Brillouin scattering from unsupported Al films

R. Bhadra and M. Grimsditch
Materials Science Division, Building 223, Argonne National Laboratory, Argonne, Illinois 60439

Ivan K. Schuller
Physics Department-B019, University of California–San Diego, La Jolla, California 90293

F. Nizzoli
Dipartimento di Matematica e Fisica, Universita di Camerino, I-62032 Camerino, Italy
and Centro Interuniversitario, Struttura della Materia del Ministero Pubblica Istruzione, I-62032 Camerino, Italy
(Received 9 November 1988)

Brillouin scattering measurements on unsupported Al films have been performed on films with thicknesses down to ~23 nm. Both the line positions and relative intensities of the Lamb modes observed in the experimental spectra are well accounted for by theory with no adjustable parameters. Fits to the experimental dispersion curves in unsupported films are found to give somewhat more reliable values for elastic constants than fits to results on supported films.

I. INTRODUCTION

The determination of elastic properties of thin films (<1 μm) poses severe experimental problems because they are considerably thinner than the shortest wavelengths that can be achieved in conventional ultrasonic experiments. As a consequence, other more elaborate techniques must be employed, the results of which are often controversial. It is safe to say that at present there is no reliable way of determining all the components of the elastic constant tensor for a material that can only be prepared as a thin film. A possible exception could be the case of transparent films where, using Brillouin scattering, both surface and bulk modes can be measured at the same time, resulting in a very rich amount of information. Metal films, however, do not meet these requirements.

One technique which has proved to be useful in the study of the elastic properties of thin films is Brillouin scattering.[1] This technique, which in opaque materials usually provides information on only one surface wave (and hence only one particular combination of elastic constants), has the potential of yielding much more information: in films considerably thinner than 1 μm it is possible to observe standing waves (generalized surface waves) between the free surface and the film-substrate interface.[2] The frequency of these modes depend on different combinations of elastic constants (C_{ij}) of the film and the substrate. In principle therefore, by fitting to the experimental results it should be possible to obtain numerous C_{ij}'s of the film.

In a recent paper we addressed the problems involved in determining elastic constants of supported thin metallic films from fits to the dispersion curves of surface waves (Rayleigh, Sezawa, and generalized Lamb waves) determined by Brillouin scattering.[3] There we reported that the errors involved in such fitting procedures are large, and that they are partly due to the presence of a substrate and uncertainties in the bonding between the two materials. Recent measurements, which have shown that measurements on unsupported films[4] are also possible, have led us to attempt fitting to the dispersion curves of Lamb waves in unsupported films in order to ascertain if the errors in the determination of the C_{ij} can be reduced.

The previous results on unsupported films were made on Au;[4] the elastic constants obtained from a fit to the dispersion curves yielded reasonable values for the C_{ij}. Unfortunately since Au has a large elastic anisotropy it was not possible to decide if the differences between fitted and literature values were due to errors or simply due to texture and/or preferential orientation of the films.

We have chosen to study Al films because since Al is almost elastically isotropic, the results should not depend strongly on growth characteristics. Films with thicknesses down to ~23 nm were studied using Brillouin scattering and the dispersion curves for the five lowest-lying Lamb modes determined. The Brillouin spectra are satisfactorily reproduced within the experimental errors, in both peak positions and intensities using a generalization of the theory described in Ref. 5 and the literature elastic constants for Al. The experimental results are also fit to obtain the C_{ij} independently and the errors involved in such fits are described in detail.

II. THEORY

The normal modes of vibration of a free-standing plate have been extensively described in the literature.[6] For isotropic and other high-symmetry materials, there are two types of modes: those polarized in the plane of the film and perpendicular to the propagation direction (Love waves) and those with a polarization in the plane containing the propagation direction and the film normal (Lamb waves). Rather than use the approach outlined in Ref. 6,

which yields analytical solutions for simple cases, we have used a generalization of the theory as it is usually used for surface waves[7] and modes in supported films.[8] This approach only yields numerical solutions but can be used for films of any symmetry. We mention that the analytical approach has also been applied to calculations of the cross section in unsupported films, but only for films which are isotropic.[9]

The coupling of light to acoustic phonons in highly reflecting materials is known to be dominated by the ripple mechanism, which corresponds to a corrugation of the sample surface due to the phonon which acts as a moving grating for the incident radiation.[10] The scattered intensity in such a process is proportional to the square of the normal component of the phonon amplitude at the surface, and hence can be easily calculated once the normal modes of a film are known. It is also clear that the Love modes, since they do not corrugate the surface, do not couple to the light via the ripple mechanism.

The calculations reported here for Brillouin peak positions and intensities are similar to those reported in Refs. 2, 3, and 5 except that the boundary conditions at the back surface are appropriate for a solid-vacuum interface. The intensity calculations for Al are based only on the ripple mechanism since the elasto-optic contributions in this material are known to be negligible.[11]

III. EXPERIMENT AND RESULTS

Al films were evaporated onto freshly cleaved NaCl substrates using feedback-controlled electron-beam guns at typical pressures better than 5×10^{-7} torr during evaporation and typical rates of 10 Å/sec. After removal from the vacuum chamber the films were placed in contact with a lightly greased grid and the NaCl dissolved in H_2O. Even for the thinnest films ($\sim$23 nm) unsupported areas as large as $\sim$1 mm in diameter could be produced in this fashion. Since the phonon wavelengths investigated with Brillouin scattering are $\sim$300 mm, these films can be treated as extending to infinity in the plane of the sample.

Film thicknesses were determined using an *in situ* quartz-crystal oscillator. Apart from small (few percent) errors in thickness which possibly arise from minor changes in sample position, background pressure, amount of material in the crucible, etc., a more severe consideration is the accuracy of calibration of the crystal monitor itself. Calibration of the quartz-crystal oscillator against a thickness profilometer; together with the uncertainties in "exact" deposition conditions imply that the error in thickness is of the order of $\sim$5%. Characterization of the films with x rays showed that they grew with strong preferential orientation with the [111] axis normal to the surface. Within experimental accuracy ($\sim$0.5%) no change was observed in the position of the [111] peak in films with different thicknesses.

The scattering geometry used in the experiments is shown in Fig. 1. The incident light was polarized in the scattering plane and no analyzer was placed in the scattered beam. Powers as low as 10 mW of 515 nm radiation were used on the thinnest films in order to avoid

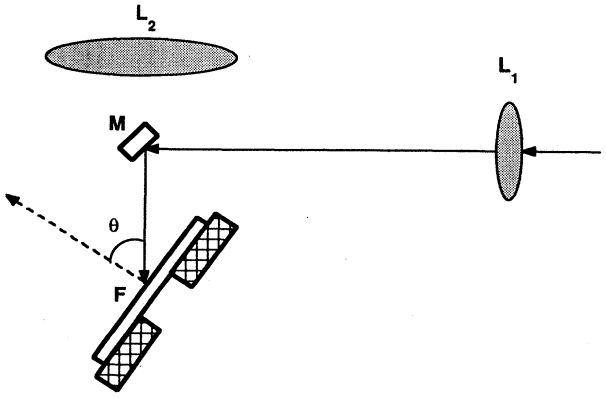

FIG. 1. Scattering geometry used for Brillouin experiments on unsupported films. L_1, laser focusing lens; M, mirror; F, film; θ, angle between radiation and surface normal; and L_2, collection lens.

burning them. The angle θ between the film normal and the incident beam was chosen in the range from 40° to 60° in different runs. The wave vector of the phonon probed is given by

$$q=\frac{4\pi}{\lambda_L}\sin\theta\ , \tag{1}$$

where λ_L is the wavelength of the incident radiation. The spectra were recorded on a tandem (5+2 pass) Fabry-Perot interferometer. There is good agreement in both line positions and intensities between experimental (Fig. 2, upper half) and calculated (Fig. 2, lower half)

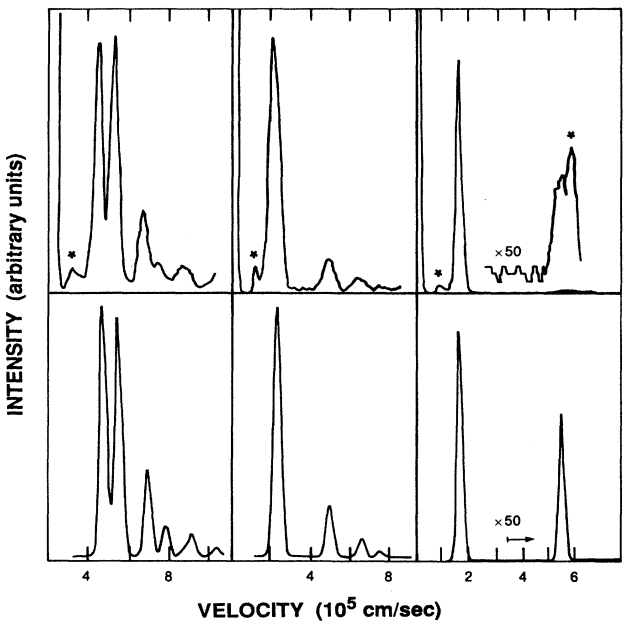

FIG. 2. Brillouin spectra from unsupported Al films. The upper half are experimental spectra the lower half are calculated. From left to right the spectra correspond to film thicknesses of 255, 125, and 60 nm, respectively; the angle θ was kept at 60°.

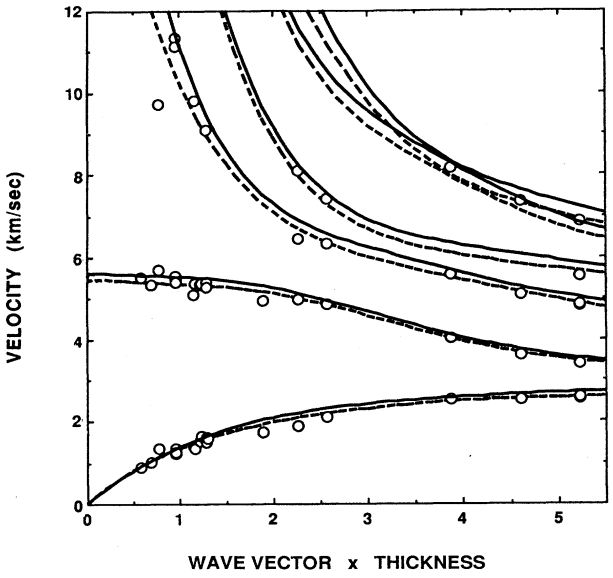

FIG. 3. Dispersion curves for the six lowest modes in unsupported Al films. The dots are experimental points, the solid lines are calculated using literature values for the C_{ij}, and the dashed lines are a fit obtained using the C_{ij} as fitting parameters.

spectra obtained using the methods outlined above. The calculated spectra were performed using the hexagonal elastic constants for a (111) preferentially oriented Al film viz., $C_{11}=113$, $C_{13}=57.9$, $C_{33}=115$, and $C_{44}=25.1$ GPa. These values were calculated from single crystal values[12] ($C_{11}=108, C_{12}=61.3$, and $C_{44}=28.5$ GPa) using the expressions for the Voigt average from Ref. 13 (the Reuss average gives values $\sim 2\%$ lower). The density was taken to be 2.69 g/cm^3. The linewidth in the calculations was chosen to reproduce the experimental resolution. Because of the strong absorption of light in the film, for the thicknesses considered in Fig. 2 the ripple contribution from the back surface turned out to be completely negligible, so that only the ripple mechanism from the front surface is relevant. We have calculated that interference effects between the light scattered from the two interfaces is appreciable only in very thin films ($d \lesssim 15$ nm). The peaks labeled with an asterisk in the experimental spectra in Fig. 2 are of instrumental origin.

The dots in Fig. 3 are the experimentally determined velocities of the Lamb modes measured on various films with thicknesses d, and at various angles [q given by Eq. (1)]. The solid lines are the dispersion curves calculated with the same literature values for the C_{ij} as used to calculate the spectra in Fig. 2. The agreement between theory and experiment is good especially considering that there is a possible systematic error in the experimental d of up to $\sim 5\%$. In the calculations we find that the fifth- and sixth-order modes exhibit a tendency to cross at $qh \simeq 3.8$. Although this is actually forbidden by symmetry, the result is a small gap in the dispersion curves, not well resolved in Fig. 3. Since the calculated intensity of the fifth-order mode is considerably smaller than that of the sixth mode in the region around $qd \approx 4$ we assign the experimental point $v \cong 7$ km/sec at $qd=5$ to the sixth-order mode. The theory is therefore capable of explaining in great detail the experimental results—mode frequencies, relative intensities, and dispersion curves.

TABLE I. Elastic constants of Al films (GPa) obtained from fits to Brillouin data. An asterisk indicates a nonfitted parameter, the last row contains literature values.

C_{11}	C_{13}	C_{33}	C_{44}	Standard deviation
104	52.7	108	22.9	16
114*	59.5	110	22.3	22
107	57.7*	116	22.9	18
104	55.7	118*	23.0	17
104	52.7	108	24.9*	23
113*	57.9*	115*	25.1*	44

We return now to the inverse problem, i.e., the determination of film elastic constants from the experimental results. A least-squares fit to the data with C_{11}, C_{13}, C_{33}, and C_{44} as adjustable parameters, gives $C_{11}=104$, $C_{13}=52.6$, $C_{33}=109$, and $C_{44}=22.9$ GPa; the associated dispersion curves are plotted as dashed lines in Fig. 3. Note that the best fit values are systematically 7—10% smaller than the literature C_{ij}. The assignment of errors to the C_{ij} determined from the least-squares fit is a nontrivial problem. Ignoring for the time being any errors arising from the thickness determination, we performed the following calculations: starting with the values from the least-squares fit we increase one of them by 10% and then perform a fit using the other three as variables. The results are summarized in Table I, where an asterisk denotes a value which is not fitted; the last row in the table gives the literature values of the C_{ij}. In all the cases considered, the change in the standard deviation is small and the differences between the dispersion relations are certainly smaller than the experimental accuracy. We therefore conclude that even in the absence of errors in the measured thicknesses, the absolute C_{ij} obtained from the Brillouin results are at best only accurate to $\sim 10\%$. Possible systematic errors in thickness would increase this uncertainty.

In summary, we find that a theory without fitting parameters which uses literature values for the elastic constants is capable of reproducing quantitatively peak positions and intensities observed in Brillouin spectra from unsupported films. The intensities of the Brillouin peaks from Al films are well reproduced taking into account only the ripple mechanism. The determination of elastic constants from experimental data can only be performed with an accuracy of $\sim 10\%$.

ACKNOWLEDGMENTS

We would like to thank B. Hillebrands for the use of his fitting program. This work was supported by the U. S. Department of Energy, Basic Energy Sciences–Materials Sciences, under Contract No. W-31-109-ENG-38, the Office of Naval Research Contract No. N00014-88K-0210, and foreign travel was provided by a North Atlantic Treaty Organization Travel Grant No. RG85/0473.

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