be significant for our understanding of the reverse isotope effect on the superconducting transition temperature T_c of PdH(D). This effect has been traditionally ignored[15] and is presently being considered for a quantitative understanding of the reverse isotope effect.

This work was supported by the U. S. Department of Energy.

---

[1] R. J. Miller and C. B. Satterthwaite, Phys. Rev. Lett. 34, 144 (1975).

[2] B. N. Ganguly, Z. Phys. 265, 433 (1973); P. Hertel, Z. Phys. 268, 111 (1974); H. Rietschel, Z. Phys. B 22, 133 (1975); B. N. Ganguly, Phys. Rev. B 14, 3848 (1976); J. P. Burger and D. S. McLachlan, J. Phys. (Paris) 37, 81 (1976).

[3] J. E. Schirber and B. Morosin, Phys. Rev. B 12, 117 (1975).

[4] J. M. E. Harper, Phys. Lett. 47A, 69 (1974).

[5] C. L. Wiley and F. Y. Fradin, to be published.

[6] J. Korringa, Physica (Utrecht) 16, 601 (1975).

[7] Y. Obata, J. Phys. Soc. Jpn. 19, 2348 (1964).

[8] P. Jena, Phys. Rev. Lett. 36, 418 (1976).

[9] M. Gupta and A. J. Freeman, Bull. Am. Phys. Soc. 22, 263 (1977), and to be published.

[10] D. A. Papaconstantopolous and B. M. Klein, Phys. Rev. Lett. 35, 110 (1975).

[11] E. Clementi and C. Roetti, At. Data Nucl. Data 14, 177 (1974).

[12] M. Manninen, R. Nieminen, P. Hautojärvi, and J. Arponen, Phys. Rev. B 12, 4012 (1975); P. Jena, A. K. Gupta, and K. S. Singwi, Solid State Commun. 21, 293 (1977).

[13] D. Ellis, private communication.

[14] P. Jena, K. S. Singwi, and R. Nieminen, Phys. Rev. B 17, 301 (1978).

[15] B. M. Klein, E. N. Economou, and D. A. Papaconstantopolous, Phys. Rev. Lett. 39, 574 (1977).

Theory of Surface-Ripple Brillouin Scattering by Solids

Rodney Loudon
Physics Department, Essex University, Colchester CO4 3SQ, England

(Received 23 December 1977)

The spectrum of light Brillouin scattered from thermally excited surface ripples on an elastically isotropic solid is calculated. The theory shows good agreement with recently measured Brillouin spectra of polycrystalline aluminum and nickel.

Mishra and Bray[1] have recently shown that the dominant Brillouin scattering mechanism for acoustoelectrically amplified transverse bulk acoustic waves in GaAs and CdS is provided by reflection from surface ripples. This contrasts with the more conventional mechanism associated with acoustic modulation of the optical susceptibility of the medium via the elasto-optic effect.

The thermal-equilibrium Brillouin spectra of a series of opaque materials have also been measured very recently.[2] The spectra show a dominating peak at the frequency of the Rayleigh surface acoustic wave, accompanied by shoulders which extend to higher frequencies where bulk transverse acoustic waves are allowed. The purpose of the present Letter is to show that the detailed spectral shapes of the thermal-equilibrium Brillouin spectra are well accounted for by the surface-ripple mechanism and provide strong evidence for its dominance.

Consider an elastically isotropic sample in the space $z<0$ with a flat surface in the plane $z=0$. The total surface area $\bar{A}$ and the illuminated area A used in the scattering experiment are both assumed to have dimensions very large compared to any acoustic wavelengths involved. Let the incident and scattered light beams in the Stokes component have frequencies ω_I and $\omega_I-\omega$, and lie in the zx plane making angles θ_I and θ_S with the z axis. The wave-vector transfer Q^x parallel to the sample surface is given by

$$cQ^x=\omega_I \sin\theta_I+(\omega_I-\omega)\sin\theta_S, \tag{1}$$

where both angles are taken positive when measured in the $z\bar{x}$ quadrant of the zx plane. Since $\omega \ll \omega_I$, Q^x is nearly constant across a measured spectrum.

The existing theory of light scattering in reflection by a surface ripple[3,4] can be extended without difficulty to obtain the thermal-equilibrium Brillouin cross section in the form

$$d^2\sigma/d\Omega d\omega_S = (A\bar{A}\omega_I{}^4/16\pi^2c^4)F^2\langle |u^z(0)|^2\rangle_{Q^x,\,\omega}, \tag{2}$$

where F is a function of θ_I, θ_S, and the relative permittivity of the scattering medium.[3,4] It is of order unity for the opaque materials considered here but its detailed form is not important for the present calculation. The final factor represents the power spectrum of the surface displacements parallel to the z axis with wave-vector component Q^x; it is this term which determines the Brillouin spectrum.

The acoustic waves which contribute to the scattering have wave vectors in the zx plane with x components of magnitude Q^x given by (1). There are in general three distinct acoustic waves of the required type for each frequency ω, one longitudinal and one transverse wave polarized in the zx plane, and a second transverse wave polarized parallel to the y axis. The transverse waves have a common velocity v_T related to the longitudinal velocity v_L by

$$v_T^2/v_L^2=(1-2\sigma)/2(1-\sigma), \tag{3}$$

where Poisson's ratio σ characterizes the elastic properties of the isotropic medium. The y-polarized transverse waves do not cause any surface ripple and are of no concern to us here.

The power spectrum needed for (2) is most conveniently obtained by linear response theory using the Green's function for acoustic displacement components $u^z(z)$ parallel to the z axis. This can be obtained from the closely related displacement-gradient Green's function used in the corresponding elasto-optic contribution to the cross section[5] or from similar Green's functions used to calculate other surface properties.[6] The result is

$$\begin{aligned}\langle\langle u^z(z); u^z(z')^*\rangle\rangle_{Q^x,\,\omega} = (i/2\rho\bar{A}\omega^2)\{&q_L^z \exp(iq_L^z|z-z'|)+(Q^{x2}/q_T^z)\exp(iq_T^z|z-z'|)\\ &-q_L^z[\Gamma_{LL}\exp(-iq_L^z z')-(v_L\Gamma_{LT}Q^x/v_Tq_T^z)\exp(-iq_T^z z')]\exp(-iq_L^z z)\\ &-Q^x[(v_T\Gamma_{TL}/v_L)\exp(-iq_L^z z')-(\Gamma_{TT}Q^x/q_T^z)\exp(-iq_T^z z')]\exp(-iq_T^z z)\},\end{aligned} \tag{4}$$

where ρ is the density,

$$q_L^z=\{(\omega/v_L)^2-Q^{x2}\}^{1/2}, \tag{5}$$

and q_T^z is the same expression with v_T replacing v_L. The Γ are surface reflection coefficients for acoustic waves: Let u_L^0 and u_T^0 be the amplitudes of longitudinal and transverse waves polarized in the zx plane with wave vectors $(Q^x, 0, q_L^z)$ and $(Q^x, 0, q_T^z)$. The amplitudes of the resulting reflected waves with wave vectors $(Q^x, 0, -q_L^z)$ and $(Q^x, 0, -q_T^z)$ are given by[5,7]

$$\begin{aligned}u_L&=\Gamma_{LL}u_L^0+\Gamma_{LT}u_T^0,\\ u_T&=\Gamma_{TL}u_L^0+\Gamma_{TT}u_T^0.\end{aligned} \tag{6}$$

The surface-displacement power spectrum is obtained by use of the fluctuation-dissipation theorem:

$$\langle|u^z(0)|^2\rangle_{Q^x,\,\omega}=(\hbar/\pi)\{n(\omega)+1\}\,\mathrm{Im}\langle\langle u^z(0); u^z(0)^*\rangle\rangle_{Q^x,\,\omega}, \tag{7}$$

where $n(\omega)$ is the Bose-Einstein thermal factor. Thus using the explicit forms of the Γ coefficients[5,7] and taking the usual high-temperature limit $k_BT \gg \hbar\omega$, we find

$$\langle|u^z(0)|^2\rangle_{Q^x,\,\omega}=\frac{k_BT}{\pi\rho\bar{A}v_T^3Q^{x2}}\,\mathrm{Re}\,\frac{\omega v_T^3Q^{x2}q_L^z}{4v_T^4Q^{x2}q_L^zq_T^z+(\omega^2-2v_T^2Q^{x2})^2}. \tag{8}$$

The shape of the spectrum in (2) for a constant or nearly constant wave-vector transfer Q^x is entirely determined by the dimensionless real factor in (8).

Three frequency ranges can be distinguished: *Range 1* corresponding to $\omega/Q^x<v_T<v_L$ has q_L^z and q_T^z purely imaginary. The real factor in (8) is zero except at the frequency ω_R of a pole corresponding to the Rayleigh surface acoustic wave.[7] The Rayleigh wave velocity is ω_R/Q^x. *Range 2* corresponding to $v_T<\omega/Q^x<v_L$ has q_L^z imaginary and q_T^z real. The real factor in (8) is zero at the two ends of the range but is otherwise nonvanishing. *Range 3* corresponding to $v_T<v_L<\omega/Q^x$ has both q_L^z and q_T^z real and the factor in (8) is nonzero except at the bottom end of the range.

Figure 1 shows the form of predicted Brillouin spectrum, the quantity plotted being the dimensionless real factor in (8). Poisson's ratio is taken to be $\sigma=\frac{1}{3}$ and (3) then gives $v_L=2v_T$. The Rayleigh peak occurs close to $\omega_R/v_TQ^x=0.933$ for this value of σ. The theory given here does not provide a shape or width for the Rayleigh line

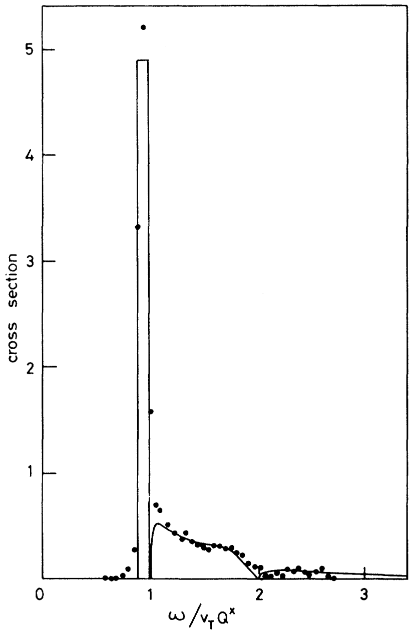

FIG. 1. The curve shows the theoretical surface-ripple Brillouin spectrum for an isotropic solid with $\sigma = \frac{1}{3}$. The experimental points from Ref. 2 are for polycrystalline Al.

and its integrated intensity is represented by the area of the rectangle in the figure. The experimental points are those of Sandercock[2] for polycrystalline Al, their vertical scale in the figure being the only adjustable parameter. A fit of similar closeness is obtained for the Brillouin spectrum of polycrystalline Ni, both metals having their Poisson's ratio close to the value assumed here.

The excellent agreement between theory and experiment provides strong evidence for the dominance of the surface-ripple mechanism. The spectra predicted by the elasto-optic mechanism[5,8] generally have a peak rather than a zero at the frequency $\omega = v_L Q^x$ and do not fit the experiments on Al and Ni. Further evidence is obtained from an estimate of the relative magnitudes of the two contributions. The real and imaginary parts of the elasto-optic coefficients of Al and Ni have not been measured, but insertion of calculated values[9] for Al produces an elasto-optic cross section which is two orders of magnitude smaller than the surface-ripple cross section. A comprehensive theory would also include interference between the two mechanisms but these effects are also negligible for the metals considered here.

There are two main general reasons for the predominance of the surface-ripple mechanism in metals. The elasto-optic cross section[5,8] is proportional to incident and scattered transmission coefficients at the surface and is inversely proportional to the square of the optical absorption coefficient when this is large, leading to severe reductions in the size of this contribution. By contrast, the surface-ripple cross section is proportional to surface reflection coefficients of order unity.

A further distinction follows from the behavior of the thermally excited acoustic waves in the vicinity of the surface. The boundary conditions force the $z = 0$ surface to be a nodal plane for many components of the strain, the acoustic variable on which the coupling of the elasto-optic mechanism relies. The amplitudes of the strain oscillations remain small within the optical skin depth of the metal where the elasto-optic scattering takes place.[10,11] The same boundary conditions, however, tend to produce maximum amplitudes at the free surface for the oscillations in the displacements, thus further facilitating the effectiveness of the surface-ripple mechanism.

I thank M. G. Cottam for helpful suggestions and J. R. Sandercock for communicating and discussing his experimental results.

---

[1] S. Mishra and R. Bray, Phys. Rev. Lett. 39, 222 (1977).

[2] J. R. Sandercock, to be published.

[3] I. L. Fabelinskii, *Molecular Scattering of Light* (Plenum, New York, 1968).

[4] E. G. Lean, in *Progress in Optics*, edited by E. Wolf (North-Holland, Amsterdam, 1973), Vol. 11, p. 124.

[5] R. Loudon, to be published.

[6] A. A. Maradudin and D. L. Mills, Ann. Phys. (N.Y.) 100, 262 (1976).

[7] L. D. Landau and E. M. Lifshitz, *Theory of Elasticity* (Pergamon, Oxford, 1970).

[8] K. R. Subbaswamy and A. A. Maradudin, to be published.

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

[10] A. Dervisch and R. Loudon, J. Phys. C 9, L669 (1976).

[11] R. Loudon, to be published.