Evidence for the Existence of Guided Longitudinal Acoustic Phonons in ZnSe Films on GaAs

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(Received 5 October 1987)

We report theoretical as well as experimental evidence for a new class of long-wavelength acoustic film excitations which we identify as longitudinal guided acoustic phonons. Despite a coupling to propagating transverse film modes, these new modes have lifetimes comparable to that of the Rayleigh mode and are the longitudinal counterpart to the well-known Lamb or Sezawa modes, with analogous existence criteria. The properties of the new modes are discussed in detail for ZnSe films on GaAs, the material system for which we were able to observe these modes by Brillouin spectroscopy.

PACS numbers: 78.35.+c, 68.55.Pr

In the field of long-wavelength acoustic surface and film excitations, the Rayleigh mode, which always exists on the surface of a semi-infinite material, is the most prominent member.[1] For a film on a substrate, higher-order modes, which are called Lamb or Sezawa modes, exist in the sound-velocity regime bounded by the transverse sound velocities of the film (v_T^f) and the substrate (v_T^s) for the case $v_T^f < v_T^s$.[2] All of these modes consist of shear vertical and longitudinally polarized partial waves whose field components propagate parallel to the surfaces and decay exponentially with distance into the substrate, thus confining the mode energy to the immediate vicinity of the film (or surface for a Rayleigh wave). Within the film, the shear partial waves are propagating in character and the longitudinal partial waves are always evanescent. Recently the longitudinal counterpart to the Rayleigh mode, the so-called longitudinal resonance with a sound velocity slightly below the longitudinal sound velocity and with longitudinal components decaying exponentially into the medium, was predicted theoretically and observed experimentally in gold.[3-5] However, since this mode radiates usually quite strongly into transverse substrate modes, the mode is quasilocalized, i.e., leaky, with a relatively large linewidth when probed by Brillouin spectroscopy. Consequently, it was expected that the longitudinal counterparts of the Lamb (Sezawa) modes, whose velocities are even higher than those of the longitudinal resonances, would have even larger linewidths and be more difficult to observe. In this Letter we report on the successful observation of these modes by Brillouin spectroscopy in epitaxial ZnSe films on GaAs substrates. Contrary to initial expectations, we also found that the lifetime of these modes can be long and the linewidths narrow, comparable to those of the Rayleigh wave itself.

The nature of the longitudinal guided acoustic waves can be easily understood by an examination of the acoustic reflections at the two film boundaries. Since the principal contribution to the total displacement field consists of propagating, longitudinally polarized, partial waves in the film, which are incident and reflected from the film boundaries at grazing angles, mode conversion into the shear-polarized standing waves in the film is weak. For $v_L^f < v < v_L^s$ where v_L^f, v, and v_L^s are respectively the film, guided wave, and substrate longitudinal wave velocities, the longitudinal fields decay evanescently into the substrate and no radiation into substrate longitudinal modes can occur. Furthermore, the grazing-incidence angle condition also ensures very weak radiative coupling into substrate shear modes. These waves can be identified as longitudinal guided acoustic waves because their properties and existence conditions are completely analogous to those of the Lamb modes. With increasing film thickness the sound velocities decrease approaching asymptotically v_L^f. Varying the substrate sound velocity in a computer simulation for a ZnSe film on a (001) GaAs substrate, we also found evidence for higher-order longitudinal guided modes.

The materials system which satisfies the above requirements on acoustic properties consisted of (001) ZnSe films grown on semi-insulating (001) GaAs substrates, as described elsewhere.[6,7] Sample thicknesses were 0.23, 0.53, 1.0, and 2.0 μm. The Brillouin measurements were performed in air at room temperature with a Sandercock-type (3+3)-pass tandem Fabry-Perot interferometer in a backscattering geometry. The light source was a single-mode 514.5-nm Ar^+-ion laser. The laser power at the sample was typically 200 mW. The light scattering geometry was chosen to investigate modes with their acoustic wave-vector component parallel to the surface, $q_{\parallel}$, lying along the [100] and [110] directions of ZnSe. The magnitude of $q_{\parallel}$ was varied by the use of angles of light incidence θ between 40° and 70° to the surface normal. The aperture of the collecting lens was 1:2.8. The incident light was p polarized; no polarization analysis of the scattered light was made.

Figure 1 shows typical spectra for $\theta = 60°$ and $q_{\parallel}$ in the [100] direction. The peak at 7 GHz is identified as scattering from the Rayleigh phonon (labeled "R" in Fig. 1). The peaks between about 8 and 12 GHz are due to the localized Lamb (Sezawa) modes (labeled "S"). A pronounced peak of magnitude comparable to the Rayleigh wave line is observed near 14 GHz in the 2- and the 1-μm films. It is identified as scattering from the longitudinal guided acoustic phonon (labeled "L" in Fig. 1). With decreasing film thickness h, its intensity decreases, accompanied by a linewidth broadening. For $h = 2$ μm, the frequency position of the mode, 14 GHz, is very close to the frequency of a longitudinal acoustic phonon in ZnSe traveling parallel to the film. With decreasing film thickness, the mode frequency shows a slight increase.

We have calculated the Brillouin-scattering cross section using a theory which takes into account both the mechanical and the electromagnetic boundary conditions at the film interfaces including both ripple and elasto-optic coupling mechanisms.[8] The elastic constants of ZnSe were evaluated from (i) the frequency shifts of the peaks associated with the longitudinal guided mode which yield to a good approximation (see below) the sound velocity of a longitudinal bulk phonon traveling parallel to the film, and (ii) the Rayleigh mode in the 2-μm film, all measured in [100] and [110] directions.[9] Their values of $c_{11} = 87.0 \pm 2.1$, $c_{12} = 54.7 \pm 2.1$, and $c_{44} = 39.1 \pm 1.6$ GPa are in good agreement with values reported for bulk ZnSe.[10-15] The photoelastic coefficients and the dielectric constants of ZnSe and GaAs have been taken from the literature.[14,16,17] In Fig. 1 the calculated cross section is shown by smooth curves. Good agreement between the theoretical curves and the experimental data was achieved by our convolving the theoretical curves with the experimental spectrometer function. Only the absolute value of the theoretical cross section has been adjusted. Since the calculation takes all boundary conditions into account, it is not surprising that the new guided longitudinal phonon is reproduced quite well.

Figure 2 shows for the guided longitudinal mode the calculated variation of the squares of the parallel and perpendicular components of the displacement field in the film (negative abscissa) and in the substrate (positive abscissa) normalized with respect to the maximum value of the parallel component in the substrate. They are a measure of the displacement energy. In the film the

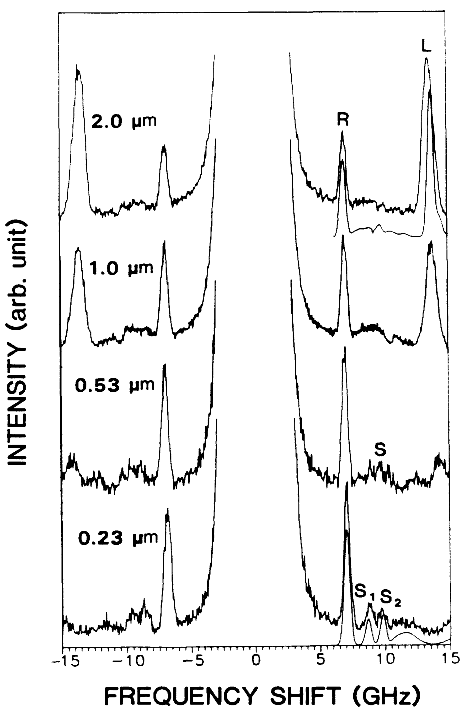

FIG. 1. Brillouin spectra of ZnSe(001) films on GaAs(001) substrate. The angle of incidence is 60°; the film thicknesses are as indicated. R, Rayleigh mode; S, Lamb (Sezawa) modes; L, longitudinal guided mode.

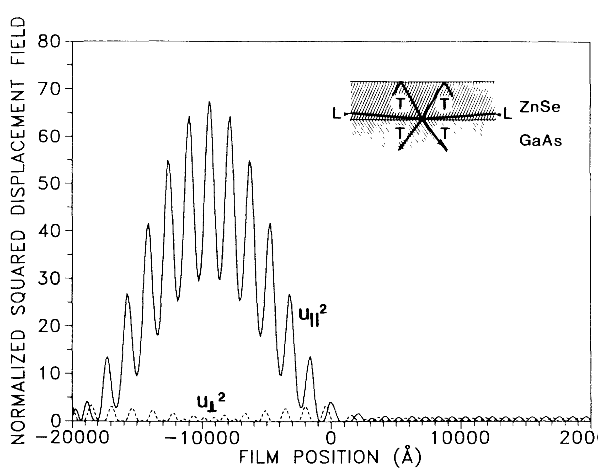

FIG. 2. Modal displacement field profile for a 2-μm ZnSe(001) film on GaAs(001), normalized with respect to the maximum value of the parallel component in the substrate. The negative values of the abscissa refer to the film, whereas the positive values refer to the substrate. The ordinate is the square of the amplitude component parallel to the film (full lines) and perpendicular to the film (broken lines) of the displacement field. Inset: Sketch of the partial waves which constitute the longitudinal guided mode. T, transverse, and L, longitudinal partial waves. Not shown is the exponentially decaying substrate longitudinal partial wave.

displacements are dominantly longitudinal and their squared values exceed those of the substrate by a factor of 70 for $h=2$ μm. The two pairs of partial waves in the film, longitudinal and shear, each form standing wave patterns. The larger periodicity is due to the longitudinal partial waves, whereas the shorter oscillations are due to the transverse components. The inset in Fig. 2 indicates the propagation directions associated with the longitudinal (L) and transverse (T) film and substrate partial waves. The longitudinal partial waves travel nearly parallel to the film whereas the transverse ones have a larger wave-vector component perpendicular to the film.

As an existence criterion for guided waves, it is necessary that the partial waves interfere constructively. Thus the wave-vector component perpendicular to the film, $q_\perp$, must obey the resonance condition

$$2q_\perp h+\delta_S+\delta_I=2n\pi, \tag{1}$$

where δ_S and δ_I are the phase shifts upon reflection at the two film surfaces, respectively, and the mode index n is a positive integer. A calculation of the phase shifts with a simplified model[18] (i.e., elastically isotropic medium) shows that Eq. (1) is fulfilled very accurately for the longitudinal partial waves with $n=1$. Equation (1) also predicts several additional properties of the mode. Because the elastic properties of the media bounding the film are dissimilar, there is a minimum film thickness below which Eq. (1) cannot be satisfied. With increasing film thickness the value of $q_\perp$ decreases in order to fulfill Eq. (1), thus decreasing the modal sound velocity. A decreasing value of $q_\perp$ implies a decreasing amount of mode conversion into shear waves, resulting in increasing localization, as experimentally observed (see Fig. 1). In the limit of infinitely thick films, $q_\perp$ approaches zero, which implies that the guided longitudinal acoustic phonon becomes an ordinary longitudinal bulk phonon traveling parallel to the film. It should be stressed that, for a film of finite thickness, no longitudinal bulk phonon traveling parallel to the film may exist because the mechanical boundary conditions at the interfaces cannot be satisfied.

Figure 3 shows results of computer simulations of the maximum of the square of the parallel displacement-field amplitude of the longitudinal guided mode, normalized as in Fig. 2, as a function of the film thickness. With increasing film thickness the sound velocity shows an overall decrease with small superimposed oscillations. The intensity of the mode increases, accompanied by strong aperiodic oscillations which exhibit monotonically increasing maxima with intervening regions of smaller maxima, accompanied by corresponding oscillations in the linewidth. The oscillatory behavior is due to the mode conversion at each reflection of transverse into longitudinal partial waves and vice versa. The transverse waves can also fulfill a resonance condition similar to Eq. (1), however with a large n which also increases with increasing film thickness. Since the total transmission of energy across the interface is proportional to the transmission coefficient at the interface and the displacement amplitude of the transverse film partial waves, a film resonance of the transverse modes implies a maximum in the energy loss into the substrate. Since the transverse-mode resonance condition is periodically fulfilled with increasing film thickness, the guided-mode attenuation also oscillates, accompanied by a corresponding oscillation in the linewidth. The two different types of maxima can be related to the odd or even value of n: A calculation in the isotropic approximation[18] of the phase factors for the reflection of the transverse film modes at the surface ($\delta_s=-\pi/2$) and at the interface ($\delta_i\cong 0.2$) yields a major maximum in the displacement field for even n and a secondary maximum for odd n.

In summary, we have found a longitudinal guided acoustic mode in ZnSe(001) films on GaAs(001). Its calculated linewidth is unusually small, typically 0.5% of its mode frequency. Despite the decay channel into transverse substrate modes, the new mode is the longitudinal counterpart to the (transverse) Lamb modes in a film. Thus the classification of long-wavelength acoustic surface and film excitations now exhibits complete symmetry with respect to shear and longitudinal waves, and includes surface modes (Rayleigh mode, longitudinal

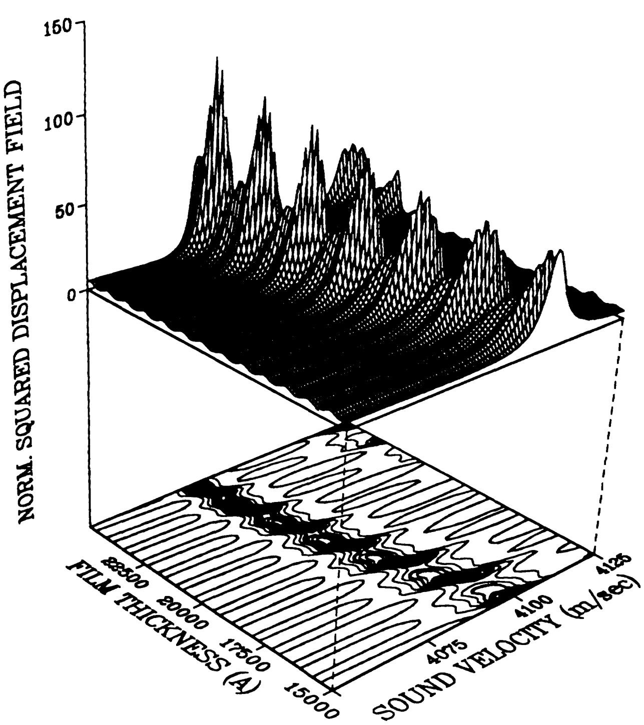

FIG. 3. Surface plot and contour plot of the square of the parallel displacement field as a function of the film thickness h and the sound velocity v.

resonance) and film modes (Lamb modes, guided longitudinal modes). The new longitudinal guided wave reported here offers a number of interesting possibilities. For example, it allows a very accurate determination of the longitudinal sound velocity in films, a parameter which is otherwise difficult to evaluate. Here we actually utilized this feature to evaluate the elastic constants of the ZnSe film. Since the displacement field is nearly completely polarized parallel to the film, the mode can be used to monitor effects which modulate only dilatational waves, and vice versa. The observation and application of the novel mode in systems other than ZnSe/GaAs is presumably only restricted by the existence criteria on the acoustic velocities described above.

This work was supported by the U. S. Air Force Office of Scientific Research–University Research Initiative Program under Contract No. F 49620-86-C-0123, and by the Defense Advanced Research Project Agency under U. S. Office of Naval Research Contract No. N 00014-85-C-0552. One of us (F.N.) acknowledges a travel grant from Consiglio Nazionale Richerche of Italy.

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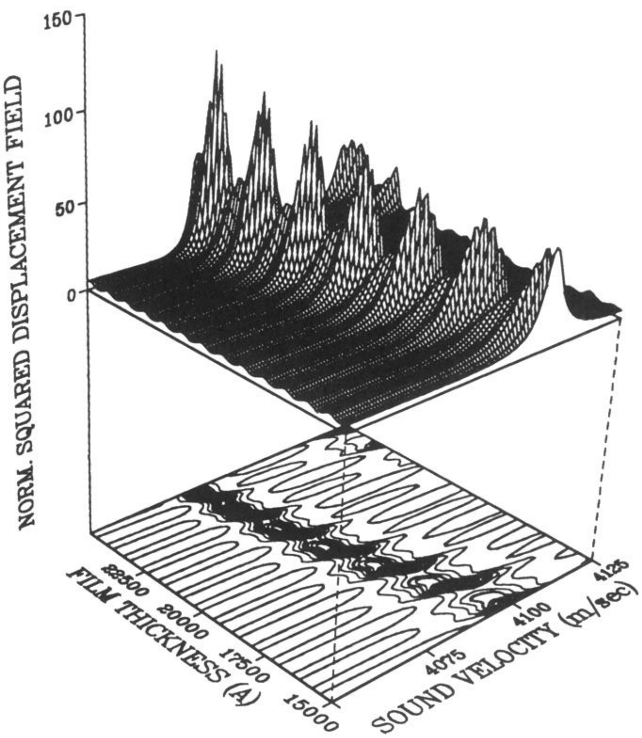

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