Biaxial elastic modulus of metallic films determined from vibrating circular membranes

A. Fartash
Physics Department, B-019, University of California, San Diego, La Jolla, California 92093, and Materials Science Division, Argonne National Laboratory, Argonne, Illinois 60439
Ivan K. Schuller
Physics Department, B-019, University of California, San Diego, La Jolla, California 92093
M. Grimsditch
Materials Science Division, Argonne National Laboratory, Argonne, Illinois 60439

(Received 3 August 1989; accepted for publication 8 October 1989)

The biaxial moduli of free-standing metallic films have been determined from their behavior as elastically tensioned membranes. Errors due to thickness determination and warping of the films are eliminated, thereby opening up the possibility of solving a number of controversial issues. The method combines the determination of stress from the frequency of the membrane modes with optical measurements of strain. Tests on Ta and Ni films yield values of the biaxial modulus in good agreement with calculated literature data.

We report measurements of the biaxial elastic modulus of metallic films by employing a novel method which is independent of film thickness and which eliminates warping of the films; possible errors in results obtained by other techniques are thereby avoided. Because thin films are used extensively in basic research and industry, the determination of their physical properties as a function of fabrication parameters[1] is important. However, because they are thin films, many of their mechanical properties cannot be measured with conventional techniques. To study films whose thickness is comparable to the shortest wavelengths attainable in ultrasonic experiments ($\sim 20\ \mu m$) it has been necessary to develop specialized techniques. A good example of the problems encountered in the determination of elastic constants (C_{ij}) of thin films is the study of the supermodulus effect in superlattices.[2] The huge enhancements in the biaxial modulus (Y_B) found using a bulge tester[3-5] have been questioned regarding the data analysis[6]: vibrating-reed studies have led to conflicting results[7-9] while Brillouin scattering[10] and transient thermoreflectance[11] have typically shown decreases in elastic moduli. It is clear from the above situation that the determination of elastic properties of thin films requires more reliable techniques.

The experimental technique employed here involves in part the determination of the resonant frequencies of a stretched circular membrane. The general solution for the resonant frequencies of such a system depends on the elastic properties of the film as well as on the applied radial tension. As will be discussed below, the dominant term in the case of thin films is the applied tension and in most cases the elastic contribution is negligible. Under these conditions the resonant frequency is a direct measure of the tension on the film; by measuring the changes in the resonant frequency as the film is stretched and by optically measuring the strain in the film, the biaxial elastic modulus is obtained. A schematic view of our experimental apparatus, which will be described in detail elsewhere,[12] is shown in Fig. 1. The body consists of three main pieces (A, B, and C) machined from brass which form an assembly that serves to rigidly clamp and stretch the film. Piece B clamps the film (F) over a knife edge on C that defines a circular boundary with a diameter of 3/8". The tension in the film is controlled by the screws that hold A in place. The coil M generates an oscillating field that causes the film to vibrate.[12] As the frequency of the current in the coil is varied, the induced vibrational resonances are capacitively detected by a shielded electrode (E) facing the film. In general it is best to operate the jig in vacuum ($< 10^{-4}$ Torr) to minimize the effects of hydrodynamic damping by air.[13] Besides air, there is another source of damping due to mechanical losses at the clamping boundary. For the apparatus shown in Fig. 1, these losses are very small, and result in minimal damping. The quality factors that were measured in vacuum ($< 10^{-4}$ Torr) were as high as $\sim 4\times 10^3$. The strain (ϵ) was measured optically by viewing the film through a high magnification microscope. The rectangular reticule (G) positioned above the film allowed particular markings

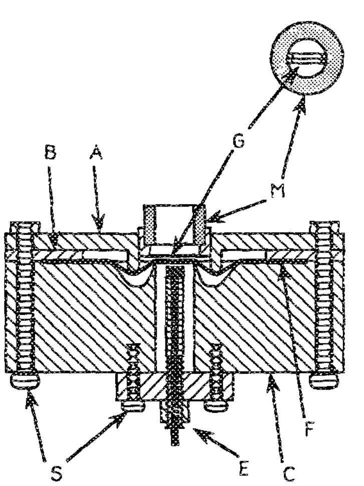

FIG. 1. High Q resonator for measurement of biaxial properties of metallic films. The film is viewed from above through a microscope. The hairlines on the reticule, shown at the upper right-hand corner of the figure, serve to locate and measure displacements of designated points for determination of strain.

to be tracked as a function of tension with a resolution of about 0.5 μm. The ratio of radial stress (tension) to radial strain yields the biaxial modulus (Y_B).

The normal modes of a film fixed on a circular boundary of radius a are discussed in many textbooks.[14,15] When the restoring force is dominated by the tension T (force per unit area) applied to the film's boundary, it is called a membrane and the resonances ν_{ij}^m are given by

$$\nu_{01}^m = 0.38274\ 1/a\ \sqrt{T/\rho}, \tag{1}$$

$$\nu_{ij}^m = \beta_{ij}^m \nu_{01}^m . \tag{2}$$

ν_{01}^m is the frequency of the fundamental mode and ρ is the mass density. For a given mode, i is the number of nodal lines and j is the number of nodal circles. The β_{ij}^m's are constants and are given as follows: $\beta_{11}^m = 1.5933$, $\beta_{21}^m = 2.1355$, $\beta_{02}^m = 2.2954$, $\beta_{31}^m = 2.6531$, $\beta_{12}^m = 2.9173$, etc. When the tension is negligible compared to the elastic rigidity of the film it is called a plate and the resonance ν_{ij}^p are given by

$$\nu_{01}^p = 0.4671\ h/a^2 \sqrt{E/\rho(1-\eta^2)}, \tag{3}$$

$$\nu_{ij}^p = \beta_{ij}^p \nu_{01}^p , \tag{4}$$

where E is the modulus of elasticity, η is the Poisson's ratio, and h is the film's thickness. β_{ij}^p's are given as follows: $\beta_{11}^p = 2.091$, $\beta_{21}^p = 3.426$, $\beta_{02}^p = 3.909$, $\beta_{31}^p = 5.019$, $\beta_{12}^p = 5.983$, etc. In order to obtain elastic properties it would appear more reasonable to work in the limit of Eqs. (3) and (4) which directly provide an elastic constant rather than Eqs. (1) and (2) which contain no implicit dependence on elastic properties. It should be pointed out that the plate modes depend critically on initial warping of the plate. Studies in the second limit (zero tension) where the film was clamped between two annular disks of polished brass and no tension was (purposely) applied, produced results which were not reproducible in successive mountings of identical films. This irreproducibility is attributed to the uncontrollable nature of the small tensions generated at the clamping boundary and make the determination of elastic properties from the plate modes impossible.

With the jig shown in Fig. 1, a typical frequency sweep yielding the resonances of a stretched 25 μm nickel film is shown in Fig. 2. The position of the resonances agrees within 1.5% with prediction of Eq. (2) but not at all with Eq. (4), clearly indicating that tension is indeed governing the behavior of the film. Although modes 2, 3, and 6 are expected to be degenerate, experimentally they turn out to be slightly nondegenerate, the degeneracy most likely being lifted by symmetry breaking perturbations due to nonuniform tension or deviations in the clamping boundary.

The measured strain versus the squared of the resonant frequency is plotted for representative nickel and tantalum films in Fig. 3. The unfilled (filled) circles in each figure represent data points recorded on increasing (decreasing) the tension. For increasing tension the slope of strain versus stress (i.e., frequency squared) monotonically increases with tension. When tension is increased close to the tensile strength of the film the slope becomes very large and signifies the imminent breakup of the film. For nickel the film breakup occurs at ~20 kHz which translates to ~5.5×10^8 N/m^2 close to the tensile strength of bulk Ni (5–9×10^8 N/m^2).[16,17] The breakup of tantalum films occurs at 16 kHz and the corresponding tension is equal to 6.6×10^8 N/m^2, comparable to tensile strengths given in literature ($\sim7.6\times10^8$ N/m^2).[16,17] On reducing the tension applied to the films, a linear slope between strain and ν^2 is obtained. From the slope and Eq. (1) we obtain the biaxial modulus; the results are summarized in Table I. Our values are compared with Y_B calculated[18] using the literature values of C_{ij} and the expressions for Y_B appropriate for the texture of our films: [100] for nickel and tantalum. The agreement between our values which have an estimated precision of 5%

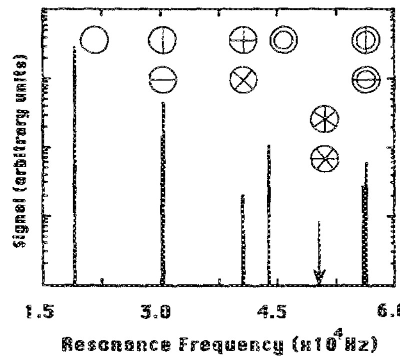

FIG. 2. Resonances of a vibrating 25 μm nickel film. The semilogarithmic plot shows the absolute values of received signal vs frequency. Directly above each peak the configurational representation of its wave function is provided. The resonant frequencies ν_{ij} in the order of increasing frequency are as follows: $\nu_{01} = 18985$, $\nu_{11} = 30221$ and 30417, $\nu_{21} = 40727$ and 40793, $\nu_{02} = 43934$, $\nu_{12} = 56328$ and 55948 Hz. The fifth resonance ν_{31} was not observed. The arrow indicates its theoretically expected value.

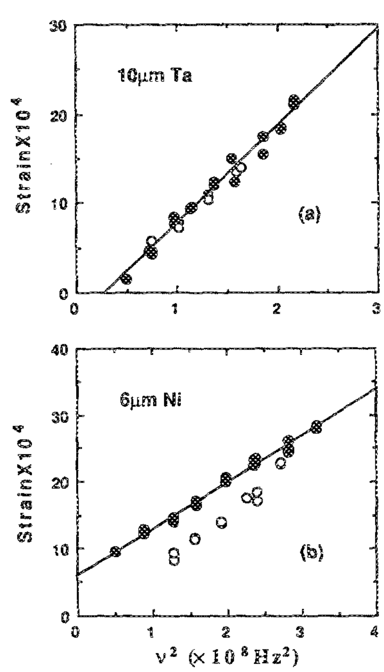

FIG. 3. Strain vs the square of the fundamental mode of (100) oriented (a) 10 μm tantalum film and (b) 6 μm nickel film. The unfilled circles represent increasing tension and are influenced by the plasticity of the film. The biaxial modulus is determined from the slope of the filled circles measured with decreasing tension.

TABLE I. Biaxial moduli of tantalum and nickel films as measured in this experiment and compared to literature values using single-crystal values of C_{ij} for the measured preferred orientation of our films.

Film	Biaxial modulus (TPa)	
	Present work	Calculated
Rolled 10 μm Ta	0.234	0.228
Electroplated 25 μm Ni	0.213	0.222
Electroplated 12.5 μm Ni	0.219	0.222
Rolled 6 μm Ni	0.204	0.222

and the literature ones is good. The use of Eq. (1), which ignores contributions from plate modes, in the analysis of our data introduces errors of less than 1% even for the thickest films.[12]

In summary, by employing a new technique we have measured the biaxial modulus of metallic films. The elastic as well as the nonelastic (plastic) behavior of nickel and tantalum has been examined. The technique involves the measurement of the fundamental mode of a high Q membrane for determination of stress; the strain is measured optically. The biaxial modulus is determined from the ratio of stress to strain with a precision mainly limited by the resolution of the optical device. The setup employed was capable of measuring values of biaxial modulus with an estimated precision of $\sim \pm 5\%$. The measured values are more reliable than those obtained by other mechanical techniques such as the "bulge tester." No knowledge of film thickness is required. The measurements are free from ambiguities due to shape and deformation (warping) of the films.

This work was supported by the ONR under contract No. N00014-88K-0210 and the U. S. DOE, Basic Energy Sciences-Materials Sciences under contract No. W-31-109-ENG-38.

[1] R. W. Hoffman, in *Physics of Thin Films*, edited by G. Hass and R. E. Thun, Vol. 13 (Academic, New York, 1966), p. 211.
[2] For a review see I. K. Schuller, in *IEEE 1985 Ultrasonics Symposium*, edited by B. R. McAvoy (IEEE, New York, 1985), p. 1903.
[3] W. M. C. Yang, T. Tsakalakos, and J. E. Hilliard, J. Appl. Phys. **48**, 876 (1977).
[4] T. Tsakalakos and J. E. Hilliard, J. Appl. Phys. **54**, 734 (1983).
[5] G. H. Henein and J. E. Hilliard, J. Appl. Phys. **54**, 728 (1983).
[6] Hideo Itozaki, Ph.D. thesis, Northwestern University, 1982 (unpublished).
[7] B. S. Berry and W. C. Pritchet, Thin Solid Films **33**, 19 (1976).
[8] D. Baral, J. B. Ketterson, and J. E. Hilliard, J. Appl. Phys. **57**, 1076 (1985).
[9] L. R. Testardi, R. H. Willens, J. T. Krause, D. B. McWhan, and S. Nakahara, J. Appl. Phys. **52**, 510 (1981).
[10] For a review see M. Grimsditch, in *Light Scattering in Solids V*, edited by M. Cardona and G. Guntherodt (Springer, Heidelberg, 1989), p. 284.
[11] B. Clemens and G. Eesley, Phys. Rev. Lett. **61**, 2356 (1988).
[12] A. Fartash, M. Grimsditch, and I. K. Schuller (unpublished).
[13] B. S. Berry and W. C. Pritchet, IBM J. Res. Develop. **19**, 334 (1975).
[14] P. M. Morse, *Vibration and Sound*, 2nd ed. (McGraw-Hill, New York, 1948), p. 172.
[15] S. Timoshenko, D. H. Young, and W. Weaver, Jr., *Vibration Problems in Engineering*, 4th ed. (Wiley, New York, 1974), Chap. 5.
[16] *American Institute of Physics Handbook*, 3rd. ed. (McGraw-Hill, New York, 1972).
[17] *Tables of Physical and Chemical Constants*, G. W. C. Kaye and T. H. Laby, eds. (Clowes, London, 1966).
[18] D. Baral, J. E. Hilliard, J. B. Ketterson, and K. Miyano, J. Appl. Phys. **53**, 3552 (1982).