

pressure-derivative data for all temperatures of calculation refer to room temperature, we are led to conclude that the pressure derivatives of elastic constants change significantly with temperature.

It is worth mentioning that de Launay's model and Sharma and Joshi's model give essentially the same results, but the latter is mathematically more straightforward and logical than the former. Bhatia's model, on the other hand, is based on a completely different approach, of screened Coulomb interaction between ions in a metal. It yields good results in spite of the

fact that only nearest-neighbor interactions are considered.

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## Electroreflectance Changes in Dielectric Constants of Au and Ag by Modulated Ellipsometry\*

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The changes induced by an applied electric field in the real and imaginary parts of the dielectric constants of Au and Ag are determined, as functions of photon energy, by ellipsometer measurements combined with phase-sensitive detection. This experiment provides all the necessary data for direct calculation of the complex dielectric constant in the absence of the field, the field-induced changes, and the depth into the sample to which the dielectric constant is modulated by the field, at given photon energy from measurements taken only at that energy. Hence the Kramers-Kronig analysis and any accompanying extrapolations to experimentally inaccessible photon energies are unnecessary. Structure in the modulated dielectric constants which is masked in the normal-incidence electroreflectance spectra, such as the structure around 3 eV in Ag, is brought out much more clearly in our more direct measurements. The results are consistent with the usual interpretation of the band structure of Au and Ag, and the modulation depths are of the same order as the Thomas-Fermi screening lengths.

WE have employed electric-field-modulated ellipsometry<sup>1</sup> to calculate changes in the real part  $\delta\epsilon_1(\omega)$  and the imaginary part  $\delta\epsilon_2(\omega)$  of the dielectric constant  $\epsilon(\omega) = \epsilon_1(\omega) - i\epsilon_2(\omega)$  of vacuum-evaporated Au and Ag. The changes are induced by a high ( $\sim 10^5$ - $10^7$  V/cm) electric field normal to the metal surface. The photon energy range measured is  $1.8 < \hbar\omega < 4.1$  eV.

The angles  $\psi$  and  $\Delta$  relating the polarization state of the obliquely reflected beam to that of the incident beam, the changes in these angles  $\delta\psi$  and  $\delta\Delta$  induced by the modulating electric field, and the total oblique incidence electroreflectance  $\delta R/R$  are obtained as functions of photon energy by the procedure of Ref. 1.

The modulating field is applied by varying the sample potential sinusoidally at  $\sim 21$  Hz between 0 and  $-1.5$  V with respect to a Pt counterelectrode, with sample and counterelectrode immersed in a 1N KCl electrolyte.

From the Thomas-Fermi theory, the field is expected to penetrate less than 1 Å into the Au and Ag,<sup>2</sup> and,

intuitively, the effective depth  $d$  to which  $\epsilon(\omega)$  is altered by the field should be of the same order as the Thomas-Fermi screening length. For the photon energies of interest here,  $d$  is much less than the light wavelength  $\lambda$ . Therefore, the approximation of Jacobsson<sup>3</sup> and others<sup>4,5</sup> can be used, replacing the dielectric constant of the field-modulated surface layer, which varies with distance into the sample, by an average over the modulation depth. The reflecting surface then consists of the transparent electrolyte with dielectric constant  $\epsilon_e$ , the modulated surface layer of thickness  $d$  and dielectric constant  $\epsilon_s = \epsilon_1 + \delta\epsilon_1 - i(\epsilon_2 - \delta\epsilon_2)$ , and the interior of the metal with dielectric constant  $\epsilon_f$ . The relationship of  $\psi$  and  $\Delta$  measured at an angle of incidence  $\phi$  to the properties of such a reflecting surface<sup>6,7</sup> is

$$\tan\psi e^{i\Delta} = \frac{p_1 + (p_2 + p_1 s_2 s_1) e^D + p_1 s_1 s_2 e^{2D}}{s_1 + (s_2 + s_1 p_1 p_2) e^D + s_2 p_1 p_2 e^{2D}} \equiv G_0, \quad (1)$$

<sup>3</sup> R. Jacobsson, in *Progress in Optics*, edited by E. Wolf (North-Holland Publishing Co., Amsterdam, 1966), Vol. 5, p. 247.

<sup>4</sup> P. Drude, *Wied. Ann.* **43**, 26 (1891).

<sup>5</sup> L. Rayleigh, *Proc. Roy. Soc. (London)* **A86**, 207 (1912).

<sup>6</sup> F. L. McCrackin, E. Passaglia, R. R. Stromberg, and H. L. Steinberg, *J. Res. Natl. Bur. Std.* **67A**, 363 (1963).

<sup>7</sup> P. Drude, *Ann. Physik* **272**, 532 (1889).

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<sup>1</sup> A. B. Buckman and N. M. Bashara, *J. Opt. Soc. Am.* **58**, 700 (1968).

<sup>2</sup> J. Feinleib, *Phys. Rev. Letters* **16**, 1200 (1966).

where  $p_1$  and  $s_1$  are the Fresnel reflection coefficients for parallel and perpendicular polarizations at the electrolyte-surface interface,  $p_2$  and  $s_2$  are the Fresnel reflection coefficients for parallel and perpendicular polarizations at the surface-interior interface, and

$$D = -(4\pi id/\lambda)(\epsilon_s - \epsilon_e \sin^2\phi)^{1/2}. \quad (2)$$

Since all changes arising in  $\epsilon$  due to electroreflectance are small, we expand in a Taylor series about values measured in the absence of a field. Forming the total differential of (1) and solving for  $\delta\epsilon$  yields

$$\delta\epsilon = \delta\epsilon_1 + i\delta\epsilon_2 = (\sec^2\psi\delta\psi + i \tan\psi\delta\Delta)e^{i\Delta} /$$

$$\left[ \frac{\partial G_0}{\partial D} \frac{\partial D}{\partial \epsilon_s} + \sum_{i=1}^2 \left( \frac{\partial G_0}{\partial p_i} \frac{\partial p_i}{\partial \epsilon_s} + \frac{\partial G_0}{\partial s_i} \frac{\partial s_i}{\partial \epsilon_s} \right) \right]. \quad (3)$$

All derivatives are evaluated at the values of the arguments in the absence of a field, i.e.,  $\epsilon_s = \epsilon_f$  and  $s_2 = p_2 = 0$ . The unmodulated  $\epsilon_f$  is

$$\epsilon_f = \sin^2\phi \left[ \tan^2\phi \left( \frac{1 - \tan\psi e^{i\Delta}}{1 + \tan\psi e^{i\Delta}} \right)^2 + 1 \right] \epsilon_e. \quad (4)$$

The partial derivatives in (3) are then given by

$$\begin{aligned} \frac{\partial G_0}{\partial D} &= 0, \\ \frac{\partial D}{\partial \epsilon_s} &= -2\pi id/\lambda(\epsilon_f - \epsilon_e \sin^2\phi)^{1/2}, \\ \frac{\partial G_0}{\partial p_1} &= \frac{1}{s_1}, \quad \frac{\partial G_0}{\partial s_1} = \frac{-p_1}{s_1^2}, \\ \frac{\partial G_0}{\partial p_2} &= \frac{(1-p_1^2)e^D}{s_1}, \quad \frac{\partial G_0}{\partial s_2} = \frac{-p_1(1-s_1)^2e^D}{s_1^2}. \end{aligned} \quad (5)$$

At each photon energy for which a measurement is made,  $\delta\epsilon$  is calculated for a preselected set of assumed  $d$ . From these results, either a theoretical total oblique-incidence or a normal-incidence electroreflectance spectrum is calculated. The assumed  $d$  which gives the best fit to experimental results is selected as correct.

Spectra obtained for  $\delta\epsilon_1$ ,  $\delta\epsilon_2$ , and calculated normal-incidence electroreflectance spectra  $\delta R/R$  are shown for Au in Fig. 1 with  $d=0.2 \text{ \AA}$ , and for Ag in Fig. 2 with  $d=0.5 \text{ \AA}$ . With these values for  $d$ , the  $\delta R/R$  spectra are in good qualitative agreement with those observed experimentally by Feinleib.<sup>2</sup> Slight changes ( $\sim 1 \text{ \AA}$ ) in assumed  $d$  from these values cause the calculated  $\delta R/R$  to change by nearly an order of magnitude. The observed modulation depth is thus of the same order as the field penetration depth predicted using the Thomas-Fermi theory.

This method yields  $\delta\epsilon_1$  and  $\delta\epsilon_2$  without recourse to the Kramers-Kronig analysis which is required when these quantities are obtained from the electroreflectance in-

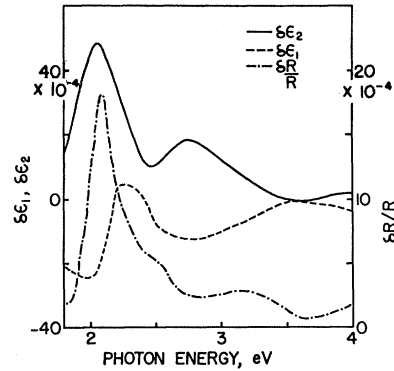


FIG. 1.  $\delta\epsilon_1$  and  $\delta\epsilon_2$ , and  $\delta R/R$  for Au as a function of photon energy.

terities alone. This feature may be particularly important for interpreting measurements such as Feinleib's and ours, where the measured photon energy range is relatively narrow. Particularly, if the oscillator strengths are not exhausted until very high photon energies (as Ehrenreich and Phillip<sup>8</sup> and Beaglehole<sup>9</sup> have pointed out for the noble metals), extrapolations of the Kramers-Kronig integral to experimentally inaccessible photon energies may yield misleading results for  $\delta\epsilon_1$  and  $\delta\epsilon_2$ .

The small peak near 3 eV in the normal-incidence Ag electroreflectance spectrum, of whose reality Feinleib expressed some doubt,<sup>2</sup> appears clearly in the calculated curve for  $\delta\epsilon_2$  obtained from our measurements. Also, the wide shoulder between 2.5 and 3 eV in the Au normal-incidence spectrum is more clearly defined in our  $\delta\epsilon_1$  and  $\delta\epsilon_2$  curves. It is therefore apparent that the ellipsometer measurements can more clearly indicate some structure which may be masked in normal- or oblique-incidence data where only the electroreflectance is measured. How such masking might occur is seen

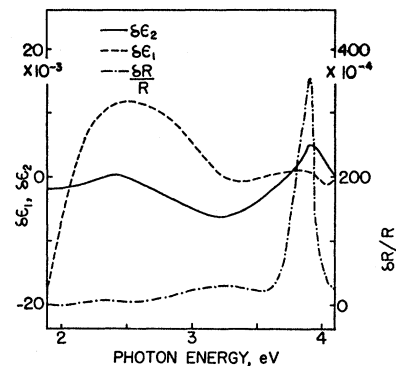


FIG. 2.  $\delta\epsilon_1$ ,  $\delta\epsilon_2$ , and  $\delta R/R$  for Ag as a function of photon energy.

<sup>8</sup> H. Ehrenreich and H. Phillip, *Phys. Rev.* **128**, 1622 (1962).

<sup>9</sup> D. Beaglehole, in *Optical Properties and Electronic Structure of Metals and Alloys*, edited by F. Abeles (North-Holland Publishing Co., Amsterdam, 1966), p. 154.

from the equation of Fischer and Seraphin<sup>10,11</sup>:

$$\delta R/R = \alpha(\phi, \epsilon_1, \epsilon_2) \delta \epsilon_1 + \beta(\phi, \epsilon_1, \epsilon_2) \delta \epsilon_2, \quad (6)$$

where  $\alpha$  and  $\beta$  are also functions of the polarization of the incident light when  $\phi \neq 0$ . It is obvious that the two terms of (6) might nearly cancel, even when the structure in  $\delta \epsilon_1$  and  $\delta \epsilon_2$  is pronounced, if  $\alpha$  and  $\beta$  had proper magnitude and sign.

The measurements are consistent with the usual interpretation of the band structure in Au and Ag. The  $L_{32} \rightarrow L_2'$  transition at 2.1 eV in Au and the hybrid interband transition and plasma resonance at 3.9 eV in Ag are clearly visible. Analysis of the  $\delta \epsilon_1$  and  $\delta \epsilon_2$  lineshapes obtained by this method should aid the study of other excitations, in these and other materials, whose identification from band theory may be less clear, since parabolic and saddle-point transitions give easily identified characteristic line shapes.

<sup>10</sup> B. O. Seraphin and N. Bottka, Phys. Rev. Letters **15**, 104 (1965).

<sup>11</sup> J. E. Fischer and B. O. Seraphin, Solid State Commun. **5**, 973 (1967).

This theory assumes scalar  $\epsilon_1$ ,  $\epsilon_2$ ,  $\delta \epsilon_1$ , and  $\delta \epsilon_2$ . The obliquely incident beam is affected by components of the tensors  $\delta \epsilon_1$  and  $\delta \epsilon_2$  both along and perpendicular to the modulating field. This is not the case with normal-incidence measurements, where the electric vector of the light is always perpendicular to the applied electric field. The agreement of our calculated  $\delta R/R$  spectra with those observed in normal-incidence experiments suggests that the anisotropy introduced is small. This may be due to the fact that, in a polycrystalline film, the crystal axes effectively assume all orientations with respect to the optical and applied fields. The tensor character of  $\delta \epsilon_1$  and  $\delta \epsilon_2$ , contained in matrix elements which in turn depend on crystal orientation with respect to the modulating field,<sup>12-14</sup> may thus be masked in polycrystalline samples.

<sup>12</sup> D. E. Aspnes, Phys. Rev. **147**, 544 (1966).

<sup>13</sup> D. E. Aspnes, Phys. Rev. **153**, 972 (1967).

<sup>14</sup> D. E. Aspnes, P. Handler, and D. F. Blossey, Phys. Rev. **166**, 921 (1968).

## Magnetoacoustic Dispersion and Attenuation; Comparison of Experiment with the Free-Electron Theory\*

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The Cohen-Harrison-Harrison formalism has been used to calculate longitudinal magnetoacoustic dispersion and attenuation curves in transverse fields for values of the parameter  $ql$  between 1.0 and 200 ( $ql = \omega v_f \tau / v$ ). The results of this calculation show that as  $ql$  is decreased from the limiting case of  $ql \gg 1$ , the oscillations in the attenuation are damped out faster than the oscillations in the dispersion. Experimental data are given for both the attenuation and fractional velocity shift in aluminum and are shown to be in qualitative agreement with the theory. An examination of magnetoacoustic-attenuation data in potassium shows that the deviation of the attenuation minima from the positions predicted in the limit  $ql \gg 1$  agrees quantitatively with theory.

### INTRODUCTION

**T**HEORETICAL treatments of the magnetoacoustic effect can be divided into those based upon the free-electron model of a metal and those which are designed to apply to a metal with an arbitrary Fermi surface. Both types of treatments are semiclassical in nature and both assume an isotropic electron relaxation time over the Fermi surface.

A comprehensive treatment of ultrasonic attenuation in real metals is given by Pippard.<sup>1</sup> He first considers the attenuation in the absence of a magnetic field and then applies the field. The effect of real metals on the attenuation is introduced by means of a deformation

parameter. This parameter is a measure of how the local Fermi surface deforms when subjected to a strain. In the presence of an acoustic wave, this strain is produced by the electric field set up between the ions and electrons by the wave. It would appear that such a treatment, which can be used for arbitrarily shaped Fermi surfaces and which includes variations in the strength of the coupling between the electrons and the acoustic phonons, should be used in interpreting experimental magnetoacoustic data. However, when this is attempted, several inherent difficulties appear. One problem is that a determination of the shape of the attenuation curve involves evaluating many integrals over the Fermi surface. Thus, while one can predict the shape of the experimental curve if the Fermi surface is known, it is far more difficult to determine the

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<sup>1</sup> A. B. Pippard, Proc. Roy. Soc. (London) **257**, 165 (1960).