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EXPERIMENTAL OBSERVATION OF SURFACE AND INTERFACE MODES BY LIGHT SCATTERING

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Resume - Un aperçu est donné des expériences récentes par diffusion de la lumière sur des surfaces et des faces séparatrices. Il a pour but de comprendre la nature des modes phonons observés et les mécanismes par lesquels ils couplent à la lumière.

Abstract - A review is given of recent light scattering measurements at surfaces and interfaces with a view to understanding the nature of the observed phonon modes and the mechanisms by which they couple to light.

The study of surface and localised phonon modes in layered materials dates back to the investigations of Lord Rayleigh /1/ into the propagation of earthquakes. More recently the development of surface acoustic wave devices lead to a resurgence of interest in these excitations. Investigations have been made using ultrasonic techniques and recently atomic and electron scattering, but the most powerful technique presently available is that of inelastic light scattering. This latter technique became possible with the development of the high contrast tandem Fabry-Perot interferometer.

This paper summarises the properties of the different surface and localized phonon states and introduces the rather complex problem of deriving the cross-section for light scattered from such states. Results of scattering measurements from a variety of different substrates and substrate/film combinations are presented. Finally, the analog problem of scattering from spin waves in magnetic substrates and films is briefly discussed.

THE LIGHT SCATTERING EXPERIMENT

For some years light scattering has been used as a tool to study phonons in bulk material. The interaction between the phonon and light was described by Brillouin /2/ in terms of the fluctuation in the dielectric constant produced by the strain associated with the phonon. Since the development of the laser, Brillouin scattering has ripened into a useful and diversified technique.

Figure 1 illustrates the technique, where for simplicity a backscattering arrangement is shown.

A transparent sample has been assumed with the scattering volume contained within the bulk of the material. The incident light of wavevector \( k^1 \) and frequency \( \omega^1 \) is scattered by a phonon \( \mathbf{q} \), \( \mathbf{q} \) into the state \( k^s \), \( \omega^s \) such that

\[
\begin{align*}
  k^s - k^1 &= \pm q \\
  \omega^s - \omega^1 &= \pm \Omega
\end{align*}
\]

(1)
The + sign refers to absorption of the phonon, the - sign to emission. (Respectively the anti-Stokes and Stokes processes).

For backscattering in a transparent medium of refractive index \( n \) we have \( q = 2n \kappa_0 \) (\( \kappa_0 \) is the vacuum wavevector of the laser light). The scattered light contains components at the frequencies \( w^1 + \Omega \) corresponding to all excitations present having the above wavevector. A normal spectrum will therefore show sharp peaks shifted by frequencies \( \Omega_L, \Omega_T, \) and \( \Omega_{TA} \) due to scattering respectively by the longitudinal and transverse phonon modes.

The scattered light is analysed using a Fabry-Perot interferometer as illustrated in fig. 1b. While this instrument is adequate for the observation of scattering in transparent materials, it is quite inadequate for studying excitations near surfaces particularly in opaque materials. In this case the light scattered elastically from surface defects swamps the much weaker Brillouin components. This problem can be overcome by the use of the high contrast multipass interferometer /3/ - however even then a difficulty remains, namely that neighbouring orders of interference are not widely separated from one another with the result that Brillouin spectra measured in neighbouring orders overlap. This is particularly troublesome for spectra containing many features or broad features - typical of the spectra to be described from surfaces and interfaces. This problem has recently been overcome with the introduction of the tandem multipass interferometer /4, 5, 6/, an instrument which combines high resolution, high contrast and a strong suppression of neighbouring interference orders. A novel scanning stage common to both interferometers automatically ensures synchronisation and forms the basis of a compact and highly stable design.

An alternative scheme relying purely on the electronic coupling of two separate interferometers /7/ has also been demonstrated.

A typical spectrum taken using the tandem interferometer is shown in Figure 2. This spectrum /6/ measured in backscattering on the surface of Ge, shows the depolarised light scattered from transverse phonons and demonstrates two important aspects of light scattering near surfaces.

a) The peaks are seen to be considerably broadened. This broadening is not related
to the phonon lifetime, but arises due to the high optical absorption /8/ which limits the penetration of the light to within a few wavelengths from the surface. The wavevector conservation condition of equation 1 only applies when all dimensions of the scattering volume are large compared to the wavelengths involved. For high optical absorption the equality is only approximate – a range of phonon wavevectors \( \Delta q \sim \alpha \) (where \( \alpha \) is the absorption coefficient) centred around \( q = k^S - k^I \) all contribute to the scattering and so give rise to the observed broadening effect.

b) Although the linewidth is in good agreement with simple calculations the marked asymmetry observed cannot be explained in terms of optical absorption. It was shown by Pine and Dresselhaus /9/ (in an albeit incorrect calculation) that the asymmetry arises from the coherent reflection of the phonons at the surface.

The above two facts, namely that the presence of the surface (and interfaces) relaxes the wavevector conservation requirement and modifies the phonon states with respect to the bulk, are of fundamental importance to an understanding of the light scattering experiments.

We consider first the modification to the phonon modes.

**PHONON MODES IN THE PRESENCE OF SURFACES AND INTERFACES**

The solutions of the equations of motion for an elastic continuum are the well known longitudinal and transverse acoustic excitations. In the presence of a surface, which introduces a stress-free discontinuity, new excitations appear, namely the Rayleigh, Lamb and Love modes.

A good feeling for the nature of these modes can be obtained following the eloquent treatment of Auld /10/ using the very simple transverse resonance technique. It is convenient to first derive the modes of a plate. The results for the plate may then be carried over to the case of a semi-infinite sample by allowing the plate thickness to approach infinity.
The symmetry of the problem with respect to the plane through the middle of the plate \((z = 0)\) requires that the waves be either symmetric or antisymmetric with respect to this plane. We thus require at least two plane waves with wavevectors \(k\) and \(\bar{k}\), where \(\bar{k}\) is obtained by reflecting \(k\) on the symmetry plane. The simplest case is that of shear horizontal waves (Fig. 3). In this case, the polarization remains horizontal upon reflection and only two waves, \(k\) and \(\bar{k}\), are required to fulfill the boundary conditions (no force in the \(z\) direction). This boundary condition requires that the displacement amplitude vanishes at \(z = \pm b/2\), i.e.,

\[
\frac{\omega}{u_t} \sin \theta = k_{zt} = \frac{m \pi}{b}, \quad m = 1, 2, 3, \ldots
\]

where the angle \(\theta\) is defined in Fig. 3. Equation 2 has a continuum of solutions with a lower cutoff for \(\omega = \frac{u_t \pi}{b}\).

The situation is more complicated whenever longitudinal waves are involved upon reflection on the plate boundary; these waves are partly converted into shear vertical waves (SV) and four plane waves (L with \(k_L\) and \(\bar{k}_L\) and SV with \(k_T\) and \(\bar{k}_T\)) are required to fulfill these boundary conditions leading to

\[
\frac{\tan(k_{zt}b/2)}{\tan(k_Lb/2)} = \frac{4B^2k_{zt}k_L}{(k_{zt}^2 - k_L^2)^2} \quad \text{(symmetric)}
\]

and

\[
\frac{\tan(k_{zt}b/2)}{\tan(k_Lb/2)} = -\frac{(k_{zt}^2 - k_L^2)^2}{4B^2k_{zt}k_L} \quad \text{(antisymmetric)}.
\]

The mode frequencies \(\omega\) are given by

\[
k_{zt}^2 = \left(\frac{\omega}{u_t}\right)^2 - \beta^2
\]

\[
k_{L}^2 = \left(\frac{\omega}{u_t}\right)^2 - \beta^2
\]

There are three frequency regimes implicit in equations 4:
a) $\omega / \beta > U_t$. In this case, both $k_{z1}$ and $k_{zt}$ are real and the solutions are normal bulk modes.

b) $U_1 > \omega / \beta > U_t$. Here, $k_{zt}$ is real but $k_{z1}$ is purely imaginary.

The solutions represent bulk transverse modes combined with a longitudinal component which is strongly localised at the surface. These modes are referred to as Lamb waves.

c) $\omega / \beta < U_t$. In this case, both transverse and longitudinal components are localised at the surfaces.

These modes are related to the Rayleigh surface mode of a semi-infinite solid. In fact, the sum of the lowest order symmetric and antisymmetric solutions in the limit $\beta b \to \infty$ is just the Rayleigh solution.

In the limit $\beta b \to \infty$, the discrete Lamb and shear horizontal (SH) solutions form continua in which the SH modes are indistinguishable from normal bulk modes. The Lamb modes, on the other hand, are bulk transverse modes with an additional longitudinal component localised at the surface.

The Rayleigh mode and the surface component of the Lamb mode are localised typically within one wavelength from the surface.

The first observation [11] of these plate modes was reported for the rather trivial case of near-normal incidence scattering from thin films of collodion and from thin crystalline platelets. For this case $\beta \to 0$ with the phonons propagating backward and forward across the plate. The Lamb solution separates into a Love wave and a longitudinal wave with $k_{z1} = m\pi / b$. The restriction of the scattering volume in the $z$ direction leads to a wavevector uncertainty $\Delta q \sim 1 / b$ with the result that typically 3 of the longitudinal modes could be observed.

A more complex example is in the spectrum of GaAs [12] shown in figure 4. Here we see the bulk phonon peaks, absorption broadened as in the case of Ge discussed above. In addition the Rayleigh surface phonon is clearly resolved together with a broad shoulder arising from scattering from the continuum of Lamb waves. The
A surprising feature of the spectrum however, is that the Rayleigh surface wave peak is more intense than the bulk phonon peak. Since the Rayleigh wave is localised within a wavelength from the surface, the interaction volume with the light is an order of magnitude smaller than for the bulk phonons. The origin of the strong Rayleigh wave scattering was explained by Mishra and Bray /13/ who pointed out that the phonons ripple the surface and that light may be directly scattered from these ripples. The total scattered intensity must be calculated therefore in terms of both, elastooptic and ripple scattering mechanisms allowing for the interference between the two. The calculations have been performed by several groups /12, 14-19/ and the theory found to be in excellent agreement with the measurements. Notice that the spectrum of figure 2 is depolarised, there being in this case no contribution from the ripple mechanism.

An important result of the calculation of the ripple scattering crosssection is that in general higher reflectivity materials will scatter more strongly. In particular, phonons in metals may be easily observed via this mechanism. In figure 5 are shown measurements on polycrystalline Al /20/. The Rayleigh wave and the Lamb continuum are clearly resolved and the inset shows the excellent agreement with theory /14/. Earlier measurements by Dil and Brody /21/ on liquid metals have also been satisfactorily explained in terms of the ripple mechanism /16, 22/.

A variety of measurements of thin films on substrates have been performed. The substrate, of course, modifies the properties of the plate modes. Before discussing these measurements we qualitatively summarise the properties of the modes of a supported plate.

MODES OF A PLATE ON A HALF-SPACE

Assuming the lower surface of the plate to be the interface with the half-space, the reflection coefficient for phonons at the lower plate surface is now no longer unity. This fact seriously complicates the analysis since the transmitted waves must be included. This section is restricted to a qualitative discussion of the modes with emphasis on their similarity to the modes of the unsupported plate.
Love Modes. The solutions of interest are those which are trapped within the slab, corresponding to total internal reflection with only an evanescent wave extending into the substrate. It can be shown that trapping can only occur for the SH waves if $U_t' > U_k$ where $U_t'$ is the transverse velocity in the substrate. The transverse resonance condition is not as simple as (2) due to the phase change occurring on reflection at the interface. The modes are referred to as Love waves.

Generalised Lamb Waves. These solutions with polarisation in the $xz$ plane also depend on the relationship between $U_t'$ and $U_k$.

a) $U_t' \ll U_k$. In this case, only one solution is possible which in the limit $b \rightarrow 0$ becomes the Rayleigh wave on the substrate.

b) $U_t' \gg U_k$. Here there is an infinite number of solutions. As before they are symmetric and antisymmetric solutions which are here labeled $M_{11}$ and $M_{21}$, respectively. Of special interest are the lowest order modes $M_{11}$ and $M_{21}$. In the limit $b \rightarrow 0$, only $M_{11}$ is trapped and this corresponds to the Rayleigh wave on the bare substrate. As $b$ increases from zero, $M_{21}$ is the next mode to be trapped. This is called the Sezawa mode. In the limit $b \rightarrow \infty$, the Sezawa mode becomes the Rayleigh mode on the top surface of the slab.

Under certain conditions for $v_t' = u_t$, the mode $M_{21}$ becomes a bound interface mode known as the Stoneley wave. The Stoneley wave velocity $U_S$ must satisfy the condition $U_R' < U_S < U_t'$, where $U_R'$ is the Rayleigh wave velocity on the bare substrate.

Following are some examples of light scattering from supported films. In general a calculation of the light scattering cross-section becomes complicated - in the most general case of a transparent film on a transparent substrate there are contributions to the scattering from both surface and interface ripples and from the elastooptic effect in both film and substrate. In addition, limitations on the scattering volume imposed by film thickness and/or optical absorption modulate the scattering cross-section as discussed above.

An example of the simplest situation in which only the surface ripple is important is shown in figure 6 for scattering from a film of Al on Si. This spectrum should be compared with that of pure Al in figure 5. Notice that the continuum of Lamb waves observed in figure 5 becomes discrete modes for the thin film. The continuous line of figure 6 is the calculated cross-section /23/. Vacher and co-workers have reported measurements of the Rayleigh wave only on Al films on different substrates /24/.

Measurements of SiO$_2$ on Si could be theoretically explained in detail /25/ using the known elastooptic and elastic properties of film and substrate. On the other hand, interpretation of spectra of Au films on Si (see subsequent paper in these proceedings) has yielded the previously unknown elastooptic constants of Au.

Of interest are measurements by Dil and coworkers /7/ on polycarbonate films on glass in which Love waves were successfully observed. Both these measurements and measurements on a Hg/glass sample which show a bound interface mode similar to the Stoneley mode are described in these proceedings.

As expected for low reflectivity materials the spectra from polycarbonate/glass samples are found to be very weak. Rowell and Stegeman have demonstrated that prism coupling techniques may be used to couple the light more effectively and obtained strong spectra of Rayleigh and Sezawa modes in optical waveguide structures.
LIGHT SCATTERING FROM MAGNETIC MATERIALS

Spinwaves in magnetic materials modulate the dielectric constant of the medium (via the spin orbit interaction) and so can scatter light. Early measurements on YIG demonstrated that the scattered intensity is comparable to that from phonons, and further that a lot of information relating to the magnetic system may be derived from the spectra. Several measurements on relatively transparent materials have been discussed in a short review by Borovik-Romanov and Kreines /26/.

Two differences between phonon spectra and spinwave spectra are of particular interest:

a) Even in the region of wavevectors accessible to light scattering (less than 1% of the Brillouin zone) spinwaves show strong dispersion due to the exchange interaction. Measurements as a function of wavevector thus yield useful information — by comparison phonons show no dispersion in this range.

b) The lack of time reversal symmetry between Stokes and anti-Stokes scattering events leads to marked asymmetry in the observed spectra.

The early measurements on relatively transparent materials showed only scattering from bulk spinwaves. More recently, measurements on opaque ferromagnets have been reported in which surface features have been observed. The surface spinwave, first described by Damon and Eshbach /27/ has the strange property of non-reciprocal propagation. More precisely on a surface with magnetisation in the plane of the surface a surface spinwave may only propagate from left to right across the field direction. As a result in a light scattering measurement (which involves absorption of an excitation in one direction and emission in the opposite direction) the surface spin wave peak will be observed only in the Stokes or anti-Stokes spectrum depending on the field direction. This strange asymmetry was first observed by Grunberg and
Metawe /28/ in EuO. Similar observations have been reported in Fe and Ni /29/.

Figure 7 shows a spectrum from Fe and demonstrates not only the marked asymmetry but also a strong broadening in the bulk spinwave peak arising from the high optical absorption. Cottam /30/ has calculated the spectral line shape for the bulk modes as a function of optical absorption, exchange constant and degree of pinning of the spins. Camley and Mills /31/ have performed a numerical calculation including scattering from the surface spinwave and obtain excellent agreement with the experimental data.

There has recently been considerable interest in scattering from thin magnetic films both theoretically /32-34/ and experimentally /35, 36/. Due to lack of space the reader is referred to the literature.

CONCLUSIONS

Brillouin scattering has ripened into a powerful tool for the study of small wave-vector excitations, both phonons and spinwaves, at surfaces and interfaces on materials ranging from transparent dielectrics to highly opaque metals.

REFERENCES