Real Crystals, edited by T. J. Bastow (Australian Academy of Science, Melbourne, Australia, 1974), p. 217.
[5]D. E. Eastman and J. J. Donelon, Rev. Sci. Instrum. 41, 1648 (1970).
[6]U. Gradmann, Appl. Phys. 3, 161 (1974), and references cited therein.
[7]S. F. Alvarado, M. Erbudak, and P. Munz, Phys. Rev. B 14, 2740 (1976).
[8]To be correct one should use terms $J\langle \vec{S}_m \vec{S}_n \rangle$, where J is the exchange integral.
[9]S. F. Alvarado, W. Eib, F. Meier, D. T. Pierce, K. Sattler, H. C. Siegmann, and J. P. Remeika, Phys. Rev. Lett. 34, 319 (1975).
[10]G. K. Wertheim, H. J. Guggenheim, and S. Hüfner, Phys. Rev. Lett. 33, 1034 (1974); D. E. Eastman and J. L. Freeouf, Phys. Rev. Lett. 34, 395 (1975).

Surface-Ripple Mechanism for Brillouin Scattering of Reflected Light from Bulk Acoustic Waves[(a)]

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We demonstrate the efficiency of scattering of *reflected* light from shear bulk acoustic phonons, acoustoelectrically amplified in the frequency range of $0.2 \leq f \leq 2$ GHz in GaAs and CdS. Bulk-phonon–induced surface ripples, as opposed to fluctuations in the dielectric constant, are shown by several experiments to provide the dominant scattering mechanism.

The conventional mechanism for the Brillouin scattering of light involves phonon-induced fluctuations of the dielectric constant in the medium.[1] In this Letter, we shall present evidence for a competitive mechanism, the scattering of light by bulk phonon-induced mechanical perturbations or ripples on the surface of the solid. This mechanism is revealed by a study of the scattering of *reflected* light. Such a technique is particularly useful for phonon studies in opaque materials, where we found the ripple mechanism to be completely dominant, at least for low-frequency shear waves (<few gigahertz).

For light-scattering studies in opaque materials, it is first necessary to distinguish[2] between the scattering of *reflected light* and the conventional *backward scattering of the penetrating component* of the incident light. There are of course similarities: Both occur in the same optical configuration; when the absorption coefficient is very high,[3-6] both involve a very small interaction length which relaxes the requirement of wave-vector conservation for the components perpendicular to the surface, thereby broadening the Brillouin lines. The distinction between them comes primarily in their dependence on the reflectivity of the material.

After we show that we are dealing with scattering of reflected light, we will consider the extent to which mechanical ripples or dielectric fluctuations are responsible for spatially modulating the reflection of light and producing an effective surface grating. A major portion of our experiments were designed to distinguish between these two possibilities.

Source of phonons and their identification.—The difficult problem in Brillouin scattering experiments with opaque materials is to detect the very small fraction of inelastically scattered light. A recent solution was provided by Sandercock's development of the high-contrast, multipass Fabry-Perot interferometer.[3,7] However, we have taken a different approach. We exploited the technique of acoustoelectric amplification of phonons from the thermal background to enhance[8,9] greatly a select portion of the acoustic-phonon spectrum in the frequency range 0.2 to 1.5 GHz.[9] High-voltage pulses were applied to rectangular samples of GaAs and CdS at 300 K, to produce intense beams of shear phonons propagating in a narrow cone of $\sim 10°$ along the length of the sample,[8,9] in the [110] direction in GaAs or perpendicular to the c axis in CdS.[10] The amplified phonons form propagating domains ~1 mm wide, which produce a pulse-modulation signal[11] when the domain passes the spot of focused light. The signals and their angular dependence can be detected by a Si photodiode. A single-pass, Fabry-Perot interferometer was used to obtain the Brillouin components and thus determine the phonon frequency at any scattering angle.[2,11] All interferometer studies were made with a single-moded Ar^+ laser.

It was essential to discriminate between light scattering from the amplified bulk acoustic phonons and from surface acoustic phonons[12] which could be generated at surface irregularities by conversion from the bulk phonon domains.[11] Where bulk- and surface-phonon velocities are different, the distinctions could easily be made from group-velocity measurements of the domain and from phase-velocity determination from the phonon-frequency measurements. Otherwise, especially in CdS, it was necessary to quench selectively the surface-phonon domains by thin strips of Duco cement applied on the surface.

Reflected-light scattering studies were made on surfaces parallel to the phonon beam and also at an angle of 60° to the beam. In both cases the direction of scattering was governed purely by conservation of the wave-vector components parallel to the incident surface, $k_{\parallel}{}^{i}=k_{\parallel}{}^{s}\pm q_{\parallel}$; here, $\vec{k}^i$ and $\vec{k}^s$ refer to the wave vectors of the incident and scattered light, and $\vec{q}$ to the phonon wave vector. At any angle of incidence of light, signals were observed on either side of the reflected beam, corresponding to Stokes and anti-Stokes components. This type of scattering, providing direct proof of a very small interaction length, was applicable at any value of the absorption coefficient, and even for nearly complete transparency (1.06-μm radiation on GaAs). For such scattering, phonons of any frequency can contribute,[6] provided only that they have the appropriate component $q_{\parallel}$. Although this permits great broadening of the Brillouin line in the Fabry-Perot scan, we observed only instrument-limited lines (~ 50 MHz) because the beamlike quality of the amplified phonons limits the range of available $\vec{q}$'s.

Distinction between reflected-light scattering and backscattering.—Backscattering is maximized when reflectivity is suppressed, which occurs when light is incident at the Brewster angle. On the other hand, for reflected-light scattering, the scattered intensity is proportional to the reflectivity, specifically to its dependence on direction of polarization and the angle of the scattered light. This is demonstrated in Fig. 1, which shows data at $\lambda = 5145$ Å for the dependence on incident angle of $I_s{}^{\perp}/I_s{}^{\parallel}$, the ratio of scattered-light intensities for polarizations perpendicular and parallel to the plane of incidence. Experiments were made on (a) polished GaAs, (b) a thin opaque layer (~ 2–300 Å) of aluminum evaporated across a narrow section of the GaAs surface, and (c) CdS. The results are characteristic of the optical properties of the reflecting material. In each case, the data fit well with the solid curve which represents the intensity ratio $I_R{}^{\perp}/I_R{}^{\parallel}$, for the reflected light calculated at the scattering angle.

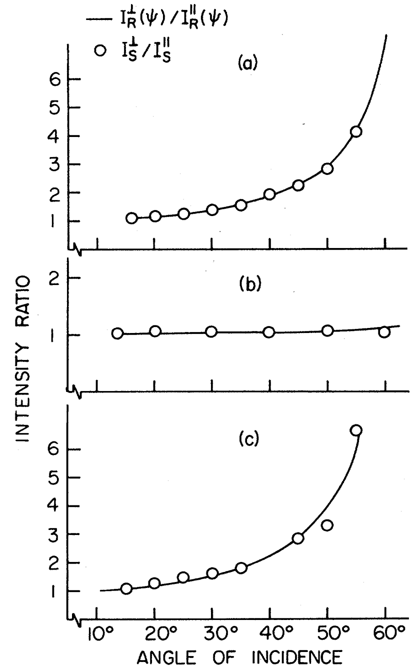

FIG. 1. Reflected-light scattering intensities on surfaces of (a) GaAs, (b) aluminized layer on GaAs, and (c) Cds for bulk acoustic waves. Ratio for perpendicular and parallel polarization eliminate various common angular factors and reveal calculated dependence on reflectivity shown by solid lines.

Evidence for a ripple mechanism.—The results obtained with aluminized surfaces on GaAs provide prime evidence for a ripple mechanism. On bare GaAs surfaces, the scattering efficiency was found to vary with the surface orientation. Yet, for each surface, the scattering efficiency on an aluminized segment was always proportional to that for the bare surface, in each case greater by the same factor of 2.3, which is the measured ratio of reflectivity of aluminum to GaAs at the angle of measurement. Furthermore, the Fabry-Perot scans on both surfaces showed the same phonon phase velocity, indicating that the aluminum film was being driven by the phonons in GaAs. These results can only be explained by a transmission of the rippling of the GaAs surface to the very thin aluminum layer. Any fluctuations of the dielectric constant on the GaAs surface could not have been transmitted through the aluminum in such a simple fashion.

Other evidence comes from analysis of the selection rules for scattering. For an elasto-optic mechanism and the geometry used for both GaAs and CdS, the polarization of the scattered light should have been rotated by 90° with respect to that of the incident light. Within experimental accuracy ($\lesssim 5\%$), we could see no contribution corresponding to rotation of polarization. This result is consistent with scattering of reflected light from a rippled surface.

A final test was the spectral dependence of the scattering efficiency on CdS. It is known that for the elasto-optic mechanism there is a strong resonant contribution to the scattering on the transparent side of the absorption edge,[13] at $\lambda > 5145$ Å at 300 K. A similar sharp resonance is to be expected[14] on the absorbing side for this mechanism. However, we found negligible variation of the scattering efficiency (I_s/I_R) for Ar^+ laser lines between 5145 and 4579 Å, a range in which any resonant contribution should have manifested itself strongly.

Analysis of scattering from ripples.—The theory of the scattering of reflected light from a periodic ripple of amplitude δ has been given by Lean[12] for a single-frequency surface acoustic wave. The peak intensity scattered in various orders is $I_s^m = I_R J_m^2(\alpha)$, with $\alpha = |(\Delta\vec{k})_n|\delta$, where $(\Delta\vec{k})_n$ is the normal component of the change in the photon wave vector. For $\alpha \ll 1$, the Bessel function $J_m(\alpha) = (\alpha/2)^m/(m!)$. We have tested the applicability of such a relation to the ripples formed by the bulk acoustic waves. To take into account the broadband character of the amplified acoustic flux, we insert a factor for the bandwidth of phonons that can be detected by the fixed aperture defining the collection optics. The frequency bandwidth decreases as $\cos\psi$ with increasing scattering angle ψ. Our aperture imposes no limitation on detecting the angular spread of phonon wave vectors.

Figure 2 shows the dependence of I_s on incident angle of light for phonons of frequency $f = 275$ MHz in GaAs. Both positive and negative orders are shown for both parallel and perpendicular polarizations of incident light. The measurements shown were made at the 5145 Å on the $(1\bar{1}0)$ face. The solid curves represent the modified theory. A good fit to the angular dependence is obtained for $m = 1$, indicating that only first-order scattering enters here. To fit both positive and negative orders, it was necessary to determine I_R at the *scattering* angle. Similar results were obtained at other phonon frequencies and on various surfaces of GaAs, both bare and aluminized, as well as on CdS.

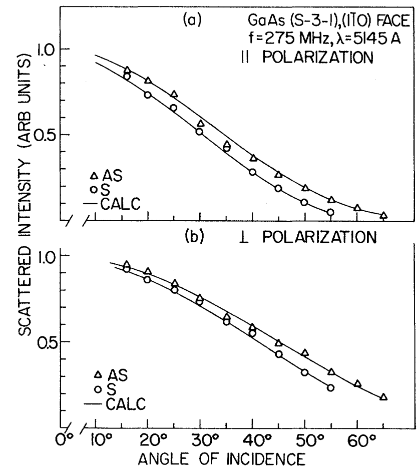

FIG. 2. Comparison of experimental data and theory for angular dependence of (+) and (−) first-order (Stokes and anti-Stokes) scattered-light intensities, for parallel and perpendicular polarizations.

Effective values of δ can be estimated from I_s/I_R. Obviously, this ratio depends on the bandwidth determined by the aperture, on the degree of phonon amplification, and on the scattering efficiency for each surface. We could readily detect values of $I_s/I_R \leqslant 10^{-7}$ for GaAs for a bandwidth of 75 MHz in the amplified phonon cone. This corresponds to $\delta \approx 0.1$ Å.

Although the dominance of the ripple mechanism of reflected-light scattering is demonstrated by every experiment described here, this dominance may not persist to very high acoustic frequencies. For a given acoustic energy density, which can be taken to be proportional to $(f\delta)^2$, we can expect the ripple amplitude to vary as $1/f$ and hence the scattering efficiency to fall off as $1/f^2$.

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[1]See, e.g., G. B. Benedek and K. Fritsch, Phys. Rev. 149, 647 (1966).

[2]S. Mishra, G. D. Holah, and Ralph Bray, in *Proceedings of the Third International Conference on Light Scattering, Campinas, Brazil, 1975*, edited by M. Balkanski, R. C. C. Leite, and S. P. Porto (Flammarion,

Paris, 1976), p. 198.

[3]J. R. Sandercock, Phys. Rev. Lett. 28, 237 (1972).

[4]J. G. Dil and E. M. Brody, Phys. Rev. B 14, 5218 (1976).

[5]A. S. Pine and G. Dresselhaus, in *Proceedings of the Third International Conference on Light Scattering, Campinas, Brazil, 1975*, edited by M. Balkanski, R. C. C. Leite, and S. P. Porto (Flammarion, Paris, 1976), p. 138.

[6]B. I. Bennett, A. A. Maradudin, and L. R. Swanson, Ann. Phys. (N.Y.) 71, 357 (1972).

[7]J. R. Sandercock, RCA Rev. 36, 89 (1975), and also in *Proceedings of the Second International Conference on Light Scattering*, edited by M. Balkanski (FLammarion, Paris, 1971), p. 9.

[8]D. L. Spears, Phys. Rev. B 2, 1931 (1970); Ralph Bray, IBM J. Res. Dev. 13, 487 (1969).

[9]T. E. Parker and Ralph Bray, Phys. Lett. 45A, 347 (1973).

[10]U. Gelbart and A. Many, Phys. Rev. B 7, 2713 (1973); M. Yamada, C. Hamaguchi, K. Matsumoto, and J. Nakai, Phys. Rev. B 7, 2682 (1973).

[11]S. Mishra and Ralph Bray, Appl. Phys. Lett. 30, 361 (1977).

[12]E. G. Lean, *Progress in Optics*, edited by E. Wolfe (North-Holland, Amsterdam, 1973), Vol. XI, p. 124.

[13]U. Gelbart and A. Many, Phys. Lett. 43A, 329 (1973); Y. Yamada, K. Ando, C. Hamaguchi, and J. Nakai, J. Phys. Soc. Jpn. 34, 1969 (1973); R. Berkowicz and D. H. R. Prince, Phys. Rev. B 11, 2316 (1975).

[14]Compare resonant behavior of Raman scattering on both sides of the absorption edge for LO phonons; R. H. Callender, S. S. Sussman, M. Selders, and R. K. Chang, Phys. Rev. B 7, 3788 (1973).

Supposed Failure of the Boltzmann Equation in Nb

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Serious doubt is cast on Allen's claim that the anomalous high-temperature electrical resistivity of Nb cannot be accounted for within the framework of the Boltzmann equation.

The lowest-order variational solution of the Boltzmann transport equation completely neglects the energy dependence and anisotropy of the electronic relaxation time, τ, and results in a constant-volume ideal electrical resistivity, $\rho(T)$, which is linear in temperature, T, for $T>\theta_D$ (the Debye temperature).[1] After correction for thermal expansion, this behavior is indeed observed for some metals, notably the alkalis,[2] but in most cases there is an appreciable deviation from linearity (d.f.l.), as in Nb, for example, where the measured resistivity[3] shows a pronounced negative deviation. In an attempt to explain this "anomalous" behavior, Allen[4] calculated the electrical resistivity of Nb taking into account two of the three mechanisms that had been proposed for d.f.l. He assumed that[5] $\tau(\epsilon)$ is inversely proportional to the electronic density of states, $N(\epsilon)$, and calculated the latter and the electronic velocity $v(\epsilon)$ from a Slater-Koster[6] fit to the augmented-plane-wave (APW) band structure computed by Mattheiss.[7] He then corrected the resulting resistivity in an ingenious way to account for the effect of shifts in the phonon spectrum with temperature. When the calculation failed to reproduce the observed behavior of $\rho(T)/\rho(\theta_D)$ for $T>\theta_D$ Allen concluded that a third mechanism, "saturation" of the electronic mean free path at high T leading to a breakdown of the Boltzmann equation, must be responsible for most of the d.f.l. observed in Nb. The purpose of this Letter is to point out that such a drastic conclusion is unwarranted, in view of the large number of uncertainties contained in the calculation. Indeed, had Allen's calculation agreed with experiment, this might have been taken as an indication that the approximations made in the calculations were reasonable and that the band structure used was sufficiently accurate for the problem, or alternatively, that a fortuitous cancellation of errors had occurred. However, lack of agreement proves nothing about the validity of the Boltzmann equation, because of the general uncertainties in the calculation. These we would like to discuss under four headings.

(1) *Reliability of the band-structure calculation.* —Allen derived his electronic parameters from the band structure calculated by Mattheiss,[7] who employed the full Slater-exchange approximation corresponding to $\alpha=1$ in the $X\alpha$ method. The work of Kohn and Sham[8] suggests that a more appropriate value of α might be $\frac{2}{3}$ and in recent times various values of α between 0.6 and 1 have been employed in band-structure calculations.