Evidence for a shear horizontal resonance in supported thin films

X. Zhang,[1] M. H. Manghnani,[1] and A. G. Every[2]
[1]*School of Ocean and Earth Science and Technology, University of Hawaii, Honolulu, Hawaii 96822*
[2]*Physics Department, University of Witwatersrand, Johannesburg, South Africa*
(Received 28 January 2000; revised manuscript received 16 May 2000)

We report evidence for a different type of acoustic film excitation, identified as a shear horizontal resonance, in amorphous silicon oxynitride films on GaAs substrate. Observation of this excitation has been carried out using surface Brillouin scattering of light. A Green's function formalism is used for analyzing the experimental spectra, and successfully simulates the spectral features associated with this mode. The attributes of this mode are described; these include its phase velocity which is nearly equal to that of a bulk shear wave propagating parallel to the surface and is almost independent of film thickness and scattering angle, its localization mainly in the film, and its polarization in the shear horizontal direction.

Surface Brillouin scattering (SBS) is proving to be of great value in the study of the acoustic excitations and elastic properties of bulk materials and thin supported layers.[1–4] Thermally excited phonons in the GHz frequency range are responsible for the light scattering in SBS. For an opaque or semi-opaque medium, the light scattering is mediated principally by the surface ripple mechanism, and involves interaction with surface excitations polarized in the sagittal plane, defined by the surface acoustic wave (SAW) propagation wave vector $\mathbf{K}_{\|}$ and the surface normal. SBS has been successful in studying a variety of surface excitations including longitudinal resonances (LR) or longitudinal guided acoustic modes (LGAM), which are surface excitations with frequencies and phase velocities that are close to those of bulk longitudinal modes propagating along the surface. These surface excitations are often difficult to detect with traditional acoustic techniques, but show up in light scattering spectra. LR have been studied extensively in recent years,[5–12] not only because of their intrinsic interest as a distinct feature in SBS spectra, but also because they can be used to determine the effective longitudinal elastic constants of a medium with unknown elastic properties.

In contrast to the success of LR, guided shear horizontal (SH) excitations, or SH resonances (SHR), i.e., transverse phonons polarized in the plane of the surface, have received relatively little attention, at least from the experimental point of view. Although calculations of Brillouin scattering cross sections for shear horizontal surface acoustic modes (called Love modes in the discrete part of the spectrum) have been performed by several authors,[13–16] SH excitations have been considered as sources of SBS in a very limited number of papers. To our knowledge, there are up to now only a few reported experimental reports on the detection of SH surface acoustic phonons using Brillouin scattering.[17–20] In these observations, the phase velocities are dispersive with scattering parameters, and these observed modes are therefore not strictly SHR's the counterpart of LR.

SH modes are polarized parallel to the surface, and the mechanism of their interaction with light is only through the volume elasto-optic effect; therefore they can be observed only for strong elasto-optic coupling media. In this paper, we suggest a scattering route as shown in Fig. 1, involving inelastic scattering of the incident beam after it has been reflected from the interface between the film and the substrate. This scattering route is similar to that of the platelet geometry described in the literature,[21] and involves collecting the light scattered by the guided acoustic excitations with wave vector $\mathbf{K}_g$ in the frequently used back-scattering configuration. The literature reports of LR in various layer systems[9–12] are believed to result from this scattering process. An additional advantage of this scattering geometry is that the phase velocity of the guided phonons probed can be calculated directly from the frequency shift of the scattered light and scattering angle, without previous knowledge of the refractive index of the film.

The samples used in our experiments are silicon oxynitride films, of thicknesses 1, 2, and 3 μm, on (001) GaAs wafers, and have been provided by Motorola Co. The films are of particular interest because of their potential use in advanced microchips and SAW device applications.

The Brillouin light scattering measurements were performed for the samples at ambient temperature using the *p*-polarized 514.5 nm line from an argon-ion laser operating in a single axial mode. The laser beam is focused onto each sample by a lens of aperture $f/2.3$ in the backscattering arrangement shown in Fig. 1. The wave vector $\mathbf{k}_i$ of the incident light makes an angle θ with the surface normal, and the light backscattered into a small cone around $-\mathbf{k}_i$ is collected

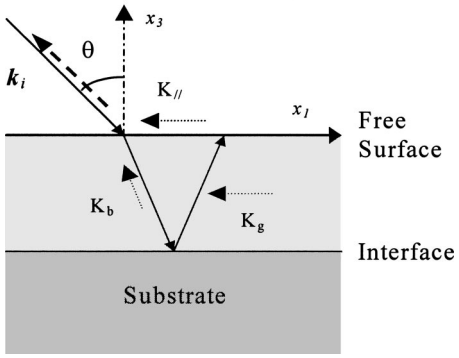

FIG. 1. Back-scattering configuration of SBS. $\mathbf{K}_{\|}$, $\mathbf{K}_b$, $\mathbf{K}_g$, and $\mathbf{k}_i$ represent the wave vector of surface phonons, bulk phonons, guided acoustic excitations, and incident laser, respectively.

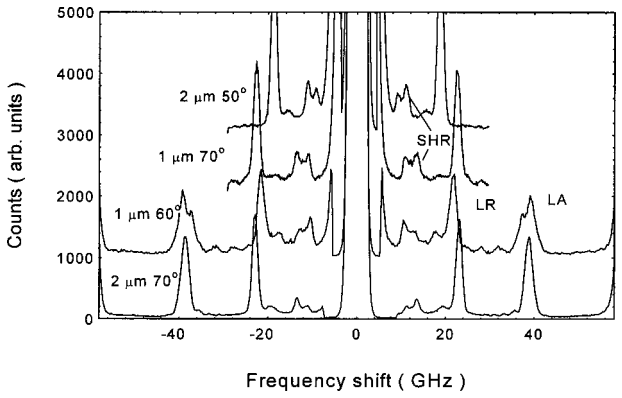

FIG. 2. Typical Brillouin spectra for 1 and 2 μm samples along the [100] direction on the [001] surface referred to the GaAs substrate at some scattering angles. The upper two and lower two spectra were collected using a free spectrum range of 31.3 and 62.5 GHz, respectively. The strong peak located around 40 GHz is due to the scattering from the bulk longitudinal phonons, while the prominent features located in the 18–24 GHz range are the scattering from LR.

by the same lens. The power of the laser beam focused onto the sample is approximately 70 mW. The scattered spectrum was analyzed by a Sandercock $(3+3)$ pass tandem Fabry-Perot interferometer. The detector was a high-quantum efficiency silicon avalanche photodiode. The spectra were taken at different scattering angles, from 30° to 75°, hence to have different wave-vector components parallel to the surface ($\mathbf{K}_\parallel$ for SAW or K_g for guided modes). In order to obtain the same measurement accuracy at different scattering angles, slits of the appropriate size were used to limit the collection aperture of the scattered light entering the interferometer.

Figure 2 shows some typical spectra taken for the 1 and 2 μm samples along the [100] direction on the [001] surface referred to GaAs substrate at different scattering angles. The spectra were collected for different scattering angles, and using different free spectrum ranges (FSR), to verify the appearances of the peaks. The strong peak located around 40 GHz is due to the scattering from bulk longitudinal acoustic modes (LA), indicated by the fact that the peak position does not shift with variation of incident angles. It is noticed that the LA peak for the 1 μm sample is broadened as a result of the scattering volume being limited by the finite thickness of the film.[22] The second prominent feature in the spectra located in the 18–24 GHz range, depending on the scattering angles, is identified as scattering from LR. It is observed for all three samples and at different scattering angles. The frequency shift varies proportionally to $\sin(\theta)$ and the corresponding phase velocity (6.32 km/s) is, within experimental error, independent of scattering angle. The intensity of the peak evidently benefits from the combined effect of the high elasto-optic coupling of the medium and the new scattering route. The high reflectivity of the interface between the film and the GaAs substrate also plays an important role. Since the film is isotropic, the phase velocity of LR is approximately equal to that of the bulk longitudinal waves, and the refractive index of the film can be obtained by comparing the frequency shift of the bulk longitudinal peak and the LR peak; for the present sample, it is estimated to be about 1.59.

In the spectrum, a third less intense peak is present in the lower frequency range (~14 GHz for $\theta=70°$). The peak is observed also for all three samples. The frequency shift of the peak is found to be proportional to $\sin(\theta)$, which means that the corresponding phase velocity (3.74 km/s) is also independent of the scattering angle. In order to identify the origin of this peak, the polarization of the scattered light was analyzed, and spectra were collected for light scattering (i) only in *s* polarization (denoted by *p-s*), (ii) only in *p* polarization (denoted by *p-p*) and (iii) unpolarized (*p-ps*). Figure 3 shows the spectra taken for the 2 μm sample at 50° (a), for the 1 μm sample at 50° (b) and 60° (c), respectively, all for the [100] direction on the (001) surface referred to the GaAs substrate. It is evident that all the features are polarized. The third less intense peak is *s* polarized. As discussed in the literature,[3,18] Brillouin scattering of a polarized photon off a pure SH phonon produces a scattered photon with a polarization rotating by 90°, hence the third peak is identified as scattering from a SH excitation.

In order to gain insight into the origin of, and to evaluate the properties of these excitations, the local density of states (LDOS) was evaluated within the Green's function formalism,[23,24]

$$D_i(\omega^2,K,x_3)=-(\pi)^{-1}\,\mathrm{IM}\,G_{ii}(K,x_3,\omega^2).$$

Here *i* refers to the mode polarization; $i=1$ for longitudinally polarized excitation, 2 and 3, respectively, for shear horizontal and sagittally polarized excitation normal to the surface. G_{ii} is the (x_i,x_i) component of the Fourier (frequency and wave vector) domain elastodynamic Green's function tensor for depth x_3. The method of calculation of the Green's function is provided in Ref. 4.

The advantages of using the Green's function method for the analysis are several. Firstly, the mode LDOS for each polarization component can be analyzed for a given frequency, allowing the relative contribution as well as the spatial distribution of each polarization component to be evaluated. Secondly, peaks in the curves of the LDOS for different film thicknesses and phonon wave vectors provide for the variation of frequencies or phase velocities with $\mathbf{K}_\parallel d$, thus enabling a direct correlation to the experimental spectra. This procedure hence avoids the need of prior knowledge of the elasto-optic coefficients to determine mode frequencies via the scattered light intensity. On the other hand, the disadvantage of the approach is also obvious: it does not incorporate the elasto-optic coefficients, therefore it cannot give a full account of the spectral intensity resulting from elasto-optic scattering.

In Fig. 3, the calculated LDOS at the free surface of the film for longitudinal and SH excitation are compared to the experimental spectra. In the calculation, the literature values of the elastic constants and density for GaAs substrate were used.[25] The two independent elastic constants ($C_{11}=95.9$ and $C_{44}=33.6$ GPa) of the amorphous silicon oxynitride films used in these calculations were obtained directly from the experimental data. The density of the films[26] is 2.40 g/cm^3.

The LDOS have two regions: a discrete part and a continuous part, separated by the threshold of the bulk shear waves in the film. The dominant feature in the discrete region of D_1 is the highly localized true Rayleigh SAW. The displacement of the Rayleigh SAW has a longitudinal com-

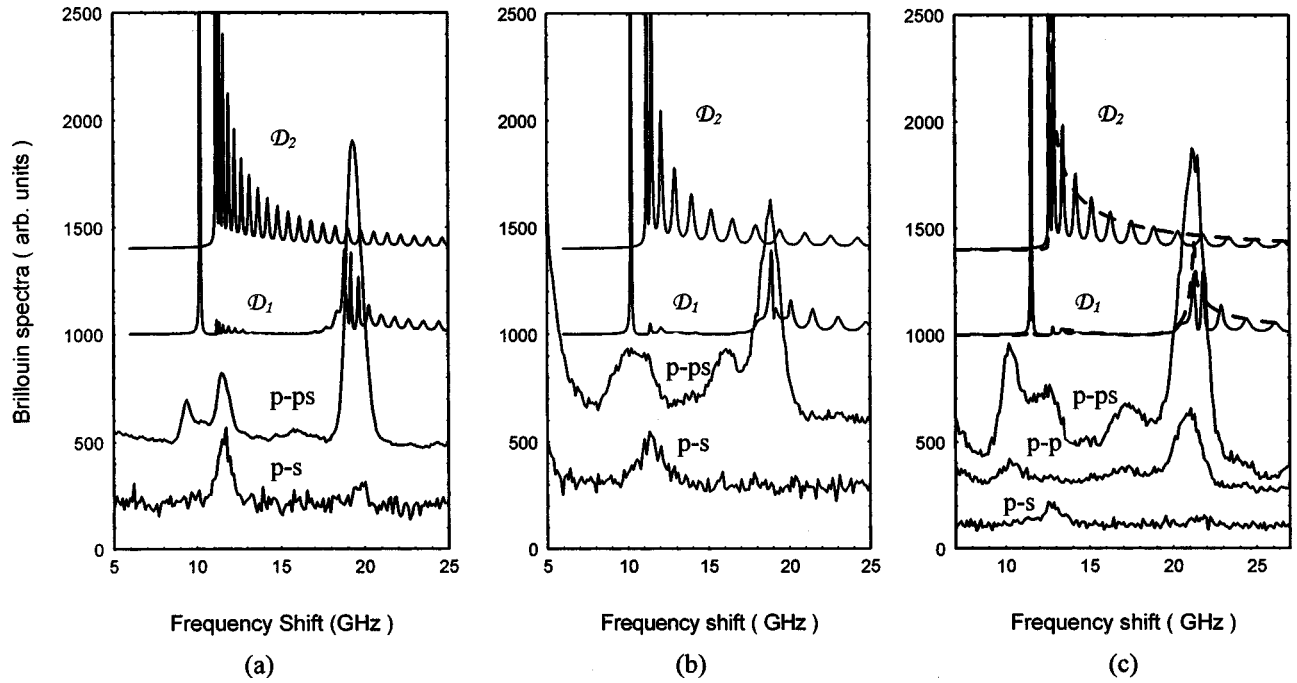

FIG. 3. The spectra for the 2 μm sample at 50° (a), for the 1 μm sample at 50° (b) and 60° (c), respectively, all taken for the [100] direction on the (001) surface referred to as the GaAs substrate. The polarization of the scattered light was analyzed by *s*-polarized (denoted by p-s), *p*-polarized (p-p), and unpolarized (p-ps). D_1 and D_2 are the calculated LDOS for longitudinal and SH components. It is evident that two resonances occur in the spectra. The dashed curves in (c) are the calculated LDOS for the film to be considered as a half space medium.

ponent; therefore it shows up in D_1. The discrete region of D_2 shows no peak. The continuous regions of both D_1 and D_2 are composed of closely spaced ripples; for D_1 these ripples are called highly attenuated pseudo-Lamb modes, while for D_2 the ripples are called pseudo-Love modes. It is evident that the ripples resonate at certain frequencies. For D_1, resonance occurs at the frequencies of 19 and 21 GHz, respectively, for a scattering angle of 50° and 60°; this resonance is called LR or LGAM in the literature.[5–12] The resonance frequency is close to that for a bulk longitudinal wave propagating parallel to the surface. For D_2, the resonance occurs near the frequencies of 11 and 13 GHz, respectively, for scattering angles of 50° and 60°. The polarization of this resonance is shear horizontal. The frequency of the resonance is also close to that of a bulk transverse wave propagating parallel to the surface. The resonance in D_2 is completely analogous to the LR and its frequencies are in a good agreement with experimental spectra; therefore we identify this resonance as a SH resonance or SH guided acoustic mode.

Considering that our films are quite thick, we may neglect the effect of the substrate to a first approximation. The dashed curves in Fig. 3(c) are calculations of D_1 and D_2 for a scattering angle of 60° when the film is regarded as a half space medium. It is seen that the closely spaced ripple structures are subdued and the resonance peaks are pronounced. In this case the positions of SHR and LR peaks are located precisely at the thresholds for shear transverse and longitudinal bulk waves, which are the respective velocities of bulk waves propagating parallel to the sample surface. Comparing with the calculations for the 2 and 1 μm samples, it is observed that the continuous peaks are fragmented into a number of closely spaced ripples when the film has a relatively small thickness, and only the ripples with frequencies close to that of the threshold have relatively large values of LDOS and hence may be observed in the experiments. What we observed in the experiments are thus in fact the envelopes of these ripples with frequencies that lie close to that of the thresholds.

Being principally a SH polarized excitation, the SHR dominates the SH response at the frequency of this mode. This is revealed in another way in Fig. 4 as the spatial distributions of the squared displacement field at the frequency of 11.2 GHz in the 1 μm film at $\theta = 50°$. It is evident that only the SH displacement component U_2 of the mode is significant. From the spatial distribution of the square of the displacement field, it is seen that the SHR is mainly localized

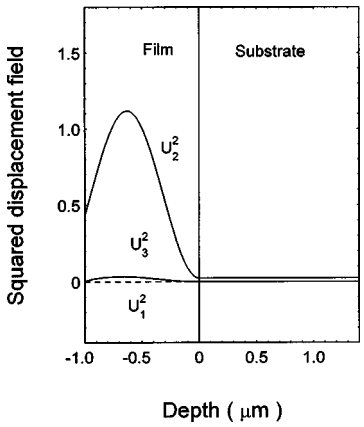

FIG. 4. The spatial distributions of square displacement field at the frequency of 11.2 GHz for the SHR mode in the 1 μm film at $\theta = 50°$. It is evident that the SHR mainly localizes in the films and only its SH displacement component U_2 mode is significant.

in the film and its displacement amplitude decreases sharply on approach to the interface between the film and substrate. Unlike the true SAW whose displacement falls off exponentially with distance from the interface in the substrate, the SHR has a small bulklike component in the substrate. This small bulk wave component carries away the mode energy into the interior of the substrate and causes the mode to decay with distance. Therefore, the mode is a quasilocalized excitation and is expected to have a relatively large linewidth when it is probed by SBS. In the present cases, the bulk wave component of the mode is very small, therefore the modes are observed with fairly narrow linewidth in the experiments.

The fitted elastic constant values for the films give a good account of the positions of the LR and the SHR. However, the calculated Rayleigh SAW frequencies, as shown in Fig. 3, are slightly higher than the observations. Although the discrepancies are small, being within 1 GHz, it may suggest that the observed peak could result from an unknown excitation. The detailed investigations of this excitation and explanations will be reported elsewhere.

In conclusion, we have presented evidence for the existence of a shear horizontal resonance or guided shear horizontal phonon in silicon oxynitrde films on (001) GaAs substrate. The mode is revealed in both experimental observations and theoretical simulations. The properties of the excitation have been investigated by calculating the associated elasto-dynamic Green's tensor that allows the local density of states as well as spatial distributions of the mode displacement to be determined. The study reported here is a valuable step in the classifying of the guided excitations of layer systems. The observation of the LR and SHR also opens up a direct means for investigating the longitudinal and shear sound velocities and hence determining the independent elastic constants C_{11} and C_{44} of films.

We are grateful to T. S. Hickernell for providing the samples, and J. Balogh for technical assistance. This paper is SOEST Contribution No. 4987.

[1] D. L. Mills and K. R. Subbaswamy, in *Progress in Optics XIX*, edited by E. Wolee (North-Holland, Amsterdam, 1981), p. 47.

[2] F. Nizzoli and J. R. Sandercock, in *Dynamical Properties of Solids*, edited by G. K. Horton and A. A. Maradudin (North-Holland, Amsterdam, 1990), p. 281.

[3] P. Mutti, C. E. Bottani, G. Ghislotti, M. Beghi, G. A. D. Briggs, and J. R. Sandercock, in *Advances in Acoustic Microscopy*, edited by G. A. D. Briggs (Plenum Press, New York, 1995), Vol. 1, p. 249.

[4] X. Zhang, J. D. Comins, A. G. Every, P. R. Stoddart, W. Pang, and T. E. Derry, Phys. Rev. B **58**, 13 677 (1998).

[5] V. Bortolani, F. Nizzoli, and G. Santoro, Phys. Rev. Lett. **41**, 39 (1978).

[6] A. M. Marvin, V. Bortolani, F. Nizzoli, and G. Santoro, J. Phys. C **13**, 1607 (1980).

[7] N. E. Glass and A. A. Maradudin, J. Appl. Phys. **54**, 796 (1983).

[8] L. Bassoli, F. Nizzoli, and J. R. Sandercock, Phys. Rev. B **34**, 1296 (1986).

[9] B. Hillebrands, S. Lee, G. I. Stegeman, H. Cheng, and J. E. Potts, Phys. Rev. Lett. **60**, 832 (1988).

[10] G. Carlotti, D. Fioretto, L. Giovanini, F. Nizzoli, G. Socino, and L. Verdini, J. Phys.: Condens. Matter **4**, 257 (1992).

[11] F. Nizzoli, C. Byloos, L. Giovanini, C. E. Bottani, G. Ghislotti, and P. Mutti, Phys. Rev. B **50**, 2027 (1994).

[12] M. Chirita, R. Sooryakumar, H. Xia, O. R. Monteiro, and I. G. Brown, Phys. Rev. B **60**, R5153 (1999).

[13] V. Bortolani, A. M. Marvin, F. Nizzoli, and G. Santoro, J. Phys. C **16**, 1757 (1983).

[14] E. L. Albuquerqu, R. Loudon, and D. R. Tilley, J. Phys. C **13**, 1775 (1980).

[15] A. P. Mayer, J. Phys.: Condens. Matter **1**, 3957 (1989).

[16] R. E. Camley and F. Nizzoli, J. Phys. C **18**, 4795 (1985).

[17] J. A. Bell, R. Zanoni, C. T. Seaton, G. I. Stegeman, W. R. Bennett, and C. M. Falco, Appl. Phys. Lett. **52**, 610 (1988).

[18] G. Ghislotti and C. E. Bottani, Phys. Rev. B **50**, 12 131 (1994).

[19] C. E. Bottani, G. Ghislotti, and P. Mutti, J. Phys.: Condens. Matter **6**, L85 (1994).

[20] G. Carlotti, G. Gubbiotti, F. S. Hichernell, H. M. Liaw, and G. Socino, Thin Solid Films **310**, 34 (1997).

[21] C. H. Whitfield, E. M. Brody, and W. A. Bassett, Rev. Sci. Instrum. **47**, 942 (1976).

[22] J. R. Sandercock, Phys. Rev. Lett. **29**, 1735 (1972).

[23] E. H. Boudouti, B. D. Rouhani, and A. Akjouj, Phys. Rev. B **55**, 4442 (1997).

[24] A. M. Marvin, V. Bortolani, and F. Nizzoli, J. Phys. C **13**, 299 (1980).

[25] G. W. Farnell, in *Physical Acoustics*, edited by W. P. Mason and R. W. Thurston (Academic Press, New York, 1971), Vol. 6, p. 109.

[26] F. S. Hickernell and T. S. Hickernell, IEEE Trans. Ultrason. Ferroelectr. Freq. Control **42**, 410 (1995).