,4 and 7,8 of Fig. 1). We shall show that this interference is quite appreciable near the threshold. [The explicit forms of the functions $\Delta I(z)/\overline{I(0)}$ are shown in Fig. 2 below.]

If the frequency ν_s approaches the threshold value ν_{th} ($\delta q \to \Delta k_0$), the oscillation contrast [the coefficient of $\cos(z + \pi/4)$ in (4)] increases. The qualitative cause of the increased contrast is that as $\gamma \to \pi/2$ the branches of the dispersion surface no longer intersect but are tangent. As a result, the US perturbation restructures the wave function of the neutron in a much larger phase-space volume. At the threshold, formally, the intensity increment $\Delta I \to \infty$, which is obviously wrong. To describe the threshold case we need a more detailed analysis, whose results are presented below.

It can be shown that the expression obtained in Ref. 5 for the neutron wave function is valid also for the threshold

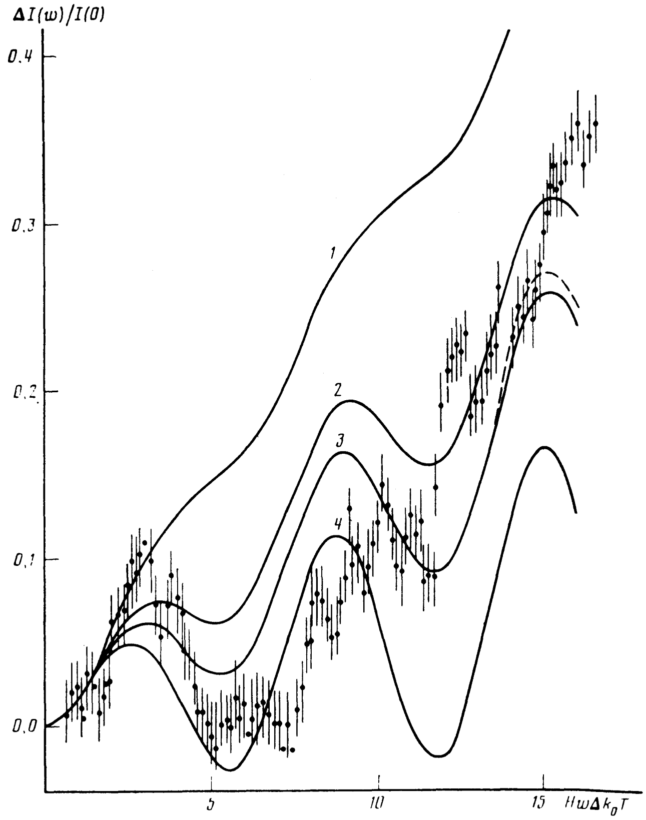

FIG. 2. Increment $\Delta I/I(0)$ to the diffraction intensity vs sound amplitude in Si(2$\bar{2}$0). Points—experiment, solid curves—calculated from Eq. (2) for various values of the parameter $p = \cos(\delta q T)$: 1—$p = -1$; 2—$p = 0$; 3—$p = 0.37$; 4—$p = 1$.

situation. Some specific features appear as $\delta q \to \Delta k_0$ because of the pairwise mutual approach of the points 1,2 and 3,4 or 5,6 and 7,8 on the dispersion surface (Fig. 1). For example, the intensity of elastic scattering of a neutron accompanied by absorption (emission) of an US quantum at threshold is equal to

$$\Delta I_{in}^{+} = \Delta I_{in}^{-} = \frac{\overline{I(0)}}{2\pi}\int d\beta \sin^2\delta \sin^2\frac{z}{\sin\delta}, \qquad \operatorname{ctg}\delta = (1 - 1/\sin\beta)|\mathbf{H}\mathbf{w}|^{-1}. \tag{5}$$

At $T \gg \tau_s$ we have

$$\Delta I_{in}^{+} = \Delta I_{in}^{-} \approx \frac{1}{4}\overline{I(0)}|\mathbf{H}\mathbf{w}|^{1/2}\left[1 - \frac{1}{\pi z^{1/4}}\Gamma\left(\frac{1}{4}\right)\cos\left(z + \frac{\pi}{4}\right)\right]. \tag{6}$$

A similar calculation for the elastic-scattering intensity leads to

$$\Delta I_{el} = \overline{I(0)}\left[\frac{1}{\pi z^{1/4}}\Gamma\left(\frac{1}{4}\right)|\mathbf{H}\mathbf{w}|^{1/2}\sin^2\left(\frac{\Delta k_0 T}{2}\right)\cos\left(z + \frac{\pi}{4}\right) - |\mathbf{H}\mathbf{w}|\cos^2\left(\frac{\Delta k_0 T}{2}\right)\right]. \tag{7}$$

The total scattering intensity at $T \gg \tau_s$ is equal to

$$I = I(0) - \frac{|\mathbf{H}\mathbf{w}|}{2}\overline{I(0)}\cos(\Delta k_0 T)\left[1 + \frac{\Gamma({}^1/_4)}{\pi z^{1/4}}\cos\left(z + \frac{\pi}{4}\right)\right]. \tag{8}$$

The depth of the scattering-oscillation intensity in the threshold situation is $\sim|\mathbf{H}\cdot\mathbf{w}|^{1/4}$, i.e., much larger than at $\delta q > \Delta k_0$, where the oscillation contrast is $\sim|\mathbf{H}\cdot\mathbf{w}|^{1/2}$ [see (4)]. The increase of the contrast near the threshold can be illustrated by using the following arguments.

In the absence of diffraction, the curvature of the free-neutron dispersion surface in momentum space is equal to $\eta_f = Q^{-1}$. In the usual two-wave diffraction the curvature η_d at the extremal points 9 and 10. (Fig. 1) is already

$$\eta_d=2\,\mathrm{tg}^2\,\theta_B/\Delta k_0, \tag{9}$$

or larger than η_f, by five or six orders. Interaction with high-frequency ultrasound ($\nu_s > \nu_{th}$) leads to an even larger curvature of the dispersion surface near the points 1–8 (Fig. 1):

$$\eta_s=\eta_d[1-(\Delta k_0/\delta q)^2]\,|\mathbf{Hw}|^{-1}. \tag{10}$$

This is due to the small value of the US gap at $\Delta k_s \ll \Delta k_0$. As the threshold is approached, the dispersion branches intersect the dispersion surface at ever decreasing angle 2θ:

$$\mathrm{tg}^2\,\theta=\mathrm{tg}^2\,\theta_B[1-(\Delta k_0/\delta q)^2]. \tag{11}$$

The curvature exactly at the threshold is $\eta_s = 0$: planar sections of the dispersion surface are produced and correspond to neutron propagation along the reflection planes. The presence of such sections leads to an increase of the contrast of the scattering-intensity oscillations.[2)]

Let us formulate a criterion for the proximity of δq to the threshold Δk_0. The difference $I - I(0)$ in (8) is transformed into ΔI (4) by acoustic displacements w_0 such that

$$1-(\Delta k_0/\delta q)^2=\cos^2\gamma\sim(|\mathbf{Hw}_0|/\Delta k_0 T)^{1/2}. \tag{12}$$

If $|\mathbf{H}\cdot\mathbf{w}|$ is smaller (larger) than $|\mathbf{H}\cdot\mathbf{w}_0|$, expression (2) or (8) must be used respectively.

3. EXPERIMENT

The measurements were made with the IRT-M reactor of the Physics Institute of the Latvian Academy of Sciences. The neutron diffractometer was described in Ref. 8. We investigated the dependences of the scattering intensity $I(w)$ in a dislocation-free silicon single crystal of 70 mm diameter and $T = 1730 \pm 5$ μm thickness. The surfaces of the plate were electrochemically polished and were parallel to planes of $\{111\}$ type. The quartz piezoconverter (Y-cut) was fastened with epoxy resin without a hardener. A transverse US wave ($\mathbf{w}\|\mathbf{H}$, $\nu_s = 19.250$ MHz) propagated parallel to the reflecting planes [the reflection $(2\bar{2}0)$. Laue symmetric case, was used]. Since the frequency ν_s was relatively close to the threshold value $\nu_{th} = 14.7$ MHz, it was necessary to take into account in the analysis of the diffraction pattern the interband interference, i.e., the terms containing cos (δqT) in expression (4). The theroetical $\Delta I/\overline{I(0)}$, curves calculated with the aid of (2) are highly sensitive to changes of the phase δqT (see Fig. 2).[3)] It was already mentioned that an important role is played in neutron diffraction by energy exchange with the US wave; corresponding to this exchange is the second term of Eq. (1) for the dispersion-curve shift that depends on the neutron wavelength $\lambda \sim 1/n_n$. In our experimental conditions ($\delta qT \approx 70$) a change of λ by 5% corresponds to a shift of the phase δqT by π. To observe oscillations of $I(w)$ it is therefore necessary to make λ rigidly steady.

We used in the measurements a quasimonochromatic neutron beam from a Pb crystal monochromator with mosaic angle $\eta \approx 4'$ [reflection (111), $\theta_{mB} \approx 11°$, maximum of spectrum corresponding to $\lambda = 1.01$ Å].[8] Under conditions of rather tight angular collimation, $\alpha \approx 5'$, the ratio $\Delta\lambda/\lambda$ is determined mainly by the parameter η and is equal to $\Delta\lambda/\lambda \approx \eta/\theta_{mB} \approx 1\%$. It is necessary also to take into account in the experiment also the inhomogeneous distribution of the sound field over the sample (see Ref. 6). From this standpoint it is necessary to decrease to a minimum the geometric dimensions of the incident beam. We used for this purpose a 2×1 mm slit on the entrance surface of the sample.

To eliminate the influence of slow neutron-flux oscillations and of the reactor background, which play a major role at low counting rate in the detector (≈ 5 count/sec), the following automated measurement regime was used. The modules of the CAMAC apparatus were used to amplitude-modulate the US oscillations by a sawtooth voltage $V \sim w$ with a period 10^{-2} s. The "sawtooth" and the time mode of the AI-4096 multichannel analyzer were turned on simultaneously. Each value of w_1 corresponded thus to a definite analyzer channel where the information $I(\omega_i)$ from the neutron detector was accumulated by multiple cycling the entire range of w ($w_{\max}$) corresponded to a voltage $V = 0.18$ V on the piezoconverter.

The normalized experimental data $[I(w) - I(0)]/I(0) = \Delta I(w)/I(0)$ are shown in Fig. 2. The tie-in on the abscissa axis (the $V \leftrightarrow w$ correspondence) was to the position of the first maximum. The growth of the diffraction intensity at small w agrees qualitatively with expression (3). Oscillations corresponding to extinction lengths $\tau_s \sim T \approx 10\tau$ are described by the factor cos $(z + \pi/4)$ were next observed. Figure 2 shows two oscillations; at large w the contrast of the oscillations decreases, apparently because the sound field is not uniform. For multimode US oscillations excited in the sample, the intensity increments due to different modes is additive, and the oscillations themselves become "smeared out." On the whole, the experimental data reveal a behavior close to that expected from theory, although not in good agreement in some sections with curve 3 corresponding to the real conditions cos $(\delta qT) = 0.37$ [the dashed line shows the results of calculating curve 3 but with allowance for the changed width of the main gap,[7] $\Delta k' = J_0(Hw)\Delta k_0 \approx \Delta k_0(1 - H^2w^2/4)$ for US excitation]. It can be stated at any rate that this experiment revealed anew Pendellösung beats over the extinction length τ_s, and the significant role of interband interference between the states near the threshold was confirmed.

Let us analyze once more interesting manifestation of interference states that differ by δq (i.e., are located on different sheets of the dispersion surface. For $z \gg 1$ (but $Hw \ll 1$) we find that the plot of the increment $\Delta I/I(0)$ of the scattering intensity is a straight line $\Delta I/I(0) = a|\mathbf{H}\cdot\mathbf{w}|$ with slope

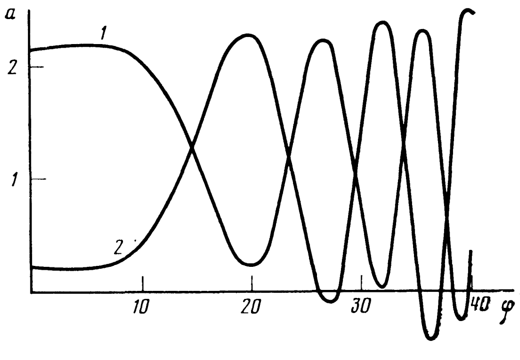

FIG. 3. Calculated $a(\varphi)$ curves for two values of $p = \cos(\delta q T_0)$: 1—$p = -1$; 2—$p = 1$.

$$a = 2|\cos\gamma| - \sin^2\gamma \cos(\delta q T)/|\cos\gamma|, \qquad (13)$$

that depends directly on $\cos(\delta q T)$.

The phase $\delta q T$ can be easily varied in the experiment by tilting the crystal plate about the direction of the vector **H**. Starting from (1), we get in this case

$$\delta q T = \delta q_+ T = k_s (\cos\varphi + v_s/v_n \cos\theta_B) T_0/\cos\varphi$$
$$= k_s T_0 (1 + v_s/v_n \cos\theta_B \cos\varphi), \qquad (14)$$

where $v_s = 2\pi\nu_s/k_s$ is the speed of sound, φ is the tilt angle and is equal to the angle between k_s and the scattering plane, $T = T_0/\cos\varphi$ is the path traversed by the neutron in the tilted plate, and T_0 is the initial real crystal thickness (at $\varphi = 0$). The $a(\varphi)$ plots calculated from (13) are shown in Fig. 3. (We call attention to the high sensitivity of the curves to the value of the initial phase $\delta q T_0$.) Note that this variant of the procedure cannot be used for x rays, since the shift $\delta q_\pm$ of the dispersion surface is specified only by the first term of (1) and accordingly the phase $\delta q_\pm T = k_s T_0$ is independent of φ.

We used the tilt method to study the oscillations of $a(\varphi)$. We measured the $\overline{I(w)}$ dependence at different values of φ in the automatic regime described above, but used in lieu of the AI-4096 analyzer the computer-based IVK computation unit. We averaged over the oscillations by partial summation of information in groups of 64 channels (in a total of 1024 channels). The cycling range was expanded for the same purpose ($V_{\max} = 0.5$ V). To increase the counting rate, we also increased somewhat the geometric dimensions

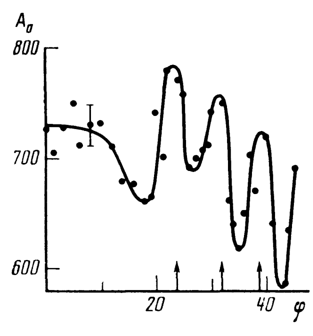

FIG. 4. $A_0(\varphi)$ oscillations. The arrows show the positions of the maxima of the scattering intensity $I(0)$ (without sound), measured in an independent "manual" experiment by the tilt method.

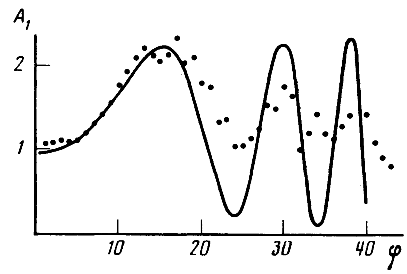

FIG. 5. Oscillations of average tilt $A_1(\varphi)$ of the function $I(w)$. Points—normalized experimental data, solid curve—calculated from Eq. (13) for $\cos(\delta q T) = 0.37$ with ultrasound.

of the incident beam (entrance slit 4×2 mm). The equations were reduced by least squares (a fit to the form $\overline{I} = A_0 + A_1 N + A_2 N^2$, where N is the number of the group of 64 channels and is proportional to w).

Figure 4 shows the results for the coefficient A_0. The $A_0(\varphi)$ ($w = 0$) oscillations are well known[9]: these are the usual extinction beats connected with $\tau = 2\pi/\Delta k_0$. Their appearance, when the plate is tilted, as a result of the phase change $\Delta k_0 T = \Delta k_0 T_0/\cos\varphi$. The period of the $A_0(\varphi)$ oscillations agrees with the theoretical predictions (see, e.g., Ref. 9).

Figure 5 shows a plot of $A_1(\varphi)$, obtained for the first time. The points correspond to the averaged values of $[A_1(\varphi) + A_1(-\varphi)]/2A_1(0)$. The figure shows also for comparison the functions $a(\varphi)$ calculated from Eq. (13) with an initial phase ($\varphi = 0$) corresponding to the real experimental conditions $\cos(\delta q T_0) = 0.37$. The position and depth of the first oscillations (which reflect the fact that the dispersion-surface branches intersect when high-frequency ultrasound is excited) agree well with the theory. The better data of Fig. 5 (compared with Fig. 2) can be readily explained, since the average slope is a less sensitive parameter than the $I(w)$ oscillation curve.

4. CONCLUSION

Noticeable progress was made in the last few years in the technique of inelastic scattering of neutrons. Spin echo spectrometers and backward-scatter spectrometers deal reliably with an energy-transfer range of order 10^{-6} eV. Even so high a resolution, however, is insufficient for a number of problems. In our case, for example, the energy transfer in inelastic scattering by an US phonon ($\nu_s \approx 20$ MHz) is $8\cdot 10^{-8}$ eV. It is all the more interesting that exchange of so low an energy with an US wave is revealed in experiment without an energy analysis of the scattered beam. In fact, since the velocities of the thermal neutrons and of the sound waves in the crystal are of the same order, absorption of the phonon energy upon diffraction shifts additionally the neutron dispersion surface by an amount comparable with $k_s \sim \Delta k_0$. In view of the importance of energy exchange, the neutron diffraction pattern in the presence of US oscillation is somewhat richer and the possibility of controlling it is somewhat better than in the case of x rays. For neutrons, in particular, there are two threshold frequencies corresponding to traveling waves with momenta $\pm k_s$. The diffraction is strongly influenced by US oscillations propagating perpendicular to the scattering plane. By tilting the crystal around the reciprocal-lattice vector it is possible to observe the oscillations of the average slope of $\overline{I(w)}$. The principal effect, however, namely the appearance of a new large extinction length $\tau_s \gg \tau$ in the crystal, is independent of the type of radiation. Additional gaps $\Delta k_s = 2\pi/\tau_s$ on the dispersion surface lead to scattering-intensity oscillations when the US oscillation amplitude is varied. The presence of strong bending of the dispersion curve near the branch intersection points gives grounds for hoping the diffraction to be more sensitive in the presence of ultrasound to distortions of the crystal lattice, compared with ordinary topography.

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1)In the case of x rays the threshold situation is realized when the ultrasound wavelength λ_s is equal to the extinction length τ.

2)The geometry of the extremal points in the vicinity of the extremal points can be described with the aid of the reciprocal-effective-mass tensor.

3)Although expression (2) was derived for the purely energetic situation, it can be used also to describe our experiments, but with δq replaced by δq_+ corresponding to the lower threshold frequency $\nu_{th} = 14.7$ MHZ, near which the measurements were made.

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