Observation of organ-pipe acoustic excitations in supported thin films

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Brillouin light scattering from supported silicon oxynitride films reveal an extended series of acoustic excitations occurring at regular frequency intervals when the mode wave vector is perpendicular to the film surface. These periodic peaks are identified as distinct standing wave excitations that, similar to harmonics of an open-ended organ pipe, occur due to the boundary conditions imposed by the free surface and substrate-film interface. The surface ripple and volume elasto-optic scattering mechanisms contribute to the scattering cross sections and lead to dramatic interference effects at low frequencies where the surface corrugations play a dominant role. The transformation of these standing wave excitations to modes with finite in-plane wave vectors is also investigated. The results are discussed in the framework of a Green's-function formalism that reproduces the experimental features and illustrate the importance of the standing modes in evaluating the longitudinal elastic properties of the films.

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Brillouin light scattering (BLS) from thermally excited, high-frequency acoustic excitations is emerging to be of great value in the study of acoustic and elastic properties of bulk and laminar structures.\textsuperscript{1-3} For an opaque or semitransparent medium, surface excitations polarized in the sagittal plane are the primary modes observed in these experiments. The corresponding scattering cross sections are controlled mainly through surface corrugations generated by the modes (ripple mechanism) and only the wave-vector component parallel to the surface (\(K_z\)) need be conserved during scattering. On the other hand, for sufficiently thick transparent materials the volume elasto-optic (E-O) interference mechanism is dominant and all components of momentum are conserved. For relatively thin films where the E-O mechanism is operative this condition on momentum is relaxed and results in the broadening of bulk phonons.\textsuperscript{4} BLS has been successful in probing a variety of near surface excitations such as Rayleigh, Sezawa, Love, Lamb modes as well as guided acoustic resonances in thin films.\textsuperscript{5-14}

Despite an extensive body of BLS experiments, standing acoustic-wave excitations in laminar structures that are characterized only by wave-vector components perpendicular to the thin-film surfaces (\(K_z\)) have received little attention. In this case due to the limited film thickness (\(d\)) and applicable acoustic boundary conditions, the allowed values of \(K_z\) are quantized to a series of discrete values. Such constraints on \(K_z\) are in contrast to modes with finite \(K_z\) components that take on a continuum of values and describe traveling wave excitations that are evident in most BLS studies. Acoustic excitations having only \(K_z\) components offer previously unexplored opportunities to probe the elastic properties of thin film structures.

In this paper we report on the observation of a series of standing wave acoustic excitations over an extended frequency range for scattering angles of 0° (\(K_z=0\)). This special scattering arrangement was undertaken to measure the photons backscattered essentially along the film normal and thus probe only \(K_z\) components of acoustic excitations. An extensive number of discrete excitations mediated by the ripple scattering mechanism are observed well below the bulk LA mode which also separate into a distinct set of modes in the vicinity of the LA mode where the elasto-optic mechanism is more effective. Our results show that the modes closely follow the behavior of the harmonics of an organ-pipe with a maximum displacement at the surface and a node at the film-substrate interface. This represents the first observation of such acoustic excitations by Brillouin light scattering and is complementary to inelastic helium-atom scattering which probe similar, albeit short wave, excitations for very thin (2–20 monolayer) films.\textsuperscript{15} We also show that when the scattering angle is changed away from normal incidence, finite \(K_z\) effects are operative and the modes become dispersive. The results are analyzed on the basis of a Green's-function formalism where calculations for \(K_z=0\) show all of the important observed characteristic BLS features including the multitude of modes, their linewidths and variations with film thickness \(d\) and scattering angle. Moreover, the periodic absence of particular Brillouin peaks from these films is traced to the destructive interference between ripple-mediated scattering amplitudes at the film boundaries.

The silicon oxynitride films used in this study were grown to thicknesses \(d=0.25, 1, 2\), and 3 μm on (001) GaAs wafers as discussed elsewhere.\textsuperscript{16} The BLS measurements were performed in backscattering at ambient temperature using approximately 70 mW p-polarized 514.5 nm radiation. The standing wave excitations were most clearly observed at small scattering angles and special care was taken to intercept the back-reflected laser beam. The signals sampled in this case are those collected from within the finite solid angle of the f/2.3 collection lens which, for the incident and back scattered beams perpendicular to the film surface, is nominally referred to as \(\theta=0°\) scattering below.

Figure 1 shows BLS spectra recorded from each sample along the [100] direction at a scattering angle of 0°. These modes thus result from excitations that nominally have a vanishing \(K_z\) wave-vector component. The most interesting

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FIG. 1. Typical Brillouin spectra recorded from the $d=0.25, 1, 2$, and $3 \mu m$ samples along the [100] direction at a scattering angle of $\theta=0^\circ$. The distinct sets of periodic peaks are standing wave acoustic excitations. The solid lines are calculations of the mode densities as described in the text for the different films. The dashed curve represents calculations for the semi-infinite film. In comparison to the calculated spectra, alternate low-frequency excitations are missing from the experimental spectra in the 1, 2, and 3 $\mu m$ films.

Aspect of the data is the appearance of a series of nearly equally spaced peaks whose frequencies extend from about 10 to 60 GHz. The spacing $\Delta \nu$ for neighboring peaks decreases from 12.2, 5.7, 2.9, and 1.9 GHz respectively for the $d=0.25, 1, 2$, and $3 \mu m$ thick layers. For the thickest film ($d=3 \mu m$), the peaks are tightly spaced and only excitations from about 30 GHz in the vicinity of the bulk LA mode (~39 GHz) are distinctly observable. At lower thicknesses, $d=1 \mu m$ and 2 $\mu m$ an entire series of modes extending from as low as 10 GHz are evident with the strongest peaks again appearing near the bulk LA frequency. Consistent with the relatively large film thickness, the bulk LA-like peak at 39 GHz in the 2 and 3 $\mu m$ samples is sharp and intense, reflecting the strong volume elasto-optic (E-O) scattering. In contrast to the thicker films, the absence of the LA-like mode in the $d=0.25 \mu m$ film is due to the weakness of the E-O contribution arising from the film thickness being less than the phonon wavelength.

Figure 2 shows spectra from the $d=0.25$ and 1 $\mu m$ samples at various scattering angles and illustrates that the finite frequencies of the modes at $\theta=0^\circ$ increase further with increasing $\theta$ (or equivalently increasing $K_\parallel$). This behavior is thus different from Rayleigh surface waves whose frequency is proportional to $\sin \theta$ and therefore vanishes as $\theta$ approaches zero. The peak widths associated with the series of low lying modes vary from sample to sample and generally increase as $d$ decreases and remains nearly constant for a given sample at different $\theta$—a behavior that also distinguishes them from surface acoustic waves whose peaks broaden considerably at small $\theta$.\footnote{\textsuperscript{1,7}} In addition, in the vicinity of the LA mode the separation between the peaks is found to be approximately one half of the peak spacing at low frequencies. As discussed below, this difference arises from interference effects that subdue alternate low-frequency excitations.

In order to gain insight into their origin and to evaluate the properties of these excitations, the projected local density of states (LDOS) $D_l$ was evaluated within the Green's-function formalism\textsuperscript{18,19}

\begin{equation}
D_l(\omega^2, K, x_s) = -\frac{1}{\pi} \text{Im} G_{ii}(K, x_s, \omega^2). \quad \text{Here } i \text{ refers to the mode polarization; } i = 1 \text{ for longitudinally polarized excitation, } 2 \text{ and } 3 \text{, respectively, for shear horizontal and sagittal polarization normal to the surface. } G_{ii} \text{ is the } (x_i, x_i) \text{ component of the Fourier (frequency and wave vector) domain elasto-dynamic Green's-function tensor for depth } x_s. \text{ The method of calculation of } G_{ii} \text{ is provided in Ref. 5.}
\end{equation}

In Fig. 1, the calculated LDOS at the free surface of each film for $D_3$ (solid curves) are compared to the experimental spectra. In the calculation, the elastic constants of the amorphous silicon oxynitride films are taken from Ref. 12, and include a small (~10%) modification to $C_{11}$ to yield the best fit to the spectra. The density of the films is 2.40 g/cm\textsuperscript{3}.\textsuperscript{16} The well-known elastic constants and density for the GaAs substrate were also utilized.\textsuperscript{20} The calculations of $D_3$ for a scattering angle of $\theta=0.1^\circ$ reveal a series of nearly equally spaced peaks that, in agreement with the measured BLS spectra, become more dense and narrower when $d$ increases. These periodic peaks are a special type of Lamb waves where the $K_\parallel$ components are essentially zero and, as discussed below, represent standing wave excitations in the films. The dashed curve in Fig. 1 is the calculation of $D_3$, for $\theta=0.1^\circ$ when the film is regarded as a half space me-
FIG. 3. Spatial distributions of the square displacement field perpendicular to the film surface \(U^2_\theta\) at frequencies of 16 and 18.9 GHz for the standing wave excitation in the 1 \(\mu\)m film at \(\theta=0^\circ\). Calculation for a nonpeak frequency of 17 GHz is illustrated for comparison.

It is seen that the closely spaced ripple structure is subdued in this case and an extended shoulder resulting from the continuum bulk waves is formed.

As an existence criterion for stable standing acoustic excitations, it is necessary that the partial waves in the films interfere constructively. Thus \(K_{\parallel}\) must obey the resonance condition

\[2 K_{\parallel} d + \delta_4 + \delta_1 = 2m \pi,\]

where \(\delta_4, \delta_1\) are the phase changes upon reflection at the two film surfaces, and are determined from the boundary conditions. The mode index \(m\) is an integer. Figure 3 illustrates the spatial distributions of the squared displacement field \(U^2_\theta\) as a function of the depth in the film (depth <0) and in the substrate (depth>0) for the 1 \(\mu\)m film at \(\theta=0.1^\circ\). (Components \(U_3\) and \(U_1\) are not shown for clarity). The displacements are calculated at frequencies of 16 and 18.9 GHz that correspond to peaks in the calculated mode density \(D_3\). It is evident that the associated displacement amplitudes are periodic where, in addition to a constant phase change of \(\pi/2\), a discrete number of half wavelengths are supported in the film. The contribution from \(\delta_4\) thus results in the above equation being satisfied and hence \(K_{\parallel}\) is quantized, leading to stable standing wave excitations. The calculations (Fig. 3) also depict that the displacement is maximum at the free surface and has a node at the film-substrate interface. This behavior closely follows that of standing wave harmonics of an organ pipe. For comparison, the displacement distribution for a nonpeak frequency of 17 GHz is also shown in Fig. 3. It is seen that not only is the displacement amplitude relatively small, but also an integral multiple of half wavelengths is not contained within the thickness \(d\). Therefore a mode at 17 GHz does not lead, in agreement with the BLS spectra, to a resonance mode in this film.

It is evident from Fig. 1 that several of the modes predicted from the above analysis are not observed in the BLS spectra, and that these missing excitations occur at regular frequency intervals. For instance, in the 1 and 2 \(\mu\)m films, alternate predicted modes are absent from the spectra until about 39 GHz, the bulk LA mode frequency. Figure 4 summarizes the calculated mode frequencies and experimental data at \(\theta=0^\circ\) as well as their variations with \(\theta\) (Fig. 2) for the 1 \(\mu\)m film. The apparent discrepancy of missing modes is due to interference of light associated with the ripple mechanism. Since the silicon-oxynitride films are transparent and the interfaces are smooth, light scattered from the top and bottom interfaces have similar intensities and interfere. The resulting scattering cross section depends on the signs of the displacement components \(U_3\) at the two film interfaces and the phase difference of the two beams. Due to the signs of \(U_3\), the only time there would not be any suppression of alternate peaks is if the phase difference of the photons scattered from the upper surface and interface were exactly in quadrature. A deviation as small as 10° from quadrature, for example, would lead one set of peaks having double the intensity of the other. The estimated phase difference between the two beams from the optical path lengths is

\[n2d/\lambda\]

in multiples of \(2\pi\), where \(n=1.68\) is the refractive index of the film and \(\lambda\) is photon wavelength. For the 1, 2, and 3 \(\mu\)m thick samples this phase difference is 1.63, and contributes to a partial suppression of alternate peaks. For the 1, 2, and 3 \(\mu\)m thick samples it is 6.52, 13.05, and 19.56, respectively, which are all almost perfectly valued (i.e., integer or half integer multiples of \(\pi\)) for the maximum suppression of alternative modes and hence account for the BLS spectra. Upon tracking the modes and their variation with \(\theta\), it is found (Figs. 2 and 4) that some modes appear at certain scattering angles and disappear at others. This behavior, not
surprisingly, illustrates that the interference is affected by the value of \( \theta \). However, in the frequency range where elasto-optic scattering is more pronounced, i.e., around the LA mode frequency of 39 GHz, there are no “missing” modes and all of the predicted excitations are evident in this range. This behavior is consistent with the E-O mechanism not being subject to the interference effects as with the ripple-mediated coupling.

Excitations with finite in-plane wave-vector components, i.e., \( K_x \neq 0 \), are traveling waves that, in contrast to the \( K_z \) components, can have a continuum of values. Figure 2 illustrates such modes observed for different \( \theta \), and hence different \( K_x \), for the 0.25 and 1 \( \mu m \) samples. The strong peaks in the vicinity of the LA mode for the 1 \( \mu m \) sample result from volume elasto-optic scattering and contain double or triple structures, similar to a previous report.\(^4\) It is evident that while some of the standing wave modes can be tracked through large \( \theta \), due to interference and the overlap with bulk excitations, others cannot be observed at finite scattering angles. At large \( \theta \), the \( K_z \) components become important and the standing wave mode may evolve into one or several decoupled (longitudinal or transverse) modes. This feature is reflected in splittings in the calculated mode frequencies that appear in Fig. 4.

It is thus seen that BLS from standing wave acoustic excitations becomes significant when the film is thin and \( \theta \) is small. Detailed calculations reveal that their position and frequency spacing are in fact very sensitive to the effective longitudinal elastic constant of films. For example, the frequencies of the standing wave excitations supported in the current samples vary by 1% if \( C_{11} \) is altered by 2% at \( \theta = 0^\circ \). As shown in Figs. 1 and 4, the best fit elastic constants \( C_{11} \) (85 GPa) provides excellent agreement for the standing wave frequencies, their frequency spacing, linewidth, and variations with film thickness and scattering angle. For very thin films, BLS is one of the few available techniques to investigate their elastic properties. In this case the often observed surface and Sezawa acoustic waves are generally most sensitive to shear elastic properties of the films.\(^5\)\(^,\)\(^24\)\(^,\)\(^25\)

Therefore, the observation of the standing modes will provide an important avenue to reliably investigate the effective longitudinal elastic properties of the films.

The series of organ-pipe type harmonics we report are to be contrasted to folded phonons observed in superlattices wherein the layering leads to Raman-active zone-center phonons.\(^26\)\(^,\)\(^27\) They are also distinct from Sezawa and Love eigenmodes localized in a film that are characterized by finite in-plane wave vector. The high resolution offered by BLS hence now opens a different avenue, along with light-atom scattering,\(^15\) to probe confined standing wave excitations that are akin to the acoustic resonances in open and closed pipes.

In conclusion, we have presented conclusive evidence for extended harmonics of standing wave acoustic excitations, in supported silicon oxynitride films. These standing wave modes, observed by Brillouin light scattering, are also revealed by theoretical simulations based on calculating the associated elastodynamic Green’s tensor that allows the local density of states as well as their spatial mode displacements to be determined. The low-frequency BLS spectra reveal that the excitations are primarily due to the ripple mediated scattering and therefore subject to interference effects associated with the film boundaries. The corresponding excitations in the vicinity of the LA frequency are largely mediated through the volume elasto-optic mechanism and thus their scattering cross sections are not strongly influenced by the interfering contributions. The study provides a valuable step in classifying guided excitations of layered systems that also opens up a direct means for investigating the longitudinal elastic properties in structures, especially when the films are very thin.

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