Study of surface Debye waves on the faces of GaAs single crystals by molecular-light-scattering spectroscopy

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Surface excitations in the hypersonic range in a GaAs crystal have been studied by Brillouin scattering. The Brillouin scattering spectra are used to study the propagation of generalized surface wave and pseudosurface modes for various directions in the (001) and (111) planes of these crystals.

Brillouin scattering is an effective method for detailed studies of equilibrium surface phonon spectra of opaque crystals.

Experimental studies of Brillouin scattering in metals and semiconductors carried out by Dill and Brody[1] and Sandercock[2] stimulated the development of a theory for the scattering of light by the surfaces of opaque crystals (Louden,[3] Subbuswamy and Maradudin,[4] Rowell and Stegeman,[5] Marvien *et al.*,[6] and Velasco and Garcia–Moliner[7]). Another direction of research has consisted of numerical calculations of the velocities and absorption of surface waves in crystals with various physical properties (Lim and Farnell[8] and Glass and Maradudin[9]).

In both cases, analysis has shown that in addition to the Rayleigh surface wave [or the so-called generalized surface wave (GSW)], with a real wave vector which lies in the plane of this surface, there may exist two other pseudosurface waves, which are characterized by a complex wave vector slanted from the surface into the crystal. These waves differ from generalized surface waves in that they are damped along the surface, since the tangential component of their wave vector has an imaginary part.

One of these waves, the pseudosurface mode (PSM), propagates in certain directions on the surfaces of anisotropic substances at velocities $V_{\rm PSM}$ which satisfy

$$V_{t2}<V_{PSM}<V_{t1},$$

where V_t1 is the velocity of a fast transverse bulk wave for the same direction, and V_{t2} is the velocity of the corresponding slow transverse wave.

The other mode, a high-frequency pseudosurface mode (HFPSM), which also exists in the case of an isotropic substance, is characterized by a velocity $V_{\rm HFPSM}$ which satisfies the inequalities

$$V_{t1}<V_{\rm HFPSM}<V_L.$$

Here V_L is the velocity of a longitudinal bulk wave in the same direction.

In this paper we report an experimental study of surface excitations in the hypersonic range in (001)- and (111)-cut cubis GaAs crystal by Brillouin spectroscopy. These crystals present a unique opportunity for observing both PSMs and HFPSMs, and the crystals themselves have a fairly high anisotropy parameter[8]

$$\mu=2C_{44}/(C_{11}-C_{12})=1{,}80\quad[8],$$

where C_{11}, C_{12}, and C_{44} are corresponding elastic moduli.

The light scattered from the surface of the GaAs crystal was observed at an angle of 180°. The conditions under which the Brillouin scattering was detected were described in detail in Refs. 13 and 14. The observations were carried out at room temperature. The electric vector **E** of the light wave incident on the crystal was parallel to the plane of incidence in all the experiments. The light source was a single-frequency argon laser with a wavelength of 514.5 or 488.0 nm, with a power ≈ 100 mW in each line.

LIGHT SCATTERED BY A GaAs (001) CRYSTAL

Figure 1 is a spectrum of the light scattered by a *z*-cut GaAs crystal for $\theta_{(001)}=0°$, where $\theta_{(001)}$ is the angle between the plane of incidence of the light and the (110) crystallographic plane. We can clearly see a line which corresponds to a GSW. The corresponding frequency shift is plotted along the abscissa, where the normalized coordinates V/V_{t1} are also introduced:

$$V=fl/(2\sin\alpha),\quad V_{t1}=(C_{44}/d)^{0,5},$$

where f is the frequency of the incident light, l is its wavelength, $\alpha(=70°)$ is the angle at which the light is incident on the sample, and d is the density of the GaAs sample.

Surface Brillouin scattering was studied for various orientations of the plane of incidence of the light on the sample. Corresponding spectra were recorded at $\theta_{(001)}$ steps of 0.5–5° over the range $\theta_{(001)}=0°-50°$.

Figure 2 shows the measurements of the surface–wave velocities on the (001) plane as a function of the angle $T_{(001)}$. Also shown here are some theoretical curves for the velocities of GSW, PSM, T1 and T2 waves.

It can be seen from Fig. 2 that in the angular interval $\theta_{(001)}=0°-23°$ the positions of the Brillouin scattering lines detected in this experiment agree well with the corresponding GSW curve, and at $\theta_{(001)}=28°-45°$ C they agree well with the PSM curve.

The "jump" from the GSW branch to the PSM branch observed in these experiments corresponds qualitatively to the results of similar studies of crystals which exhibit scattering by surface deformations: Ge (Refs. 14 and 15), Si (Ref. 2), and Ni (Ref. 16). Note, however, that crystals with an elastooptic mechanism for surface scattering of light, such as GaAs, differ from these other crystals in that no theoretical analysis has been carried out of the jump from the GSW branch to the PSM branch with a change in $\theta_{(001)}$.

A series of spectral measurements with high frequency resolution for intermediate angles $\theta_{(001)}$ ($\theta_{(001)}=26°$

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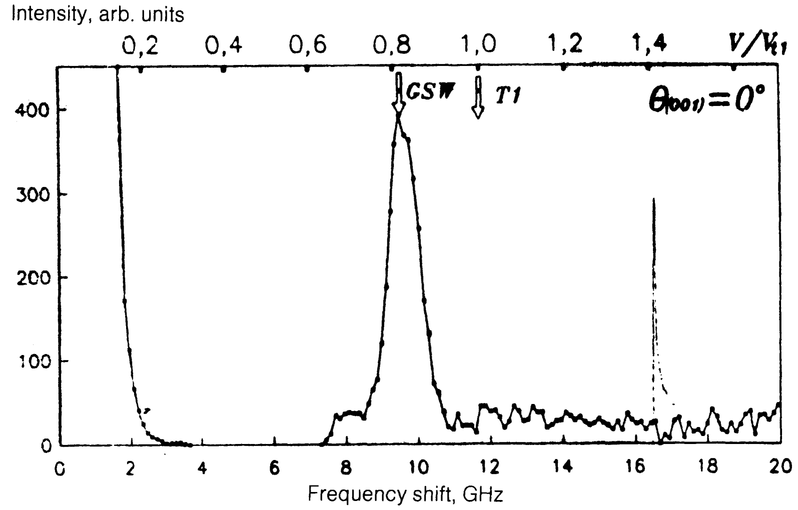

FIG. 1. Spectrum of the light scattered by a GaAs crystal for $\theta_{(001)} = 0°$, $\alpha = 70°$, $i = 514.5$ nm. The frequency shift, plotted along the abscissa, is also shown in normalized form as V/V_{t1}. The theoretical (dotted) curve was calculated with the help of material properties from Ref. 18.

$-28°$) allowed us to simultaneously detect spectral lines corresponding to GSWs and PSMs (Fig. 3). It can be seen from this figure that for $\theta_{(001)} = 25°$ (Fig. 3a) the scattered light spectrum contains only a satellite corresponding to a GSW. As $\theta_{(001)}$ increases, the competition between the scattered light due to GSWs and PSMs results in progressive replacement of the GSW component of the spectrum by a satellite associated with PSM (Fig. 3,b–d).

The simultaneous observation of both GSW and PSM components in the angular region $\theta_{(001)} \approx 25°$ in the GaAs crystal is very similar in a qualitative sense to the simultaneous detection of GSW and PSM satellites in the case of a Ge crystal,[14] for which there is a deformation mechanism for surface scattering.

When the conditions for observing and recording the Brillouin spectra were comparable to those of the corresponding experiments of Refs. 2 and 14, at no value of the angle $\theta_{(001)}$ were we able to detect a component corresponding to an HFPSM. The presumed position of the HFPSM satellite in the Brillouin spectrum for $\theta_{(001)} = 0°$ is shown by the maximum of the theoretical curve (Fig. 1). This curve was calculated from Eq. (2) of Ref. 11.

LIGHT SCATTERED BY A GaAs (111) CRYSTAL

In a (111)-cut GaAs crystal we also studied the surface Brillouin scattering, for various values of the angle $\theta_{(111)}$, which was varied from 5° to 40° in steps of 1°–5° (here $\theta_{(111)}$ is the angle between the plane of incidence of the light on the sample and the $[1\bar{1}0]$ crystallographic direction).

Figure 4a shows the spectrum of the light scattered by

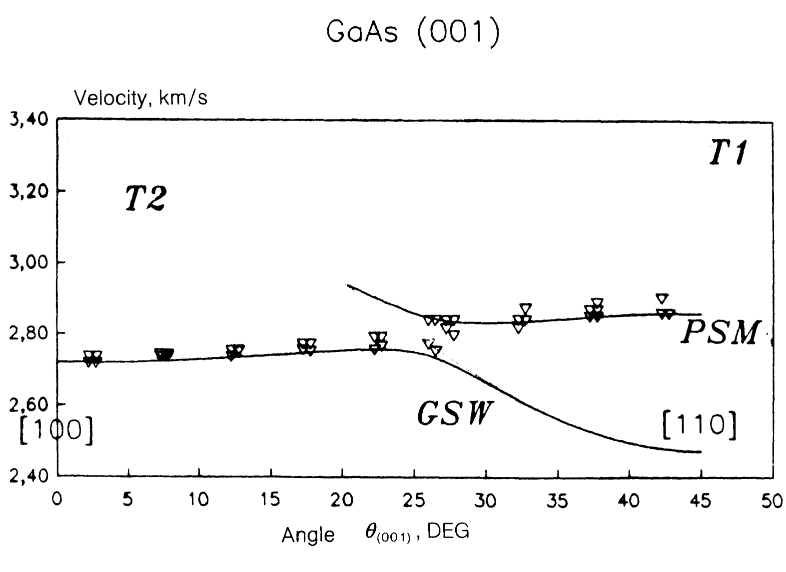

FIG. 2. Velocities V_{GSW}, V_{PSM} V_{t1} and V_{t2} for the (001) plane versus the angle $\theta_{(001)}$. Triangles—Experimental values of V_{GSW} and V_{PSM}; solid lines—theoretical for GSW and PSM; dotted lines—for T1 and T2. The values of the material properties for these curves were taken from Ref. 18.

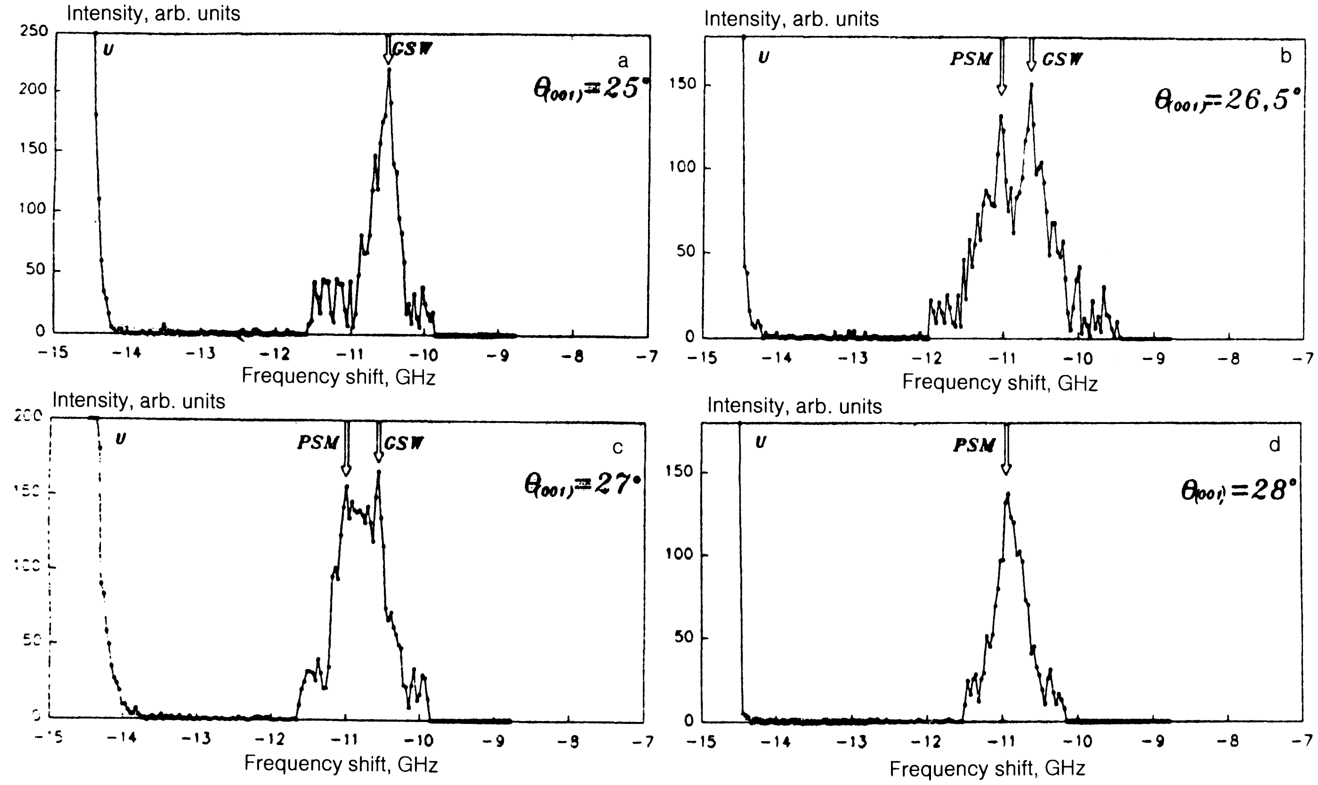

FIG. 3. Spectra of light scattered by a z-cut GaAs crystal ($\alpha = 70°$, $l = 488$ nm) a—$\theta_{(001)} = 25°$; b—26.5°; c—27°; d—28°. The arrows mark the positions of the Stokes Brillouin scattering satellites corresponding to GSW and PSM modes; U is an unshifted component of the spectrum in the neighboring order.

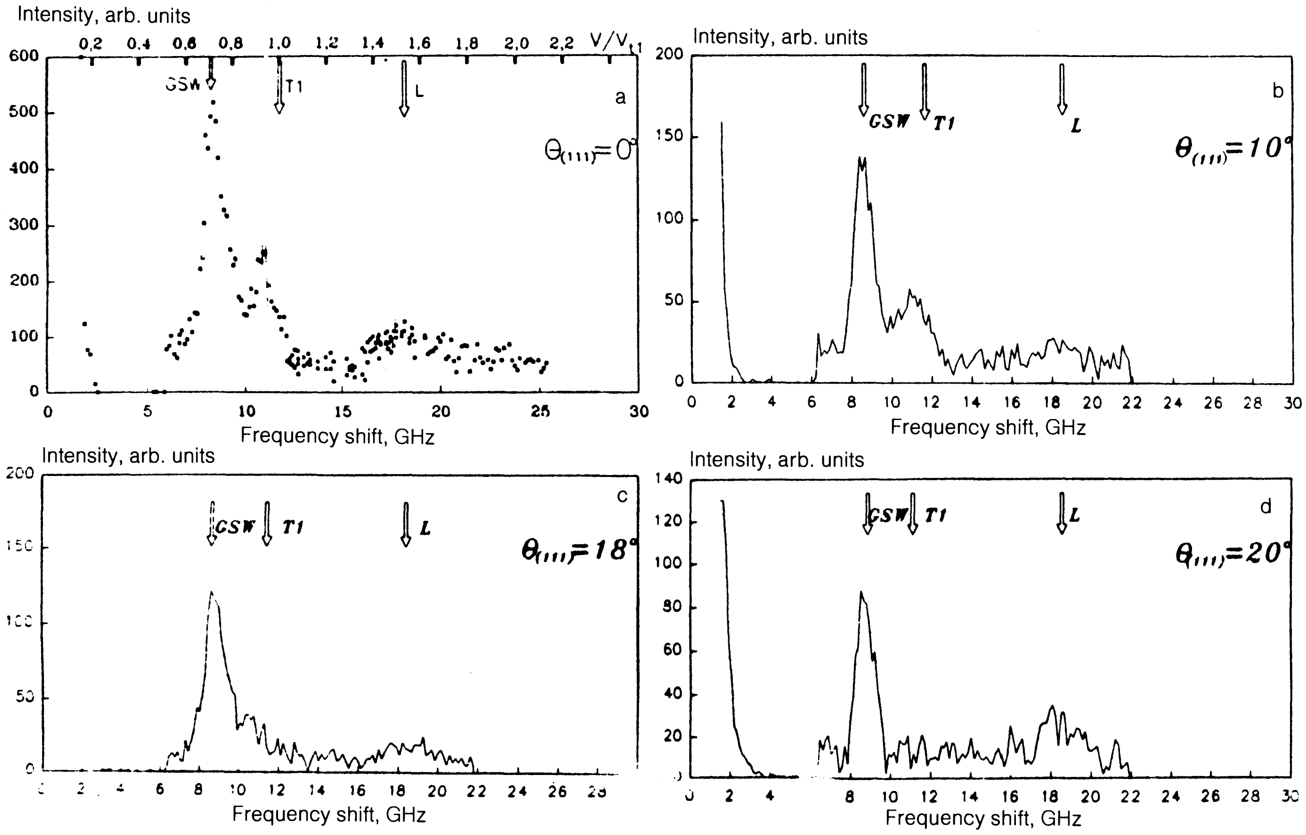

FIG. 4. Spectrum of the Brillouin scattering by a GaAs (111) crystal for various values of $\theta_{(111)}$. a—0°; b—10°; c—18°; d—20°. The frequency shift in frame a is also shown in normalized form, as V/V_{t1}. The theoretical (dotted) curve was reproduced from Ref. 11 $\alpha = 70°$, $i = 514.5$ nm).

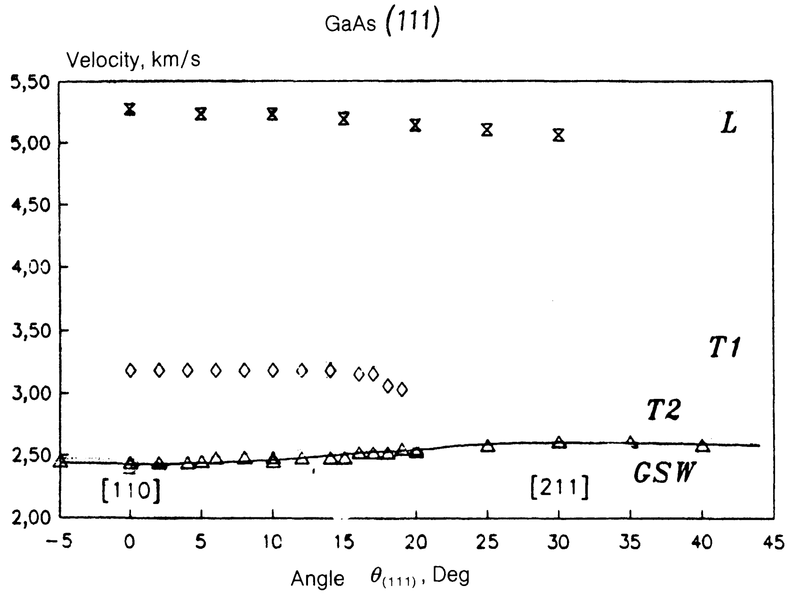

FIG. 5. Velocities of surface and bulk acoustic waves for the (111) plane of a GaAs crystal versus the angle $\theta_{(111)}$. The triangles, rhombi, and "hourglasses" are experimental velocities for GSW, PSM, and HFPSM modes, respectively. The theoretical GSW curve was taken from Ref. 19; the values of the material properties for the curves corresponding to V_{t1}, V_{t2}, and V_L were taken from Ref. 18.

the surface of the GaAs for $\theta_{(111)} = 0°$. The frequency shift of the Brillouin components in the spectrum is also shown in terms of the coordinate V/V_{t1}. In addition to the main Brillouin satellite at the frequency $f = 8.87$ GHz (this satellite is due to a GSW), we can clearly see two additional lines in this spectrum, at 11.6 and 18.7 GHz. These other lines correspond to pseudosurface modes.

The possibility of simultaneoulsy detecting components of the Brillouin scattering corresponding to both PSMs and HFPSMs in the case of a GaAs (111) crystal with $\theta_{(111)} = 0°$ was predicted in Ref. 11. The corresponding theoretical curve from that paper is reproduced in Fig. 4a (the dotted line). There is good qualitative agreement between the positions of the PSM and HFPSM components in the spectrum, on the one hand, and the maximum on the theoretical curve, on the other. The possibility of simultaneously detecting the main Brillouin scattering component and lines corresponding to PSMs in (111)-cut crystals with a large anisotropy parameter was also pointed out by M. Grimsditch.

Figure 4,b–d, reproduces the spectra of light scattered by a GaAs sample at $\theta_{(111)} = 10°$ (b), 18° (c), and 20° (d). We see that at $\theta_{(111)} = 10°$, as in the case $\theta_{(111)} = 0°$ (Fig. 4, a and b), the spectrum of the scattered light contains some peaks corresponding to both pseudosurface modes along with the GSW component.

With increasing $\theta_{(111)}$ (Fig. 4, c and d), the PSM line gradually fades away, merging with the high-frequency edge of the main Brillouin satellite at $\theta_{(111)} \approx 18°$. We observed components of the scattered light corresponding to GSWs and HFPSMs over the entire range of the angle $\theta_{(111)}$. In the interval $\theta_{(111)} \approx 12° - 22°$, the intensity of the main Brillouin satellite, associated with the GSW, decreased slightly. The PSM component in the spectrum was clearly observed over the interval $\theta_{(111)} = 0°–18°$.

Figure 5 shows the results of measurements of the velocities of the surface waves for various angles $\theta_{(111)}$. Also shown in this figure are corresponding curves for GSW, T1, T2, and L waves.

Over the entire range of $\theta_{(111)}$ the values found for V_{GSW} from the experimental data agree well with the corresponding theoretical curve, and the PSM and HFPSM velocities agree completely with the behavior of the curves for T1 and L waves, respectively.[10,11]

While preparing this paper for publication, we received a report of the simultaneous detection of both PSM and HFPSM pseudosurface modes in a GaAs (111) crystal.[17] The results we obtained in the present study are in good quantitative agreement with the theoretical and experiment results in Ref. 17.

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