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BRILLOUIN SCATTERING NEAR RIGID INTERFACES

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Abstract - A review is given on Brillouin-scattering measurements taken in the vicinity of liquid-solid interfaces with a variety of ratios of acoustic impedances. For this purpose we have taken ethanol, polycarbonate and the liquid metals mercury and gallium in close contact with solid glass substrates. Some refined measurements on Lamb and Love modes in thin polycarbonate films on Pyrex glass are reported. A Stoneley-type wave at the liquid metal-glass interface is analysed in some detail. Our improved double-pass tandem Fabry-Pérot interferometer is described briefly.

1. Introduction

In 1975 Dresselhaus and Pine showed that the thermal phonons reflected from a stress-free material surface significantly influence the Brillouin spectra taken in the vicinity of such a surface. The implementation of the acoustic boundary conditions in the derivation of the theoretical Brillouin spectra taken near a stress-free interface removed an extra doublet from the spectra that appeared in the original derivations.

In 1980 Dil and van Hijningen showed that this extra doublet does appear in the Brillouin spectrum when it is taken near a rigid interface. Such a scattering geometry is realized, for example, near a liquid-solid interface. The thermal phonons (or rather density fluctuations) in the liquid are then clamped by the relatively rigid surface of the solid.

In this paper we review the situations where different acoustic loads are put on solid glass substrates, and where the effects of these loads are probed by the Brillouin scattering technique. We begin by analyzing the Brillouin spectra of ethanol "loading" a glass surface. Then we increase the load by changing to polycarbonate (PC). We show that both the longitudinal and transverse phonons in PC are clamped at the glass-polymer interface. The difference in acoustic impedances between PC and glass makes it possible that a thin film of the polymer on glass will support thermal Lamb and Love waves. These waves are observed as peaks in
Finally, we increase the impedance on the glass even further by loading it with the liquid metals mercury and gallium. The interface wave between the glass and liquid metals is of the Stoneley type and is also observed in the Brillouin spectra.

2. Experimental setup

In order to perform Brillouin-scattering measurements in the vicinity of liquid-solid interfaces, a Brillouin spectrometer with a tandem Fabry-Pérot interferometer is needed. Such a tandem arrangement eliminates the problem of overlapping orders, so that the larger Brillouin shifts which originate from the solid do not fold back into the first order and confuse the spectra with the smaller shifts from the liquid.

Our spectrometer with the tandem interferometer was described in Ref. 8. We have introduced some modifications to increase the throughput of the instrument. This increase was accomplished by changing from triple to double passing, so that the aperture of the signal beam could be increased to 25 mm. A simplified layout of the modified spectrometer is given in Fig. 1. Note the cats-eye retroreflectors used for the multipassing. These reflectors are more convenient for routing such large-diameter beams than cube-corner prisms.

The contrast of the double-pass tandem interferometer is of the order of $10^{10}$, and its finesse normalized to the first order is about 100. All experiments reported in this paper were performed with the mirror spacing ratio set to $d/d'=1.1$ with $d=8.2$ mm. This results in an FWHM of the instrumental profile of
\[ \frac{\Delta \Omega}{2\pi} = 0.2 \text{ GHz}. \]
Finally, the transmission of the spectrometer from the sample to the photomultiplier is approximately 10%.

Fig. 1 also shows the sample cell. It consists of a hemisphere made of glass with a sample chamber glued at the back of it for holding the liquid. By properly choosing the index of refraction of the glass, one is able to generate evanescent waves along the glass-liquid interface. The penetration depth \( \delta \) of these evanescent waves into the liquid can be varied by adjusting the angle \( \theta_0 \) which the incident laser beam makes with the glass-liquid interface:

\[ \delta = \left( n_g^2 \cos^2 (\theta_0) - n_l^2 \right)^{-\frac{1}{2}} \lambda_0 / 2\pi. \]  

In Eq. (1), \( \lambda_0 = 514.5 \) nm is the free-space wavelength of the argon-ion laser, and \( n_g \) and \( n_l \) are the indices of refraction of the glass and the liquid, respectively. For the experiments reported here, we have used fused silica (\( n_g = 1.46 \)), pyrex (\( n_g = 1.47 \)) and GGG (gadolinium-gallium garnet, \( n_g = 1.98 \)) as our solid glass substrates. The samples of interest were ethanol (\( n_l = 1.36 \)), mercury (\( n_l = 1.4 + i 4.4 \)), gallium (\( n_l = 0.89 + i 5.6 \)) and polycarbonate (\( n_l = 1.59 \)). The smallest penetration depth was achieved at the GGG-ethanol interface, where \( \delta \) could be made as small as 60 nm.

3. Brillouin scattering near rigid interfaces

In 1980 we showed that the Brillouin spectra taken in the vicinity of a solid-liquid interface and due to the evanescent incident field inside the liquid contains two doublets. One doublet is just a broadened version of the usual Brillouin spectrum of the bulk liquid. Its frequency is given by

\[ \pm \Omega_B = (q^2 + K^2 - A^2)^{\frac{1}{2}} \nu, \]  

where

\[ q_\pi = k_{s,\pi} - k_{o,\pi}, \]  

\[ K = \text{Re}(k_{s,z} - k_{o,z}), \]  

\[ A = \text{Im}(k_{s,z} - k_{o,z}), \]

and \( \nu \) denotes the velocity of sound of the liquid. In Eqs. (2)-(5), \( k_0 \) and \( k_s \) are the complex wavevectors of the incident and scattered light inside the scattering medium, and \( q \) is the wavevector of the scattering phonon. The subscripts \( \pi \) and \( z \) denote vector components parallel and perpendicular to the interface. Later we introduce the superscript \( > \), which indicates components of the wavevectors outside the scattering medium.

The doublet of Eq. (2) appears also in the Brillouin spectra taken with evanescent waves along a stress-free interface. This is, for example, the case in backscattering experiments on free semiconducting or other opaque materials. The FWHM of these peaks is inversely proportional to the penetration depth of the incident light and takes the form

\[ \Delta \Omega_B = 2 (2AK)^{\frac{1}{2}} \nu. \]
However, in the case of scattering near a rigid interface, an extra Brillouin doublet appears, the frequency shift of which is given by

\[ \pm \Omega_\pi = \Omega_\pi \nu . \]  

The FWHM of these peaks is proportional to the damping coefficient \( \Gamma \) of the phonons, and is given by

\[ \Delta \Omega_\pi = \Gamma q_\pi^2 . \] 

Hence, we have here the peculiar phenomenon of a doublet of narrow peaks from a very thin interaction region. The origin of these peaks stems from a singularity in the density of states, which is, in turn, due to the asymmetry introduced by the clamped interface at \( z = 0 \).

The evolution of the two doublets as a function of the penetration depth of the incident light is shown in Fig. 2. The scattering geometry is simply the liquid ethanol clamped by the solid glass \((n_g = 1.55)\) demi-sphere as shown in the inset.

Fig. 2. Observed and calculated (full curves) Brillouin spectra taken near an ethanol-glass interface (inset) for different penetration depths of the incident light.

One clearly observes the increasing shift \( \Omega = \cos(\theta_0) \) of the narrow peak with the decreasing angle \( \theta_0 \). The broad peak, however, remains centered at \( \Omega_b = \sqrt{2}n_1v_2\pi/\lambda_0 \), but it broadens as \( \Delta \Omega_b = (n_2^{\cos^2(\theta_0)} - n_1^2)^{1/4} \).

It should be noted that the directions of incidence and observation are reciprocal. Hence, when \( k_s \) and \( k_0 \) are interchanged in Fig. 2 but taken in opposite directions, the spectra remain the same which was experimentally verified. In that case the interaction region is made small as the direction of observation selects a complex \( k_s \).

A new situation arises when \( \Omega_s \) becomes larger than \( \Omega_b \). This happens when \( k_s \) turns imaginary as in a 180°-backscattering geometry. In that case, only one doublet centered at \( \pm \Omega_\pi \) remains. We verified this experimentally by taking spectra near a GGG-ethanol interface, where we were able to confine the interac-
tion region to 60 nm. The two spectra of Fig. 3 illustrate the two situations with $\Omega_b > \Omega$ and $\Omega_b < \Omega$ ($\theta_0 = 7^\circ$ for $\Omega_b > \Omega$ and $\theta_0 = 28^\circ$ for $\Omega_b < \Omega$, $n_0 = 1.98$, $n_1 = 1.36$ in Eq. (1)). Note that in the lower part of Fig. 3 a Brillouin spectrum is taken from ethanol with a $q$-vector which is $\Omega_b / \Omega_b = 1.5$ times larger than a corresponding $q$-vector in a bulk-scat-tering experiment.

Fig. 3. Brillouin spectra from an ethanol - GGG interface for the cases $\Omega_b > \Omega$ (upper) and $\Omega_b < \Omega$ (lower). The curves are smoothed data points.

Another interesting case arises when a polymer hardens against the rear surface of the GGG demi-sphere. We took polycarbonate as polymer, as it is known for its large elasto-optic coefficient of the shear waves. The spectra of Fig. 4 illustrate that for completely hardened PC, the longitudinal and transverse phonons are still clamped at the interface, because both spectra peak at $\Omega = q_V 1_s$ and not at $\Omega_b$. The clamping indicates a good contact between the PC and the glass.

The observed fact that the phonons experience the glass as a rigid substrate can be understood from the difference in acoustic impedances, $Z = \rho v$, $Z_{eth}$ $Z_{PC}$ $Z_{SiO_2}$ $Z_{Pyrex}$ $Z_{GGG}$ $Z_{Hg}$ $Z_{Ga}$ $1.95$ $2.9$ $13.0$ $13.1$ $48.2$ $20.1$ $16.2$ (10$^6$kg/m$^2$s$^3$)

The impedances of the liquid metals mercury and gallium are included in Eq. (9) for later reference. Eq. (9) learns that there exists still a relatively large impedance mismatch between PC and the glasses. This implies that a relatively thin film of PC on a glass substrate will support all kinds of propagating acoustic modes. These modes are called generalized Lamb waves when their polarization lies in a plane perpendicular to the film. The observation of such Lamb modes has been reported by us in Ref. 8.
Their appearance in the Brillouin spectra usually shows up as a rather strong peak due to the zero order Lamb wave, followed by a set of weaker peaks due to higher order modes. A theoretical study of these spectra will be given in a conference paper by Nizzoli et al. 13.

The thin PC films on glass support in addition to the Lamb waves a set of acoustic modes which are polarized parallel to the film. These modes are called Love waves12, and we reported the observation of the zero-order Love wave in Ref. 8. With our improved spectrometer we were now able to observe the higher order modes, as is shown in Fig. 5. The scattering geometry is shown in the inset.

The position of these peaks can be calculated from the proper dispersion relations given in Ref. 12. The lower part of Fig. 5 shows that the frequency shifts of the observed Love modes fall rightly on the dispersion curves.

Finally we report on some new scattering experiments taken near glass-liquid metal interfaces. The acoustic impedances of these liquids are comparable to the corresponding values of the glass demi-spheres, as is illustrated by Eq. 9. The Rayleigh surface waves on the stress-free glass surfaces are then renormalized by the acoustic load introduced by the liquid metal. Such a Stoneley-type wave was observed in the scattering experiments of Ref. 8 for a glass-liquid mercury interface. Figs. 6 and 7 show our new results on fused silica-liquid mercury and gallium interfaces. The scattering geometries are shown in the insets. The upper parts of the figures show examples of the
Fig. 5. Brillouin spectra of polycarbonate films on Pyrex substrates, showing Love-wave peaks (upper) and calculated dispersion curves of the Love waves (lower); the asterisk was taken from Ref. 8.

Brillouin spectra of these interface phonons, whereas the lower parts show the shifts as a function of $q_x$. The spectra were taken with an incident laser power of 25 mW, so that laser heating is estimated to be less than 20 K.

Fig. 6. Brillouin scattering from a glass-liquid mercury interface, showing the interface phonon of the Stoneley type (upper) and the theoretical and experimentally observed dispersion relations (lower); the asterisk is taken from Ref. 8.

The lower parts of Figs. 6 and 7 also show the theoretical curves for $\Omega$ vs. $q_x$. These curves can be calculated from the dispersion relations of Stoneley waves, such as derived by Albuquerque14. We conclude that our Stoneley-wave velocities $v_0 = \Omega /q_x$ are considerably smaller than the corresponding theoretical values. The reason for this is not clear. We have heated the samples up to 550 K but not seen any anomalous effect. The only interesting phenomenon was, that for both liquid mercury and gallium the width of the peaks remained at the constant value of
$\Delta \Omega = 0.9$ GHz with decreasing $q_f$. This may be an indication of some kind of relaxation $^{10}$.

Fig. 7. Brillouin scattering from a glass-liquid gallium interface, showing the interface phonon of the Stoneley type (upper) and the theoretical and experimentally observed dispersion relations (lower).

4. Conclusions

We have shown that interface effects due to different acoustic loads on solid glass substrates can be probed by the Brillouin-scattering technique. When the load is small, such as in the case of a low-density liquid, the Brillouin spectra show that the thermal phonons in the liquid are clamped by the solid surface. A similar clamping occurs at a polymer-glass interface. This results in the thermal generation of Lamb and Love waves in thin polymer films on glass substrates. These waves are observed as peaks in the Brillouin spectra. Finally, we have increased the load by taking the relatively heavy liquid metals mercury and gallium. The presence of thermal interface waves of the Stoneley-type showed up as strong peaks in the Brillouin spectra.

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