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Problems with the determination of elastic constants from higher-order surface waves: Results for Al on NaCl

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(Received 15 June 1987)

Using Brillouin scattering we have studied the surfacelike generalized Lamb modes in Al films on NaCl. Using a theory in which the elastic constants (C_{ij}) of Al are taken from bulk values, reasonably good agreement is found for the positions of the peaks. However, we were unable to fit our experimental results to obtain the C_{ij} independently, since different combinations of the C_{ij} lead to similar spectra and dispersion curves. The changes in the spectra brought about by varying each one of the C_{ij} are investigated theoretically, but in no case could the experimental linewidths be reproduced. The fact that the experimental linewidths are much narrower than the calculated ones is very hard to explain unless it is assumed that the Al films are not completely bonded to the substrate. Calculations on unsupported films support this explanation. Our results clearly show the difficulty in obtaining information on elastic constants from an analysis of generalized Lamb modes of a film on a substrate and illustrate some of the problems which can be encountered.

INTRODUCTION

The determination of the elastic properties of materials that are in the form of thin films poses severe experimental problems, since conventional ultrasonic techniques cannot be used when the film thickness becomes comparable to or less than the wavelengths attainable in these experiments. In spite of the effort that has gone into developing special techniques for measuring thin films, it is not yet possible to reliably determine all the elastic constants of a material which can only be prepared in the form of a thin film. A technique which has proved to be particularly well adapted to the study of thin opaque films is Brillouin scattering. The development of the multipassed and tandem Fabry-Perot interferometer¹ has made it possible to observe scattering from surface (Rayleigh) waves as well as from a continuum of bulk excitations.¹ The sharp feature associated with Rayleigh waves makes it convenient to obtain information on a shear elastic constant related to a bulk transverse wave polarized normal to the free surface. The method has been used quite extensively in the investigation of shear elastic properties of superlattices² and metallic glasses.³ Extracting information from the broad features arising from bulk excitations is considerably harder and has only been used to obtain information on longitudinal elastic properties when elastic isotropy can be assumed.³

Apart from the sharp feature associated with the Ray-

leigh wave, additional sharp features appear at higher frequencies in Brillouin scattering experiments from supported films thinner than $\sim 3000 \text{ \AA}$.^{1,4-12} These sharp features can be shown to be due to generalized Lamb modes which are related to the Lamb modes of a free-standing plate. The frequencies of these additional modes, as well as that of the Rayleigh mode, are dependent on a number of parameters, viz., the elastic properties of the film as well as those of the substrate, the thickness of the film (d), and the densities of the film and of the substrate. Experiments performed to date^{1,4-7} find that for a system in which all these parameters are known, the peak positions and intensities are well accounted for by theory.

In a number of recent articles⁸⁻¹² the positions of the generalized Lamb modes have been analyzed to yield information on the elastic constants. However, it was realized in these articles that all C_{ij} could not be fitted simultaneously; a number of the C_{ij} had to be taken as input parameters. In this paper we describe a procedure to obtain the elastic constants from the observed spectra, in what was expected to be a very simple case. We have chosen to study Al films deposited on the (100) surface of NaCl. When deposited at room temperature on NaCl, Al grows with preferential orientation along the [111] direction but with random orientation in the plane. Our Al films therefore have hexagonal symmetry and their elastic constants can be estimated reasonably well from those of single-crystal Al. (This is particularly true

for Al, since in the bulk form it is almost isotropic.) The elastic constants of the substrate are, of course, well known.

THEORY AND EXPERIMENT

The Brillouin experiments were carried out using ~ 200 mW of 514.4-nm radiation from a single-mode Ar-ion laser. The incident beam, polarized in the scattering plane, was collinear with the scattered beam and they subtended an angle θ with the surface normal. The angle θ was changed in different experiments but was always in the range of 35° – 60° . The focused laser spot on the sample was typically ~ 75 μm in diameter. We were not able to measure the temperature of the sample in the scattering volume; however, since most of the incident light is reflected, and since the thermal conductivity of Al is very high, we estimate that the temperature in the scattering volume is not likely to be more than a few tens of degrees centigrade above room temperature. The scattered light was analyzed using a 5 + 4-pass tandem Fabry-Perot interferometer.

Figures 1(a) and 1(b) show the experimental spectra obtained for films with thickness of 1400 and 2000 Å and a component of the wave vector parallel to the sur-

face, $q_{\parallel} = 2.12 \times 10^5$ and 1.87×10^5 cm^{-1} , respectively. The peaks labeled with an asterisk are instrumental and are due to a filter which attenuates the intensity of the unshifted radiation. In a strict sense the features in Fig. 1 are not really localized modes, since their frequencies lie within the continuum of modes of the NaCl substrate, and they should be regarded as resonances within this continuum.¹³ This distinction implies that slightly different techniques must be used for their calculation; these techniques have been described in Refs. 7 and 13. The results of such a calculation (which includes not only the frequency of the modes but also the strength of the coupling to the incident radiation) are presented in Fig. 2. The spectra in this figure are calculated for a film 2000 Å thick and $q_{\parallel} = 1.87 \times 10^5$ cm^{-1} , in order to reproduce the experimental conditions used in obtaining Fig. 1(b). The upper and lower curves in Fig. 2 represent spectra for two different sets of elastic constants which will be described in detail later. The most striking difference between the experimental results for the 2000-Å film in Figs. 1(b) and the calculated spectra of Fig. 2 is the width of the Brillouin peaks. This is of particular concern since the calculated spectra contain essentially no broadening and the linewidths are determined only by the resonance with the substrate. As will be discussed later, we believe that this is evidence that the films are not firmly bonded to the substrate.

Ignoring, for the moment, the problem of linewidths,

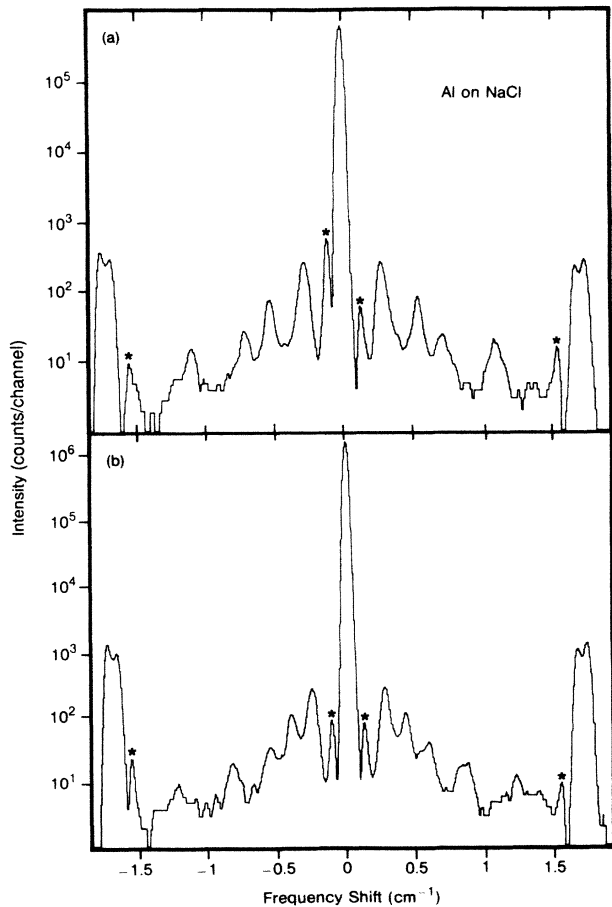


FIG. 1. Brillouin spectra of polycrystalline Al on NaCl substrates. (a) Film thickness 1400 Å, $q_{\parallel} = 2.12 \times 10^5$ cm^{-1} , and (b) film thickness 2000 Å, $q_{\parallel} = 1.87 \times 10^5$ cm^{-1} . The peaks labeled with asterisks are of instrumental origin.

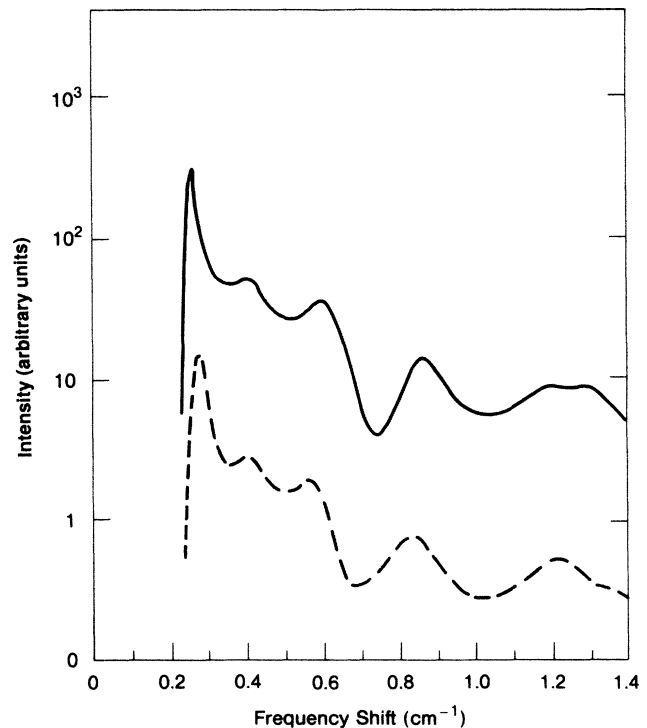


FIG. 2. Computed spectra for a 2000-Å film of Al with $q_{\parallel} = 1.87 \times 10^5$ cm^{-1} . The elastic constants for the upper spectrum (solid line) are $C_{11} = 11.3$, $C_{13} = 5.8$, $C_{33} = 11.5$, and $C_{44} = 2.5$; and those for the lower spectrum (dashed line) are $C_{11} = 14.0$, $C_{13} = 6.6$, $C_{33} = 11.1$, and $C_{44} = 2.0$. Although the elastic constants changed by as much as $\sim 20\%$, the spectra are very similar.

the velocity associated with each peak can be calculated from spectra like those in Figs. 1 and 2. In Fig. 3 these velocities are plotted as a function of $q_{\parallel}d$ where d is the film thickness. The crosses are experimental values. The solid and dashed lines are from calculated spectra like those in Fig. 2. The solid lines (Figs. 2 and 3) refer to values from the literature for the elastic constants of a preferentially oriented film, viz., $C_{11}=C_{22}=11.3$, $C_{33}=11.5$, $C_{44}=C_{55}=2.5$, $C_{66}=2.6$, $C_{13}=C_{23}=5.8$, and $C_{12}=6.0$ in units of 10^{11} dyn/cm². The dashed lines (Figs. 2 and 3) are calculated using elastic constants which were chosen in an attempt to produce a better fit, viz., $C_{11}=14.0$, $C_{33}=11.1$, $C_{44}=2.0$, and $C_{13}=6.6$ in units of 10^{11} dyn/cm². We mention that the spectra depend only on C_{11} , C_{33} , C_{44} , and C_{13} . The constants of the NaCl substrate C_{11} , C_{12} , and C_{44} were taken as 4.91, 1.28, and 1.28×10^{11} dyn/cm² respectively, and the densities of Al and NaCl as 2.702 and 2.165 g/cm³, respectively. In the calculations, only the ripple mechanism of coupling to the light has been taken into account, since for Al the elasto-optic contribution can be shown to be negligible.^{5,6} In Fig. 3 the absence of a data point for some of the calculated spectra indicates that the features were so broad that they could not be assigned with any certainty. It can be seen that the calculations with both sets of C_{ij} describe the experimental results reasonably well. However, since the elastic constants used to produce the fits differ by as much as 20% it is clear that it is not possible to obtain all the elastic constants with any reasonable accuracy. This is consistent with the previous results of Refs. 8–12.

It should not be concluded from the above comments that the peak positions do not depend on the individual elastic constants. In Fig. 4 we show the variations produced by a change in only one of the pertinent elastic constants. The solid line in each case is the calculated spectrum for a 2000-Å film, $q_{\parallel}=2.12 \times 10^5$ cm⁻¹, and values from the literature for the C_{ij} . The dashed line is the spectrum obtained by decreasing the indicated C_{ij} by

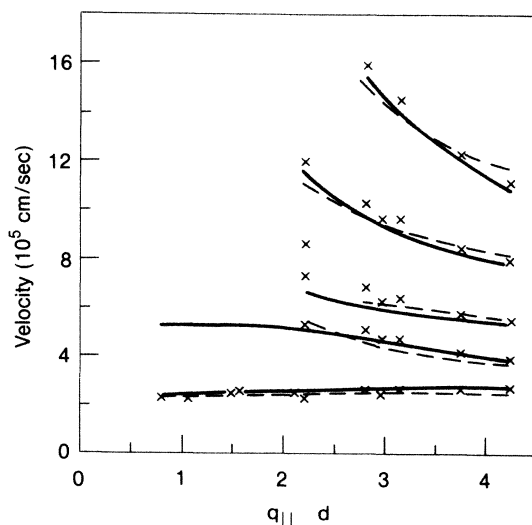


FIG. 3. Velocity of sound as a function of $q_{\parallel}d$. Crosses are the experimental values; the solid and dashed lines are calculated using the elastic constants as in Fig. 2.

20%. By comparing calculated spectra like those shown in Fig. 4 with experimental results like those in Fig. 1, we attempted to produce better agreement between theory and experiment. After iterative changes in all pertinent elastic constants a "best fit" was obtained for a specific set of C_{ij} : these are listed in the caption of Fig. 2. The "best fit" was judged by eye since the theoretical spectra contain broad features which make a computerized least-squares fit very difficult.

A rather surprising feature in the experimental spectra is the sudden disappearance of higher-order peaks between $q_{\parallel}d=2.1$ and 2.2, as evidenced by the crosses in Fig. 3. The spectra at which this sudden change takes place are shown in Figs. 5(b) and 5(e) and correspond to two different samples with $d=1000$ Å, $q_{\parallel}=2.12 \times 10^5$ cm⁻¹ and $d=1400$ Å, $q_{\parallel}=1.57 \times 10^5$ cm⁻¹, respectively. Figures 5(a) and 5(d) are the calculated spectra for the above films on NaCl while 5(c) and 5(f) are for corresponding unsupported films. From the similarity of Figs. 5(a) and 5(b) and that of 5(e) and 5(f), we conclude that films thicker than 1400 Å are no longer firmly bonded to the substrate. This idea is supported by the previously mentioned fact that the observed linewidths in films with $d \geq 1400$ Å are narrower than those calculated for supported films and by the fact that the frequency of the lowest mode in a 1400-Å film is lower than that in the 1000-Å film. This conclusion makes the

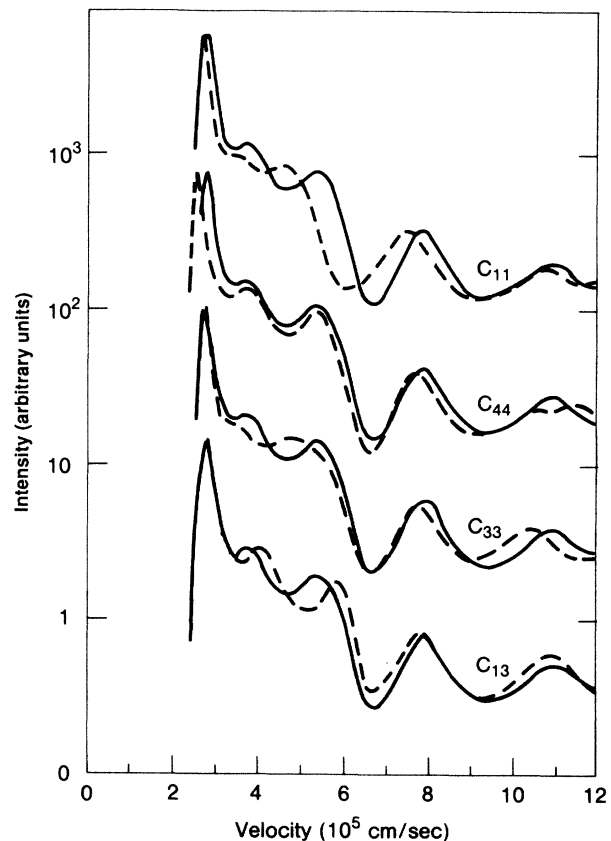


FIG. 4. Spectra computed for a 2000-Å Al film with $q_{\parallel}=2.12 \times 10^5$ cm⁻¹. The solid line is for spectra computed with values from the literature. The dashed lines are spectra computed by decreasing the indicated elastic constant by 20%.

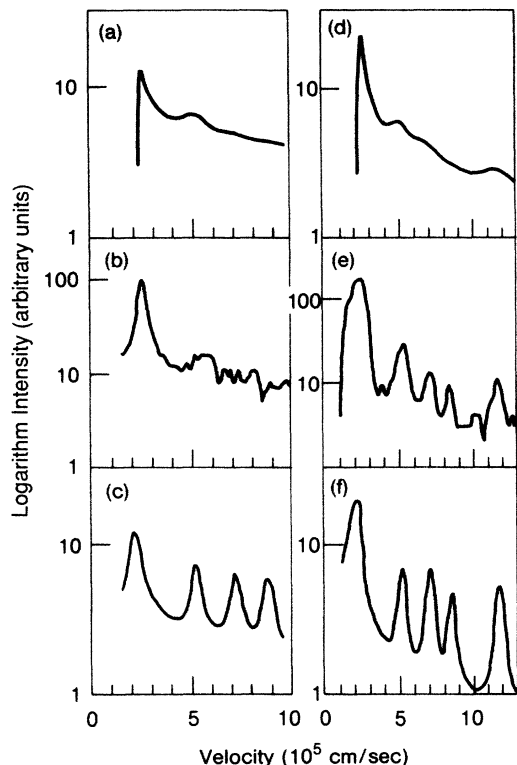


FIG. 5. (a), (b), and (c) are spectra for 1000-Å films and $q_{\parallel} = 2.12 \times 10^5 \text{ cm}^{-1}$. (d), (e), and (f) are for 1400-Å films and $q_{\parallel} = 1.57 \times 10^5 \text{ cm}^{-1}$. (a) and (d) are calculated for supported films. (b) and (e) are experimental spectra. (c) and (f) are calculated for unsupported films.

already uncertain determination of elastic constants even more open to possible errors. We point out that if our Al samples had been deposited on a harder substrate so that the higher-order modes were not within the continuum of bulk modes, we would not have been able to detect the “nonbondedness” of our films, because sharp peaks would have been observed for both bonded and nonbonded films and the only difference would have been a small difference in peak position which could al-

ways be attributed to experimental errors.

In a recent Letter¹⁴ Brillouin scattering from free-standing films with thicknesses down to 200 Å were presented. It was shown that for Au films on NaCl (the higher-order modes in Au lie below the continuum) the spectra from supported and unsupported films with $d \gtrsim 1000 \text{ Å}$ are quite similar, differing mainly in small changes in the peak positions.

CONCLUSIONS

By attempting to fit the positions of higher-order surface modes in Al films on NaCl, we have shown that different combinations of elastic constants can produce similar results. Consequently, we conclude that a fit of these mode positions to obtain the elastic constants is not reliable. However, if some of the C_{ij} 's could be determined by another technique, this method may prove useful to determine the remaining ones. It should also be noted that even if a quantitative determination of the C_{ij} 's is not possible, a comparison of spectra of two different samples may yield some information as to whether changes exist in the C_{ij} 's.

We also provide evidence that in some cases the films may not be firmly bonded to the substrate. This feature, although important, may not always be easily detectable but it can lead to serious errors in any type of fitting procedure. Conversely, in those cases where it can be detected (viz., shear velocity in films greater than in substrate), Brillouin scattering may prove to be a convenient, nondestructive technique to provide information on the bond between film and substrate.

ACKNOWLEDGMENTS

This work was supported by the U.S. Department of Energy (Basic Energy Sciences Program, Division of Materials Sciences), under Contract No. W-31-109-ENG-38 and by North Atlantic Treaty Organization Travel Grant No. RG85/0473. One of the authors (O.F.L.) acknowledges financial support from the Conselho Nacional de Pesquisas of Brazil.

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