PROC, 15TH INT. CONF. PHYSICS OF SEMICONDUCTORS, KYOTO, 1980
J. PHYS. SOC. JAPAN **49** (1980) SUPPL. A p. 711–714

SURFACE BRILLOUIN SCATTERING: COMPARISON OF RESONANT ELASTO-OPTIC AND RIPPLE MECHANISMS

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We report the relative strengths of the surface elasto-optic and surface ripple mechanisms of Brillouin scattering for the acoustoelectrically amplified bulk shear waves in CdS, as a function of acoustic frequency. The measurements were facilitated by strong resonant enhancement of the weak elasto-optic mechanism, which yielded the first resonant Brillouin spectrum on the opaque side of the absorption edge.

The Brillouin spectrum taken in a reflection geometry displays not only sharp lines associated with the conventional bulk, in-depth, interactions, but also a broad structure associated with light scattering interactions which are localized at the incident surface. The surface contribution is due to acoustic and optical boundary conditions. These are responsible for (1) introducing new modes (surface acoustic waves) and modifying the existing bulk modes through promotion of coupling of the transverse and longitudinal waves at the surface, and (2) generating localized light scattering mechanisms which effectively create amplitude and phase gratings at the surface. The localization of the interactions relaxes conservation of wave vector components perpendicular to the surface. The resulting broad spectrum serves to distinguish the surface Brillouin scattering contributions.

This paper presents the first quantitative determination of the relative strengths of the different surface scattering mechanisms. Furthermore, surface scattering is exploited to obtain the first resonant Brillouin scattering response on the opaque side of the absorption edge in semiconductors.

Two surface scattering mechanisms have been analyzed: the ripple, which involves dynamic corrugations generated on the surface by the atomic displacements associated with the bulk and surface acoustic waves, and the elasto-optic, which is based on phonon-induced modulation of the dielectric constant. The initial analyses of surface Brillouin scattering for bulk waves contemplated only the elasto-optic mechanism [1-3]. However, it was soon found that the ripple mechanism is usually overwhelmingly dominant [4-6]. Only very recently, has any evidence of surface elasto-optic contributions been reported [7-9].

The opacity of the material dictates which of the various contributions is dominant. The bulk contribution is sharply peaked and dominant in weakly absorbing material, then broadens with increasing opacity [10], and finally becomes indistinct in highly opaque metals [3]. On the other hand, the surface contribution is inherently broad and much less affected by opacity, eventually becoming dominant at high opacity. The relative changes are best illustrated in theoretical Brillouin spectra [3] for the elasto-optic mechanism. The ripple mechanism is relatively unaffected by opacity, except in so far as it affects the reflectivity [7,11], whereas the surface elasto-optic mechanism does diminish, but not until the absorption depth becomes smaller than the optical wave vector in the material [3]. These results explain the dominance of the ripple mechanism in highly opaque metals, and the possibility of competition between the two surface mechanisms in the less opaque semiconductors.

The specific objectives of the present work were (1) to isolate and detect the weak surface elasto-optic mechanism without interference from the ripple mechanism, and (2) to determine its strength relative to the ripple mechanism for the same bulk acoustic modes, as a function of acoustic frequency. The measurements were made with argon-ion laser radiation near the intrinsic absorption edge of CdS, both to take advantage of resonant response to enhance the elasto-optic mechanism and to obtain, as a possible by-product, the first resonant spectrum on the opaque side of the absorption edge. There is no resonant response for the ripple mechanism because its strength depends on reflectivity parameters [4,11] and not on the modulation of the dielectric constant.

To attain these objectives, it was necessary to find some means of observing the two surface scattering mechanisms completely separately, yet for the same phonons. This was accomplished for the bulk shear waves (T2) propagating in the basal plane of CdS, by taking advantage of differences in selection rules for the two mechanisms. For these waves, the atomic displacements are polarized parallel to the c-axis, and can be expected to generate ripples on the c-face but not on the orthogonal a-face. On the other hand, for the elasto-optic mechanism (p_{44} coefficent), the scattering cross section is expected to be zero on the c-face and maximum on the a-face [12]. Thus the two scattering mechanisms should appear completely separately, on orthogonal faces. Further discrimination is provided by analysis of the polarization of the scattered light. There should be no depolarization for light scattered by the ripple mechanism, but 90° rotation of polarization for the light scattered by the elasto-optic mechanism.

There remains the question of whether we can observe the two mechanisms for the T2 modes in isolation from other transverse and longitudinal bulk modes. For thermally-excited phonons, it is not necessarily possible to focus only on the T2 modes, because there can be overlapping contributions from the broadened spectra of the other phonon polarizations [3,5]. However, by resorting to the acousto-electric process, we were able to selectively and intensively amplify the piezo-electrically-active T2 waves from the thermal background [4]. This is accomplished by applying high voltage pulses (1.5 μsec long, repeated at 1 Hz) to CdS samples cut with their length in the basal plane. The amplified phonon pulses are so intense that the scattered light signals can easily be observed with only a single-pass Fabry-Perot interferometer. The scanning mode was employed for determination of the phonon frequency. The filter mode was used for quantitative measurements of the scattering intensity. Measurements were made at 300 K in the phonon frequency range 0.2 to 0.9 GHz in CdS with ~ 10 ohm-cm resistivity. All scattering was done in the reflection configuration, through a fixed angle of 90°, with polarized incident and scattered light.

The Fabry-Perot scans illustrated in Fig. 1, were taken in the opaque regime at λ = 4965 Å. The scan in Fig. 1(a) presents the ripple scattering on the c-face. Both the elastically scattered laser light and the very large phonon signal appear at the same polarization. In Fig. 1(b), the scan presents the much weaker elasto-optic signal on the a-face. It appears with its polarization rotated through 90° with respect to the laser signal, which is completely suppressed by an appropriate analyzer. The phonon lines are narrow here, in spite of the localization of the scattering mechanism, because the bulk phonons are intensively amplified in a tight cone (< 10°) along the axis of the sample. Thus there are no phonons available with a wide spread of wave vector components perpendicular to the surface, to contribute to the broadening of the signal.

Further confirmation that we have observed the two scattering mechanisms completely independently is obtained from the spectral response obtained with argon-ion laser lines between 4579 and 5145 Å. The scattered light intensity I_S was normalized to an attenuated, specularly reflected intensity I_R, with both signals measured after being passed through the Fabry-Perot interferometer to the photomultiplier. This normalization simultaneously corrects for laser intensity, instrumental response, reflectivity, etc. The results shown in Fig. (2) are for the

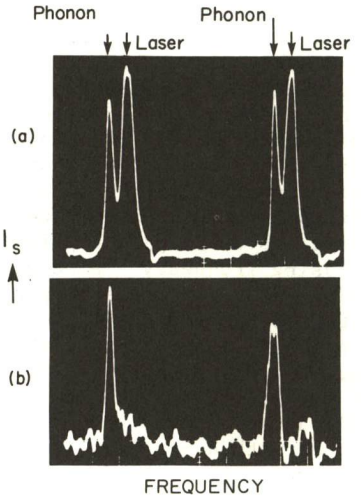

Fig. 1 Fabry-Perot scans for:
(a) the surface ripple mechanism on the c-face of CdS, and
(b) the surface elasto-optic mechanism on the a-face

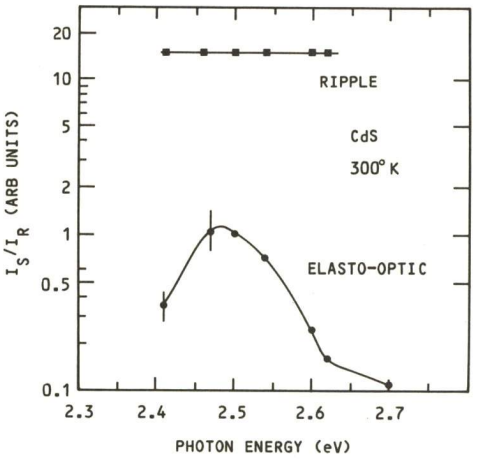

Fig. 2 Quantitative comparison of non-resonant surface ripple scattering and resonant surface elasto-optic scattering for shear waves in CdS at f = 524 MHz

ratio I_S/I_R in arbitrary units, but to scale for the two mechanisms. The data show a flat, non-resonant response for the ripple mechanism on the c-face, and a strongly resonant response for the elasto-optic mechanism on the a-face.

The resonant response shown here represents the first such determination on the opaque side of the absorption edge. Previous resonant spectra were restricted to transmission measurements on the transparent side of the edge, at $\lambda > 5145$ Å. They involved the bulk elasto-optic interaction with the same amplified shear modes as used here and the same p_{44} elasto-optic coefficient [14]. We note that the bulk shear waves have polarization parallel to the incident a-surface and should be uncoupled from other modes at the surface [3]. It would therefore be reasonable to assume that an extension of the surface interaction measurements to the transparent regime, would yield the same form for the resonant spectrum as obtained from the transmission measurements. However, it would require considerable enhancement of sensitivity to detect the surface elasto-optic scattering in the region of very rapidly decreasing strength on the long wavelength side of the resonant peak. This serves to illustrate the complementary nature of the bulk and surface Brillouin scattering techniques.

It is interesting to consider whether the shape of the resonant structure in Fig. 2 is influenced by the optical absorption. According to the theory for surface elasto-optic scattering, derived for an isotropic medium [3], the cross section varies with absorption coefficient α, as $[1+(\alpha\lambda/2\pi m)^2]^{-1}$. For CdS at 300 K, the refractive index n = 2.8 and α is nearly constant at $\sim 8 \times 10^4$ cm^{-1}, for $4500 < \lambda < 5000$ Å. In this range, the cross section is reduced by less than $\sim 5\%$, a negligible factor.

The relative strengths of the ripple and elasto-optic mechanisms are obtainable directly from Fig. 2, and refer to the acoustic frequency f = 524 MHz. Even at the peak elasto-optic response, the ripple scattering is stronger by a factor of 15. We estimate this factor to become ~ 700 in the nearly flat, non-resonant regime, which exists only at $\lambda > 7000$ Å for the T2 modes. The extrapolation from 15 to 700 includes the resonant contribution in the transparent regime for the bulk elasto-optic mechanism [13].

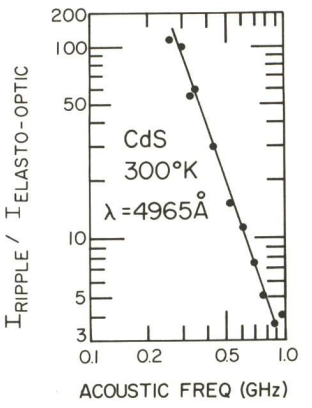

Fig. 3 Ratio of surface ripple to surface elasto-optic scattering strengths, as a function of acoustic frequency

The ratio of ripple to elasto-optic scattering strength is strongly dependent on acoustic frequency. This is illustrated in Fig. (3) by data taken at λ = 4965 Å. In the frequency range 0.2 to 0.9 GHz, the ratio of ripple to elasto-optic scattering varies as $1/f^3$. Thus the strong disparity between the two mechanisms may well disappear and even reverse at sufficiently high acoustic frequencies. Qualitatively, this result reflects mainly the strong decrease in ripple scattering, as the ripple amplitudes become smaller with increasing frequency. However, an analysis, using theories [3,5] for an isotropic material, yields a $1/f^2$ dependence for the ratio of the two mechanisms, for acoustic flux striking the surface at a small angle. The discrepancy may be due in part to the great sensitivity of the scattering strengths to the angle at which the phonons strike the surface, combined with a possible frequency dependence of the angular distribution of amplified phonon intensity.

We acknowledge the electronics assistance of A. Moretti. The work was supported by National Science Foundation Grant No. 7914618 and the National Science Foundation/Materials Research Laboratory Program No. DMR77-23798 at Purdue University.

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