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[5]We thank D. J. Bromley for naming the Pan-Am texture by pointing out that this texture resembles a recognized commercial logo.

[6]Galileo Electro-Optics Corp., Sturbridge, Mass. 01518.

[7]S. Takagi, private communication.

[8]The symmetrical placement of resonance peaks bounding a region of finite absorption may indicate that the NMR coil is somehow coupling into a continuum of excitations in the superfluid's texture. Another possibility involving the Pan-Am texture is suggested in our contribution in Proceedings of the Fifteenth International Conference on Low Temperature Physics, Grenoble, France, 23–29 August 1978 (to be published).

[9]It is possible that for any given texture, the field and temperature dependences *will* separate, but that in our experiment by varying the field and temperature we are modifying the texture itself, and its corresponding $R_T(t)$. Since we have no way of locking the texture, however, we cannot determine if this speculation is correct.

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Brillouin Scattering from Surface Phonons in Thin Films

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Brillouin scattering from thermal surface phonons has been observed in thin films deposited on semi-infinite media. The velocity of Rayleigh and Sezawa waves was measured and interpreted in terms of the film elastic constants. Surface phonon attenuations were also measured and it was found that the lifetimes of surface and bulk phonons of the same wave vector are comparable in the glass studied.

The scattering of light by various types of thermally excited sound waves at surfaces of solids has been recently reported[1-4] and interpreted.[5-8] Brillouin scattering from bulk phonons has been investigated at the surfaces of opaque media[1,2] as well as in transparent free-standing films[3] by Sandercock, and in thin films deposited on semi-infinte media by Rowell, So, and Stegeman.[4] In the past year Sandercock[2] has also observed Brillouin scattering from thermal surface phonons on reflection from the surface of opaque materials such as metals and semiconductors. However, his attempts to study surface waves by Brillouin scattering in transparent media have proved unsuccessful to date.[2] Furthermore, Sandercock[2] found the measured surface phonon velocities to be (1–5)% lower than theoretical values calculated from bulk elastic constants. In this Letter we report the Brillouin scattering measurements of both the velocity and attenuation

of surface acoustic excitations (i.e., Rayleigh and Sezawa waves)[9] in thin glass films deposited on semi-infinite media.

The present experiment utilizes the techniques of integrated optics[10] to confine the incident and acousto-optically scattered light in and near the film. The optical waveguides were fabricated on fused silica microscope slides (refractive index = 1.4616) by the rf sputtering of a Corning 7059 glass film of thickness 3005 Å and refractive index 1.578, both measured by ellipsometry. As illustrated in Fig. 1, the incident light was inserted into the TE_0 waveguide mode using standard prism techniques[10] and the light scattered at ~90° into the TM_0 mode was coupled out by a second prism (~10% efficienty for each prism). The films were of excellent optical quality and waveguiding losses were typically 4 dB/cm.

The acousto-optic interaction for this scattering geometry has been discussed previously.[11] Since the dimensions of the scattering region are large in the plane of the film, but only a few optical wavelengths perpendicular to the surface, wave-vector conservation occurs only in the plane of the interfaces. Thus, the phonon wave vector parallel to the surface, i.e., $\vec{\kappa}_{\parallel}$, is given by $\vec{\kappa}_{\parallel} = \vec{K}_{\parallel}^{M} - \vec{K}_{\parallel}^{E}$ where $\vec{K}_{\parallel}^{M}$ and $\vec{K}_{\parallel}^{E}$ are the optical wave-vector components parallel to the surface for TM_0 and TE_0 modes, respectively. (Note that $|\vec{K}_{\parallel}^{M}| \neq |\vec{K}_{\parallel}^{E}|$ at the same optical frequency since the TE_0 and TM_0 modes satisfy different electromagnetic boundary conditions at the two film interfaces.) The scattering cross section has contributions[11] from both the elasto-optic and corrugation effects. The film thickness and scattering geometry for this experiment were chosen on the basis of a maximum in the scattering efficiency predicted using the techniques of Ref. 11.

The frequency spectrum of the scattered light coupled out by the second prism was analyzed using standard Brillouin-scattering instrumentation.[12] Approximately 200 mW of light at 0.5145 μm from a single-frequency argon-ion laser was focused by a 30-cm lens into the waveguide. Frequency analysis was achieved by a piezoelectrically scanned triple-pass Fabry-Perot interferometer with a contrast in excess of 10^8 and a long-term finesse > 50. Spectra were accumulated for periods of time of 4–24 h in a prototype DAS-1 stabilized data-acquisition system.[12]

A low-resolution spectrum (instrumental linewidth ≃ 0.5 GHz) is shown in Fig. 2. Four distinct spectral lines were observed and, to avoid confusion, additional components in the spectrum due to the overlapping of spectra from different Fabry-Perot orders have been removed. The signal at the peak of the 11.72-GHz line (see Fig. 2) was ~0.3 counts/sec channel (1 channel = 200 MHz).

The interpretation of the Brillouin spectrum requires an analysis of the phonon modes of a thin film on a semi-infinite medium. There are three orthogonal bulk modes, each characterized by $\vec{\kappa}_{\parallel}$ and the acoustic frequency Ω. The displacement fields are linear combinations of standing waves made up of the usual plane-wave phonons associated with an infinite medium and the relative amplitudes and phases are determined from the acoustic boundary conditions at both film in-

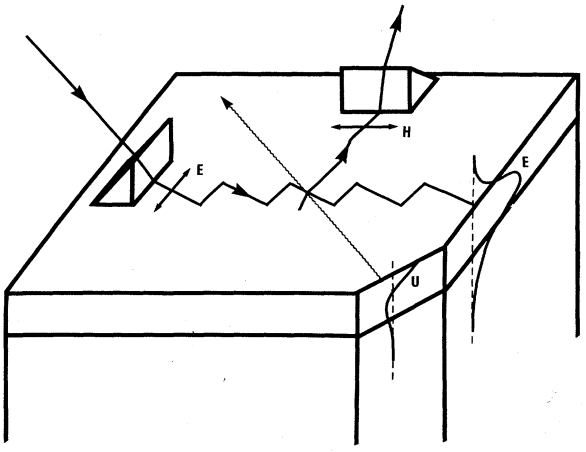

FIG. 1. Experimental sample geometry.

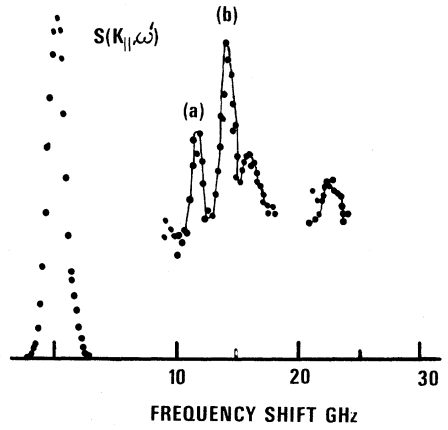

FIG. 2. Low-resolution Brillouin spectrum for the $TE_0 \rightarrow TM_0$ scattering geometry. Structures *a* and *b* identify the Brillouin lines associated with the Rayleigh and Sezawa modes, respectively. The remaining peaks are due to bulk phonons.

terfaces. For isotropic media two of these modes consist of shear and longitudinal standing waves in both the film and the substrate with displacement fields polarized in a plane defined by $\vec{\kappa}_{\parallel}$ and the normal to the surface, i.e., the sagittal plane. (The third mode which is not studied in this experiment is a standing shear wave polarized normal to the sagittal plane.) The present $TE_0 \rightarrow TM_0$ geometry corresponds essentially to depolarized scattering from shear strains associated with the sagittal-plane modes. Maximum scattering efficiency can be shown[11] to occur when the bulk phonon wave vectors are in the plane of the surface which corresponds to shear waves traveling parallel to the film. Therefore the spectral peak at 15.5 GHz corresponds to a frequency shift of $\Omega = \kappa_{\parallel}(v_T)_s$ where $(v_T)_s$ is the shear-wave velocity in the substrate. If $(v_T)_f$, the shear-wave velocity in the film, is less than $(v_T)_s$, the shear wave vectors in the film are always larger than in the substrate and bulk modes of the film-substrate system characterized by shear wave vectors in the film parallel to the surface do not exist. Since the film represents a negligible fraction of the total sample volume, bulk phonon modes are thermally excited only if the shear wave vectors in the substrate are real (rather than complex) quantities. Therefore the spectral lines at $\Omega < \kappa_{\parallel}(v_T)_s$ must correspond to scattering from thermal surface phonons. Since the wave vectors associated with the TE_0 and TM_0 modes in the film are not parallel to the surface, weak scattering also occurs at $\Omega \simeq \kappa_{\parallel}(v_L)_f$, i.e., from longitudinal waves in the film traveling approximately parallel to the surface. This is the origin of the spectral line at a frequency shift of ~22 GHz.

Surface waves in thin films on semi-infinite media have been discussed by Adler and Farnell.[9] In the substrate $\kappa_{\parallel} > \Omega/(v_T)_s$ and the sagittal-plane–confined fields decay exponentially into the substrate. There always exists one Rayleigh wave, the velocity of which varies with film thickness from the Rayleigh velocity of the substrate to that of the film material (see Fig. 3). Higher-order surface modes commonly called Sezawa waves can also occur if $(v_T)_s > (v_T)_f$, depending on the film thickness. They are propagating shear waves trapped in the film and occur at discrete frequencies (given $\kappa_{\parallel}$ and h) since the acoustic phase must change by an integer multiple of 2π in one roundtrip transit of the film. Their surface wave velocities lie between that of shear waves in the film and substrate. The dispersion relations shown in Fig. 3 were calculated using

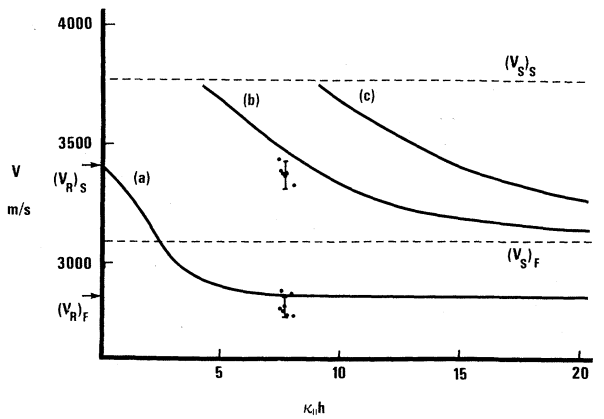

FIG. 3. Dispersion in surface wave velocity with normalized film thickness ($\kappa_{\parallel}h$). Curves *a*, *b*, and *c* refer to the Rayleigh wave, lowest-order Sezawa wave, and the next-order Sezawa wave, respectively. Typical error limits are shown.

values of $(v_T)_f$ and $(v_L)_f$ for bulk Corning 7059 glass. Note that for the value of $\kappa_{\parallel}h$ appropriate to this experiment (> 7), the Rayleigh-wave amplitude is very small at the film-substrate interface, and the surface phonon velocity corresponds very closely to that of a semi-infinite medium of Corning 7059 glass.

The measured velocities of the Rayleigh and Sezawa waves were found to be lower than the calculated values as shown in Fig. 3. It was not possible to accurately measure surface phonon velocities over a reasonable range of $\kappa_{\parallel}h$ because of the strong stray light scattering from inhomogeneities in the film at angles away from 90° and because scattering cross section was strongly peaked at the thickness used. Subsequently $(v_S)_f$ was evaluated by Brillouin scattering in a geometry in which the light is scattered in a direction normal to the waveguide.[4] [In addition $(v_L)_f$ was measured from the film longitudinal mode "cutoff" in a $TM_0 \rightarrow TM_0$ geometry at a 90° scattering angle.] The longitudinal velocity was found to decrease by ~0.6% to $(v_L)_f = 5560$ m/s but the depolarized spectrum was unfortunately relatively insensitive to $(v_S)_f$ and it was found to correspond to the bulk value to an accuracy of ~5%. However we found that by changing the value of $(v_S)_f$ from 3090 m/s to 3010 m/s, i.e., by 2.9%, both the Rayleigh and Sezawa wave velocities could be made to agree with calculations to better than the experimental uncertainty of ~1.5%. Noting that changes as large at 10% have been reported previously for other materials,[4] we attribute the lowering of the surface wave velocity to changes

in the film elastic constants during fabrication. We also note that mechanical polishing is known[13] to produce surface damage layers and dispersion in surface phonon velocities. Although neither of the film interfaces were polished in the present experiment, surface damage layers may be a partial cause of the discrepancies found by Sandercock.[2]

The surface-wave lifetimes were evaluated by measuring the spectral breath of the Brillouin lines under high resolution (instrumental linewidth $\simeq$ 70 MHz, full width at half-intensity). Since a surface wave can be completely described by $\kappa_{\parallel}$ (which is specified uniquely by the scattering geometry), then the Brillouin line shape is proportional to $[(\omega_0 \pm \Omega)^2 + (1/\tau)^2]^{-1}$ for perfectly monochromatic incident light of frequency ω_0 where τ is the phonon lifetime. The observed linewidths were 180 and 360 MHz for the Rayleigh and Sezawa waves, respectively. After correction for the instrumental line profile, the finite acceptance angle at the interferometer and the finite width of the incident light in the film (all by successive convolution with a Lorentzian line profile), the lifetimes given in Table I were deduced. Since the Sezawa mode is guided by both film interfaces, we attribute the shorter lifetime (and hence larger attenuation) of this mode relative to the Rayleigh wave as due to inhomogeneities in the film boundaries.

The attenuation of shear and longitudinal waves in both a silica microscope slide and a bulk sample of Corning 7059 glass was also measured at a 90° scattering angle. (Unfortunately, the spectral lines at 15 and 22 GHz in the guided-wave spectrum of Fig. 2 are spectrally broadened because of scattering from a phonon continuum characterized by a single $\kappa_{\parallel}$ but different values of $\kappa_{\perp}$, the wave-vector component normal to the surfaces.[1,4]) The results are listed for comparison in Table I. Assuming the usual $\tau \propto \Omega^{-2}$ dependence, surface phonons have the shortest lifetimes of all acoustic waves at a common frequency. However, when compared at approximately the same wave vector, the lifetimes are comparable. We note that such a relation could be indicative of strain- or impurity-dominated damping. (The waveguides were not annealed after fabrication.)

In summary we have measured the velocity and attenuation of high-frequency (> 10 GHz) Rayleigh and Sezawa waves using Brillouin scattering. Surface wave velocities lower than those calculated from bulk elastic constants are attributed to changes in film acoustic velocity during preparation. This type of measurement may well prove useful in the future for studying phase transitions in thin films, anisotropic compressibilities in molecular films, etc.

TABLE I. Phonon frequencies and lifetimes for bulk and surface waves at a 90° scattering angle.

Phonon mode	Frequency (GHz)	τ (10^{-9} s)
Rayleigh	11.7	2.6 ± 0.3
Sezawa	13.7	1.0 ± 0.1
Longitudinal (SiO_2)	24.1	2.6 ± 0.3
Shear (SiO_2)	15.0	2.2 ± 0.4
Longitudinal[a] (Corning 7058)	26.6	2.0 ± 0.2
Shear[a] (Corning 7059)	14.9	2.1 ± 0.4

[a]Measured with 0.488-μm incident light.

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