Brillouin scattering study of pseudosurface acoustic wave propagation in (110)- and (111)-cut PbTe crystals

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The propagation of surface and pseudosurface acoustic waves in the (110) and (111) planes of a PbTe crystal was studied for the first time by the method of Brillouin spectroscopy. Investigation of the azimuthal dependence of the width of the component corresponding to the pseudosurface modes for the (110) plane revealed the existence of an additional isolated direction (different from $[1\bar{1}0]$) in which the pseudosurface modes degenerate into a Rayleigh surface acoustic wave.

Among treatments of the dynamical properties of the surface of opaque solids, high-resolution Brillouin studies of the surface phonon density of states SPDS for long-wavelength phonons are of significant interest.[1,2]

Theoretical analysis of the SPDS shows that the spectrum should contain, in addition to a peak corresponding to generalized surface waves (GSW),[3,4] an entire continuum of phonon states corresponding to internal elastic Debye waves.[2,5–10]

In addition to the Brillouin satellite corresponding to GSW, a wide plateau adjoining the high-frequency edge of the GSW satellite is observed in the spectrum of the light scattered by the surface of the sample. The form of this plateau strongly depends on the anisotropy of the elastic properties of the experimental material as well as on the geometry of the experiment. Thus, in many cases the continuous spectrum can contain intense spectral components corresponding to pseudosurface modes.[2,9,11] These waves are characterized by the fact that their wave vector tilts away from the surface into the crystal and, as a result, it may be possible to record the corresponding Brillouin lines in a Brillouin experiment. The spectral components corresponding to both GSW and pseudosurface modes have been detected simultaneously in (111)-cut GaAs,[11,12] InSb,[13] and InAs[14] crystals in a wide range of azimuthal angles in the (111) plane. Intense Brillouin components corresponding to pseudosurface modes have also been observed in the case of *z*-cut Ge,[15,16] Si,[1,17] InSb and GaAs,[14,17] and Ni[18] cubic crystals for azimuthal directions close to [110].

The object of the present work was to use Brillouin spectroscopy to study the SPDS and the propagation of pseudosurface waves in a PbTe crystal.

Lead telluride crystals are characterized by a sharp anisotropy of elastic properties, characterized by the parameter $\mu=2C_{44}/(C_{11}-C_{12})=\mu_{\text{PbTe}}=0.307$,[14,19] where C_{ij} are the corresponding moduli of elasticity. Only three observations of the spectral composition of Brillouin light in crystals having an elastic anisotropy of this type ($\mu<1$) have been reported in the literature,[1,20,21] and the pseudosurface Brillouin satellite was detected only in Ref. 21. We also note that in these crystals a pseudosurface branch is characteristically present for the (111) and (110) planes, while for samples with $\mu>1$, for example, Si or GaAs, a pseudosurface branch is characteristically present for the (001) and (111) planes.[4]

A Burleigh spectrometer, described previously in Ref. 22 and equipped with a five-pass piezoelectrically scanned Fabry–Perot interferometer, was used to record the surface scattering spectra of the PbTe single crystal. A single-frequency argon laser ($\lambda=514.5$ nm, ~ 60 mW) was used for the scattering experiment. The light scattered by the crystals was observed at an angle of 180° with respect to the beam incident on the crystal. In all experiments the electric-field vector **E** of the incident light wave was parallel to the plane of incidence of the light.

The SPDS was calculated for the (110) and (111) planes of the PbTe crystal by the method developed in Ref. 9 for calculating the spectra of equilibrium Debye elastic surface excitations of cubic crystals. The computed SPDS were compared with the experimentally recorded Brillouin spectra.

EXPERIMENTAL STUDY OF THE SURFACE PHONON DENSITY OF STATES FOR THE (110) PLANE OF A PbTe CRYSTAL

Examples of the spectra of light scattered by a (110)-cut PbTe crystal for the angles $\theta_{(110)}=0°$ (a), 76° (b), and 90° (c), where $\theta_{(110)}$ is the azimuthal angle between the plane of incidence of the light and the $[00\bar{1}]$ crystallographic axis, are displayed in Figs. 1a–c. The section of the samples by the plane of incidence is shown schematically in the inset in Fig. 1a. Here $\alpha=70°$ is the angle of incidence of the light and $\mathbf{k}_i$, $\mathbf{k}_s$, and $\mathbf{q}$ are, respectively, the wave vectors of the incident and scattered light and the GSW. The velocities

$$V=\Delta f\frac{\lambda}{2\sin(\alpha)}$$

are plotted, together with the frequency Δf, along the abscissa. The pedestal in the Brillouin satellites is due to the conditions under which the spectra were recorded.[19]

Figure 2 displays the measured velocities of surface waves in the (110) plane as a function of the angle $\theta_{(110)}$. The theoretically computed curves for the velocities of

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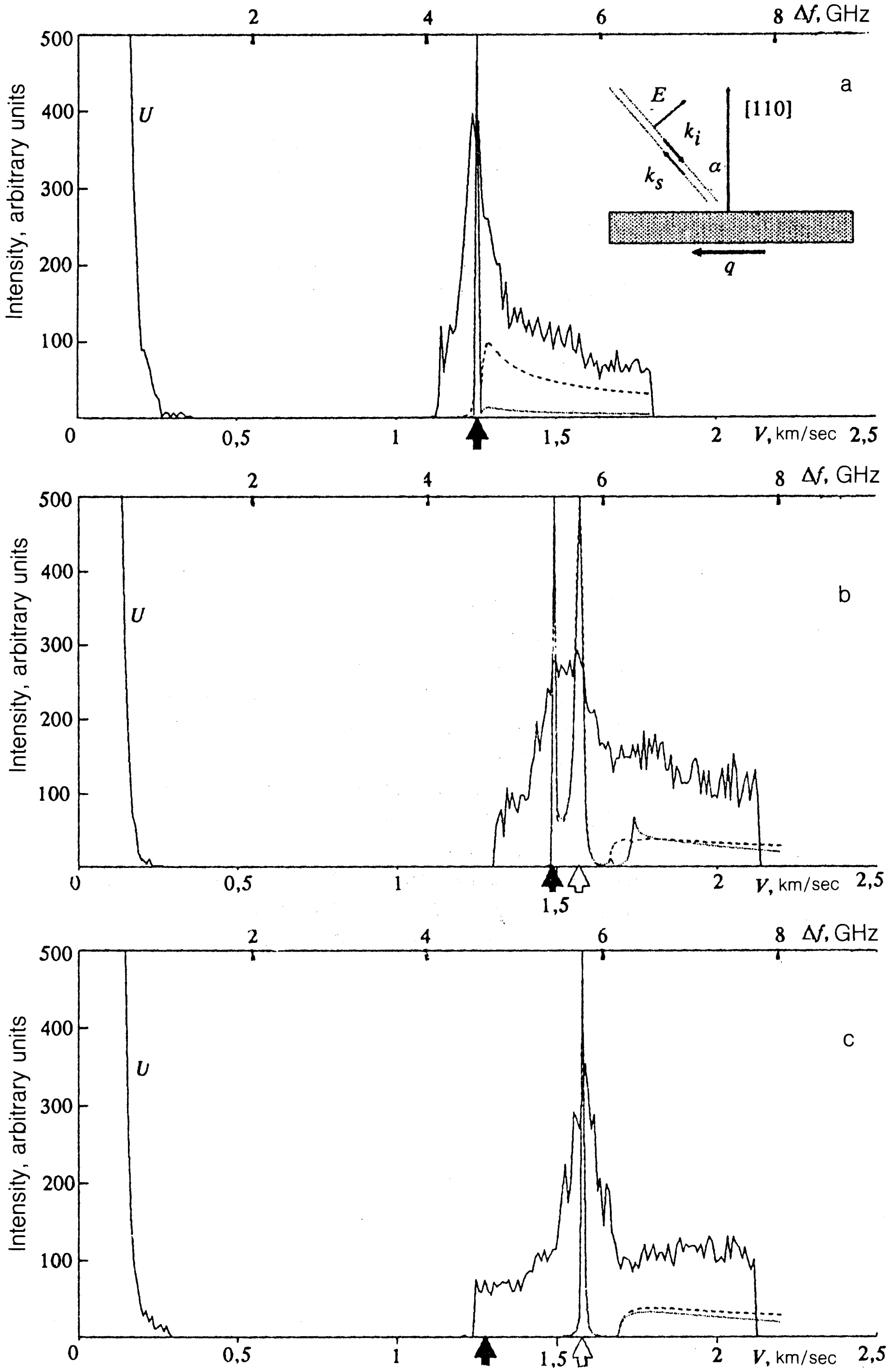

FIG. 1. Spectra of light scattered by a (110)-cut PbTe crystal for $\theta_{(110)}=0°$ (a), 76° (b), and 90° (c); $\alpha=70°$; and, U is the initial spectral line. The computed SPDS spectrum (the dotted and dashed lines correspond, respectively, to the normal and tangential components of the surface excitations) was calculated using the values given in Ref. 19 for the material parameters and is plotted in arbitrary units along the ordinate. The arrows on the abscissa correspond to the values of the GSW (dark-colored) and pseudosurface mode (light-colored) velocities.

GSW, pseudosurface modes, and the quasitransverse acoustic waves $T1$ and $T2$ are also displayed in Fig. 2.

As one can see from Figs. 1 and 2, for angles in the range $\theta_{(110)}=0°–74°$ the position of the fundamental Brillouin line in the spectrum corresponds to GSW and for $\theta_{(110)}=78°–90°$ it corresponds to pseudosurface modes.

This change in the position of the Brillouin line from GSW to pseudosurface modes as a function of the azimuthal angle is described well by redistribution of intensity in the acoustic SPDS spectra for both normal and tangential excitations of the surface of the sample (the dotted and dashed lines in Figs. 1a–c). For the deformation mechanism of surface scattering of light the normal component of the surface oscillations determines the spectral composition of the Brillouin light, while in the presence of strong elastooptic surface interaction the tangential component influences the spectral composition of the scattered light.[2]

The calculations also show that for intermediate angles $\theta_{(110)}\approx 75°–78°$ the SPDS spectra can contain two intense spectral components corresponding to both GSW and pseudosurface modes. We were not able to resolve them separately, but in this frequency range the broadening of the Brillouin line in the spectrum is noticeable (Fig. 1b; $\theta_{(110)}\approx 76°$).

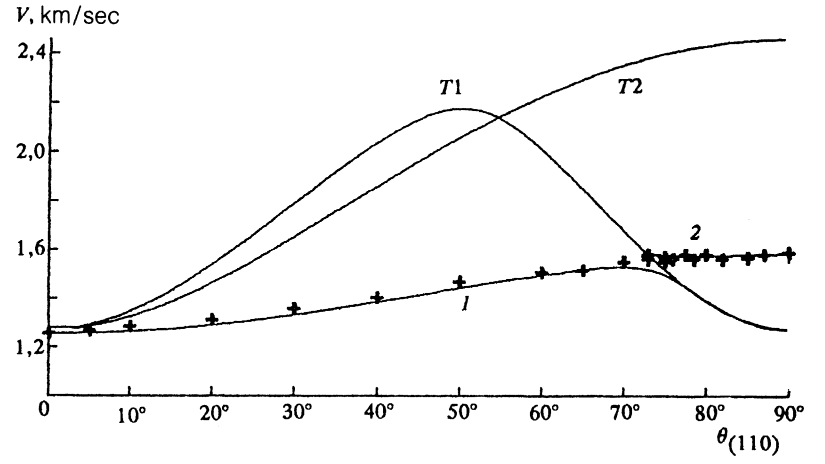

FIG. 2. Velocities of GSW, pseudosurface, $T1$, and $T2$ acoustic waves for the (110) plane in PbTe as a function of the angle $\theta_{(110)}$. The crosses are the experimental values of the GSW and pseudosurface mode velocities; the solid lines are the theoretical curves for GSW (*1*), pseudosurface (*2*), $T1$, and $T2$. The values of the material parameters for the curves presented were taken from Ref. 19.

In Ref. 15 we observed the change in the spectral composition of the Brillouin light scattered by a PbS crystal ($\mu_{\rm PbS}=0.508^4$) with increasing azimuthal angle $\theta_{(110)}$. We were able to follow not only the transition from the GSW to pseudosurface modes, but we also resolved experimentally at $\theta_{(110)} \approx 74°$ experimentally both Brillouin spectral components simultaneously (Fig. 6a in Ref. 15).

It turns out that the intensity redistribution in the SPDS acoustic spectrum for normal and tangential surface waves as a function of $\theta_{(110)}$ in cubic crystals with $\mu < 1$ is analogous to the corresponding intensity redistribution in the SPDS spectrum for z-cut cubic crystals with elastic anisotropy parameter $\mu > 1$,[9,14] where for azimuthal directions close to [100] the position of the fundamental Brillouin satellite in the spectrum of the light scattered by the crystal corresponds to GSW and for directions close to [110] it corresponds to pseudosurface modes. In addition, both Brillouin lines, corresponding to GSW and pseudosurface modes, are recorded in a narrow range of directions.[14,16,18]

Besides determining the velocities of the generalized and pseudosurface waves from the Brillouin spectra, we also measured the Brillouin widths δf_B of the pseudosurface mode lines for different azimuthal angles of the directions of propagation of these waves. We found that the width δf_B passes through a minimum at $\theta_{(110)} \approx 82°$ and 90° (see Fig. 3). In order to determine δf_B we performed the corresponding deconvolution of the obtained spectrum (we do not explain here the method that we employed for processing the obtained spectra).

The calculations showed that over a wide range of azimuthal angles $\theta_{(110)}$ a Lorentzian curve describes the profile of the pseudosurface line in the SPDS well. Our calculations for the case of z-cut cubic crystals[14] as well as the calculations presented in Ref. 11 show that the absorption constants $\alpha_{\rm attn}$ obtained for pseudosurface waves by solving the equations of motion[4] are in good agreement with the values of $\alpha_{\rm attn}$ obtained by fitting a Lorentzian profile to the pseudosurface profile. This is justified in the computed spectra for both normal and tangential surface excitations. For this reason, in the present work $\alpha_{\rm attn}$ was determined from the profile of the pseudosurface mode line in the SPDS spectrum.

The computational results for $\alpha_{\rm attn}$ as a function of $\theta_{(110)}$ are also displayed in Fig. 3 (solid curve).

According to Fig. 3, the positions of both minima of δf_B agree well with the positions of the zeroes of $\alpha_{\rm attn}$.

This shows that the minimum values of δf_B corresponds to pseudosurface mode propagation directions for which the volume partial component, responsible in the case of an acoustic signal introduced from outside the sample, for the energy flux directed into the medium, vanishes and the component itself degenerates into a Rayleigh wave.

The minimum of δf_B at $\theta_{(110)} = 82°$ corresponds to an

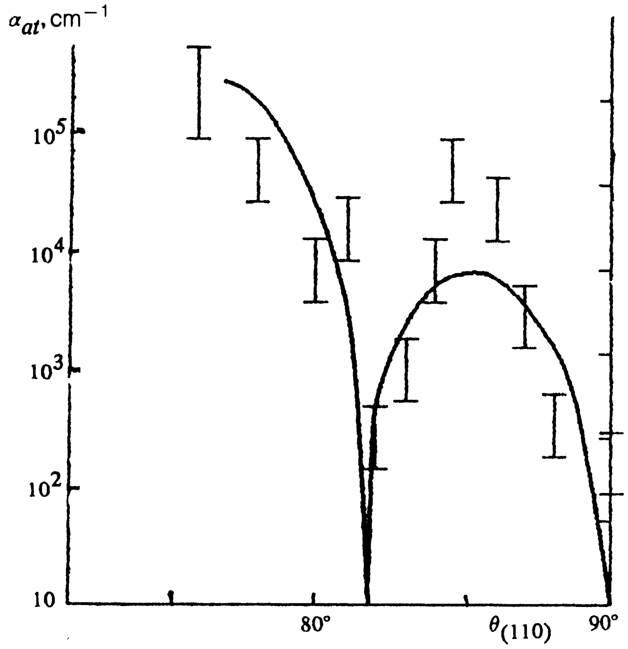

FIG. 3. Pseudosurface mode absorption as a function of the angle $\theta_{(110)}$ for (111)-cut PbTe. The experimental error is indicated. The solid line was computed. The values of the material parameters for the curve were taken from Ref. 19.

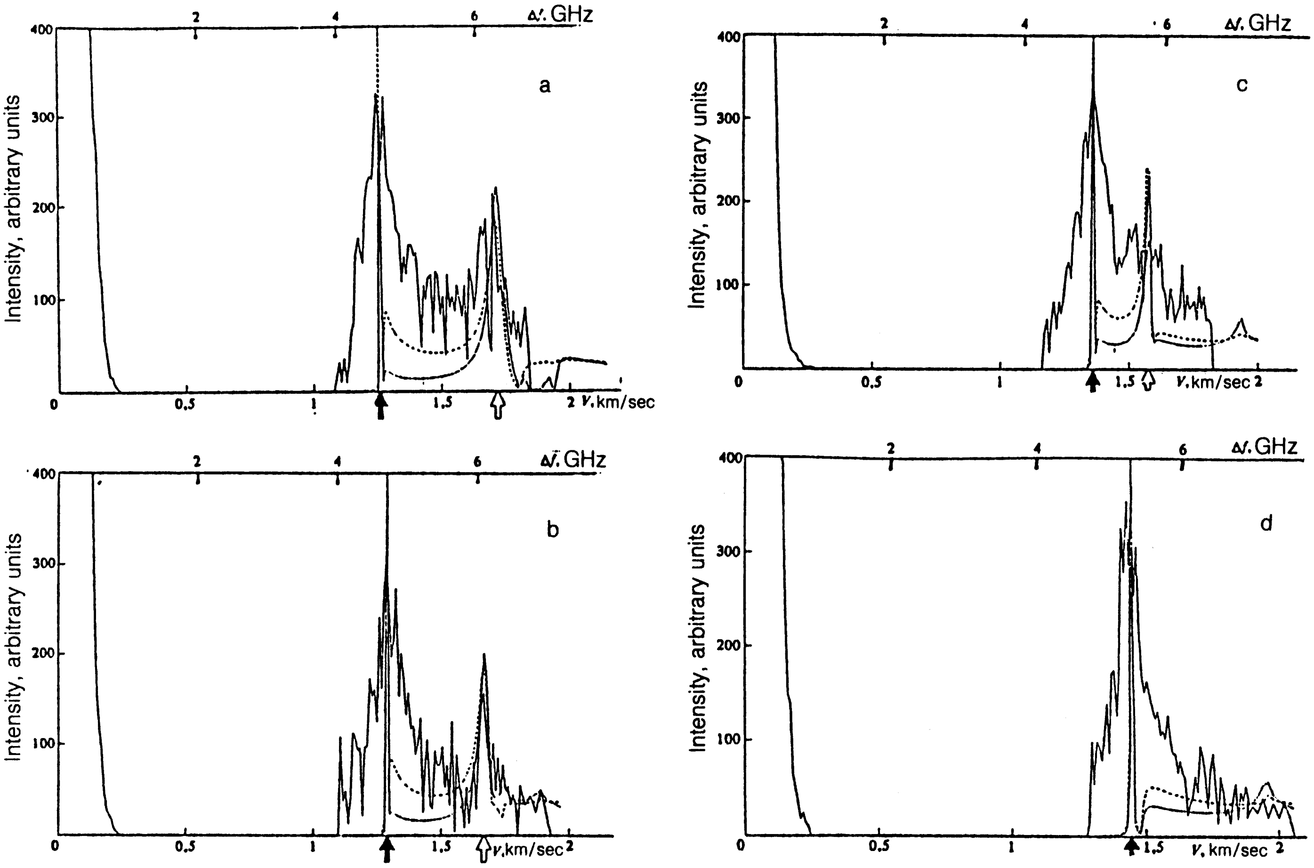

FIG. 4. Spectra of light scattered by a (111)-cut PbTe crystal for $\theta_{(111)}=0°$ (a), 10° (b), 20° (c), and 30° (d); $\alpha=70°$. The arrows on the abscissa correspond to the GSW (dark-colored) and pseudosurface mode (light-colored) velocities. The computed SPDS spectrum of equilibrium surface excitations was calculated using the values given in Ref. [19] for the material parameters and is plotted in arbitrary units along the ordinate.

additional isolated direction in the (110) plane of the PbTe crystal along which the pseudosurface mode is once again two-part wave.[4]

Such an additional isolated direction was previously predicted for KCl crystals ($\mu_{KCl}=0.375$) for $\theta_{(111)}=79.4°$.[23] We previously observed experimentally such additional isolated directions in z-cut Si, Ge, GaAs, and InSb crystals.[15,17] Such a direction was also obtained in the (001) plane of a GaAs crystal by the ultrasonic method.[24]

EXPERIMENTAL STUDY OF THE SURFACE PHONON DENSITY OF STATES FOR THE (111) PLANE OF A PbTe CRYSTAL

Examples of spectra of light scattered by a (111)-cut PbTe crystal for $\theta_{(111)}=0°$ (a), 10° (b), 20° (c), and 30° (d) are displayed in Figs. 4a–d. Here $\theta_{(111)}$ is the angle between the $[1\bar{1}0]$ crystallographic direction and the plane of incidence.

The line corresponding to GSW is clearly seen in all spectra. The B component corresponding to pseudosurface modes is recorded separately in the range $\theta_{(111)}=0°$–20° (Figs. 4a–c). As $\theta_{(111)}$ increases further, pseudosurface mode component merges with the high-frequency edge of the fundamental B satellite (Figs. 4c–d). The spectral composition of the computed SPDS, for both normal and tangential surface excitations, is correlated with the spectral composition of the scattered light for all values of $\theta_{(111)}$ (see Figs. 4a–d, dashed and dotted lines).

It was found that over a wide range of angles $\theta_{(111)}$ the behavior of the B satellites corresponding to pseudosurface mode is qualitatively similar to the behavior observed in crystals with $\mu>1$, where the pseudosurface Brillouin satellite was recorded reliably in GaAs in the range $\theta_{(111)}=0°$–20° (Ref. 11) and 0°–18° (Ref. 12) and in InSb and InAs in the range 0°–15°.[13,14]

The measured velocities of surface waves in the (111) plane as a function of the angle $\theta_{(111)}$ are displayed in Fig. 5. The figure also displays the theoretical curves for the velocities of GSW, pseudosurface modes, and $T1$ and $T2$ waves.

The experimentally determined values of the GSW velocity agree well in the entire range of angles $\theta_{(111)}=0°$–30° with the corresponding theoretical curve, and in the case of a pseudosurface wave they agree in the range $\theta_{(111)}=0°$–20°. We note that, in contrast to crystals with $\mu>1$, for PbTe the pseudosurface and $T2$ curves characteristically intersect ($\theta_{(111)}\approx 20°$). This feature of cubic

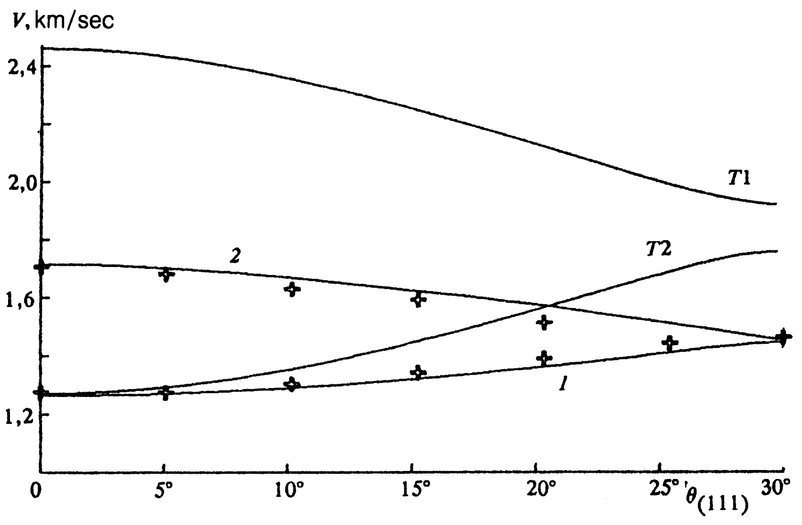

FIG. 5. Velocity of surface and volume acoustic waves for the (111) plane of a PbTe crystal as a function of the angle $\theta_{(111)}$. The crosses are the experimental values of the GSW and pseudosurface mode velocities. The values of the material parameters for the theoretical curves, corresponding to GSW (*1*), pseudosurface (*2*), *T*1, and *T*2, were taken from Ref. 19.

crystals with $\mu < 1$ was first noted in Ref. 25 for KCl.

In Ref. 1, where *B* scattering in a Cr crystal was observed ($\mu_{Cr} = 0.70$), it was found that the high-frequency part of the *B* component is appreciably deformed. This deformation probably occurs because the pseudosurface mode line was not separately resolved. This fact was already indicated in Ref. 26.

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