Optical Plasma-Resonance Absorption in Thin Films of Silver and Some Silver Alloys

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Plasma oscillations in Ag, *Ag*-Au, *Ag*-Cd, and *Ag*-In have been studied by optical transmittance measurements using *p*-polarized light at oblique incidence. The plasma resonance is observed as an increased absorption at the plasma frequency. The resonance disappears at about 40 at.% Au and at as little as about 3 at.% Cd or In. The energy of the resonance was found to be almost constant with varying impurity content. For the Ag-Au system this is in contrast with results obtained from electron energy-loss experiments and from optical data. It is argued that in some cases optical plasma-resonance absorption measurements are a more correct measure of the plasma frequency than are other experimental methods. The plasma resonance in pure Ag has also been studied in photoemission yield.

I. INTRODUCTION

THE free electrons and the ion cores in a metal form a stable plasma. This can be caused in different ways to perform oscillations. The quanta of these are the plasmons. Most experimental studies of these oscillations have been made using electrons in the keV range for excitation in so-called characteristic energy-loss experiments.[1]

During the last decade there has been a considerable interest in the role of plasmons in optical studies of solids. There is a close connection between the optical constants of a solid and the inelastic scattering of electrons. In principle, the plasmon energy can be calculated from optical data.[1,2] However, there is generally no interaction between a photon and the plasma, because of the transverse nature of the electromagnetic field and the longitudinal character of the plasma waves in the bulk of an optically isotropic and homogeneous solid. Plasmons, therefore, cannot usually be excited by light, nor can the plasmons decay into photons. If the specimen consists of a thin film, however, there are a number of electronic vibrational modes, some of which can be excited by light. Steinmann[3] has given an excellent review on the topic of optical plasma resonances.

The theory of collective electronic motion in a thin slab was originally treated by Ritchie[4] and has recently been developed independently by Kliewer and Fuchs,[5] by Sauter, Sturm, and Forstmann,[6–9] and by Melnyk and Harrison.[10,11]

One of the radiative modes can be experimentally observed under special conditions as an increase in the optical absorption of the film and this phenomenon is therefore called optical plasma-resonance absorption. Compared with electron energy-loss experiments, relatively few elements have been studied with regard to optically excited plasmons. (So far only the following elements have been investigated: Ag,[12–15] Al,[16–18] Mg,[19] K,[20–22] Ge,[23] Bi.[23]) In this paper we describe some studies of this type on silver films, and, for the first time on metallic alloys, on the following systems *Ag*-Au, *Ag*-Cd, and *Ag*-In. The effect on plasmon excitation on the photoelectric yield of silver films has also been investigated using a cesiated film.

II. THEORY

The detailed theory for calculating the collective electronic motion in a metallic film has been derived in Ref. 5. In the following we will give only a brief outline of those features of the theory which are of relevance to the interpretation of our experimental results.

There exist two types of long-wavelength collective excitations of the conduction electrons in a metallic film. These can be classified as radiative and nonradiative excitations, depending on the electromagnetic fields outside the slab. The dependence of the collective waves on the material is best described by the complex dielectric constant which for a free-electron gas can be

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[2] D. Pines, *Elementary Excitations in Solids* (W. A. Benjamin, Inc., New York, 1963).
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[23] T. Sasaki and A. Ejiri, in *Optical Properties and Electronic Structure of Metals and Alloys*, edited by F. Abelès (North-Holland Publishing Co., Amsterdam, 1966), p. 417.

written as

$$\epsilon(\Omega,\gamma)=1-(\Omega^2+i\Omega\gamma)^{-1}, \quad (1)$$

where

$$\Omega=\Omega'+i\Omega''=\omega/\omega_p. \quad (2)$$

Here, ω_p is the plasma frequency and γ is the intrinsic damping constant.

It can be shown that the nonradiative excitations correspond to two kinds of surface modes with exponentially decaying electromagnetic fields outside the film ($\Omega''=0$). The dispersion curves can be drawn with Ω' as a function of the reduced wave vector k/k_p (the k direction is in the plane of the film). They both fall below the free-space photon dispersion line, $\Omega'=k/k_p$, and depend on the thickness of the film, since the two surface modes couple with each other. In the limiting case of a thick crystal, one obtains $\Omega'=1/\sqrt{2}$ and $\epsilon=-1$. Thus this corresponds to the same frequency $\omega_p/\sqrt{2}$ as the ordinary longitudinal surface plasmon. Because of the nonradiative character of these surface modes, they cannot, in general, be excited by light. However, on a rough surface or metallic grating, the irregularities serve the purpose of preserving momentum so that surface modes can be excited by light.[24,25]

The nonradiative or virtual modes, however, correspond to energy transport of electromagnetic waves out of the film, resulting in a radiative decay of the vibrational mode. The appropriate expression for the frequency is therefore given by Eq. (2), where Ω' gives the oscillation frequency and Ω'' is a measure of the frequency spread due to decay into radiation. Of the two types of polarization of the virtual modes p and s, only the p-type is of interest in our case, since only this can be excited by light strongly enough to be detectable. The solution of Maxwell's equations for this case gives many modes of vibration of which one, denoted OT_1 in Ref. 5, is of particular interest, being the most well-defined mode. This corresponds to the excitation that was considered by Ferrell when predicting the plasma radiation.[26] In what follows, only this mode will be considered.

Dispersion curves of the OT_1 mode for two values of film thickness d have been calculated using Ref. 5. These are shown in Fig. 1. For $\theta=0$ (θ is the angle of incidence in the optical experiments and is related to the wave vector of the plasmon through $\theta=\arcsin kc/\omega$), the frequency of the mode is independent of thickness and occurs at ω_p. The dependence of frequency on the angle θ and the reduced thickness W, where $W=\omega_p d/c$, is, for a thick film,

$$\Omega'\approx(\cos\theta)^{-1}, \quad \Omega''\approx-\sin^2\theta\,(W\cos\theta)^{-1}, \quad (3)$$

and for a thin film,

$$\Omega'\approx 1+\tfrac{1}{8}W^2\tan^2\theta, \quad \Omega''\approx -W\sin^2\theta\,(4\cos\theta)^{-1}. \quad (4)$$

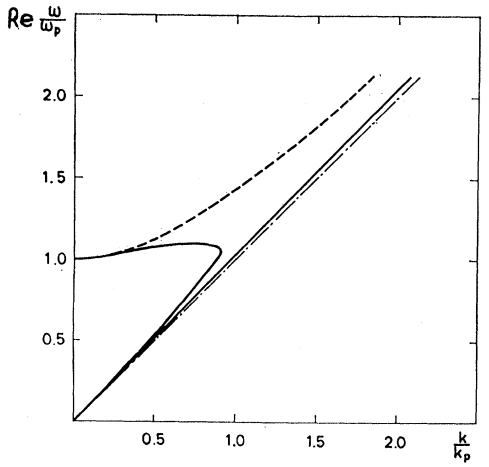

FIG. 1. Dispersion relation for the OT_1 mode calculated from Ref. 5. $\mathrm{Re}\omega/\omega_p$ is the normalized real part of the oscillation frequency, and k/k_p is the normalized wave vector of the oscillation in the plane of the film. The dash-dotted curve is the free-space photon dispersion line. The solid curve corresponds to the reduced thickness $W=1.40$ and the dashed curve to $W=10.0$.

These expressions are good approximations for angles less than about 45°.

In the optical plasma-resonance absorption experiments, polarized light is incident on a metallic film and the transmittance as a function of polarization, angle, and photon energy is recorded. Figure 2 shows such recordings for a silver film. A strong increase in absorption can be observed for non-normal incidence of p-polarized light at a photon energy of 3.78 eV. This corresponds to ω_p for silver, and is due to the excitation of the OT_1 mode mentioned above.

In the expression for the transmittance, two kinds of damping mechanisms have to be considered. First, there is an internal damping corresponding to interband and intraband transitions. Optical absorption occurs only if this damping is different from zero. In the dielectric constant above, it was represented by γ, which so far has been set equal to zero. The other kind of damping is associated with the decay of the plasmon into a photon and is represented above by Ω''.

Starting from Fresnel's laws for reflectance and transmittance, McAlister and Stern[13] have derived a simple formula for the transmittance around the plasma frequency of an unsupported thin film. It has subsequently been shown[6,27] that Fresnel's laws are not valid when plasma oscillations occur, but that the correction to the final formula is small. The formula for the transmittance of p-polarized light has the Lorentzian form

$$T_p=\frac{4(\omega-\omega_p)^2+\tau_d^{-2}}{4(\omega-\omega_p)^2+(\tau_d^{-1}+\tau_r^{-1})^2}. \quad (5)$$

Here,

$$\tau_d^{-1}=2\epsilon_2(d\epsilon_1/d\omega)_{\omega=\omega_p}^{-1} \quad (6)$$

[24] R. H. Ritchie and R. E. Wilems, Phys. Rev. **178**, 372 (1969); and references given therein.
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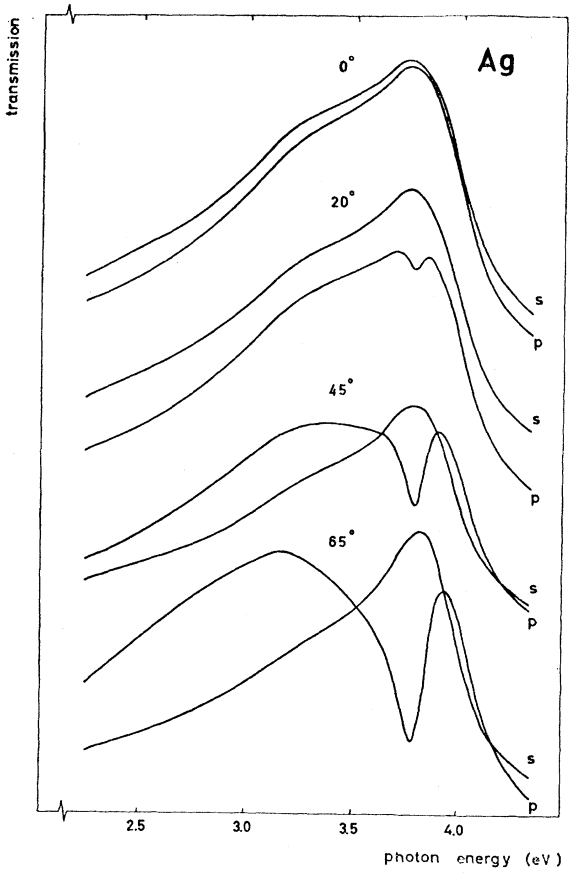

FIG. 2. Transmittance curves of Ag films for s- and p-polarized light at different angles of incidence. The dip at 3.78 eV for p-polarized light is caused by the plasma resonance.

is the internal damping frequency and

$$\tau_r^{-1} = W(d\epsilon_1/d\omega)_{\omega=\omega_p}^{-1} \tag{7}$$

is the radiation damping frequency. Since the derivation of this result is not available in published form, the essential features of it, together with the approximations made, are given in the Appendix. For a film on a substrate, we have also derived an expression for the transmittance. The result is that Eq. (5) is multiplied by a correction factor due to the presence of the substrate

$$C = 4A \Big/ \epsilon_s^{1/2}\left(1+\frac{(\epsilon_s-1+\cos^2\theta)^{1/2}}{\epsilon_s\cos\theta}\right)^2, \tag{8}$$

where ϵ_s is the dielectric constant of the substrate and $A \approx 1$. The derivation of this formula is also given in the Appendix. The expression for the radiation damping constant τ_r^{-1} should also be multiplied by a correction factor:

$$c = \frac{2}{1+(\epsilon_s-1+\cos^2\theta)^{1/2}/\epsilon_s\cos\theta}, \tag{9}$$

while τ_d^{-1} is left unchanged by the presence of the substrate.

III. EXPERIMENTAL

The specimens were prepared by vacuum evaporation (10^{-6} Torr) onto quartz disks. The Ag-Au and Ag-In samples were annealed for 3 h at 300°C in the vacuum evaporator to remove concentration gradients. The annealing temperature for the Ag-Cd samples had to be reduced to 100°C because higher temperatures caused a surface deterioration. The elementary composition of the samples was determined from lattice parameter and atomic flame-spectroscopy measurements. The optical transmittance measurements were made in air using a double-beam spectrophotometer equipped with a Glan-Thompson prism polarizer. The film thickness was measured by the Tolansky interference method, which gives an accuracy of 20–30 Å. To obtain optimal transmittance the films were made 350–450 Å thick.

In the theory of collective oscillation modes described above, it is important to notice the assumption that the films consist of plane parallel homogeneous layers. For very thin films, however, an island structure is very often formed during evaporation and annealing of the films. This gives rise to anomalies in the transmission spectra in such a way that the plasma frequency is now determined by the geometrical shape of the islands. We observed this phenomenon for very thin films ($d<100$ Å) of Ag-Au. The same behavior has been reported for thin K films,[22] where the anomalies have been explained by the assumption that the islands consist of rotation ellipsoids with the symmetry axis perpendicular to the foil surface.

IV. RESULTS AND DISCUSSION

The transmittance T as a function of photon energy for a pure silver film is shown in Fig. 2 for different polarizations of the incident light and for different angles of incidence. At normal incidence ($\theta=0$), there is a maximum in T at about 3.8 eV for both s- and p-polarized light. At lower photon energies, the transmittance is reduced because of free-electron absorption, and for higher energies because of the onset of interband transitions from the d band to the Fermi level and across the conduction band gap at the L point. Changing the angle of incidence does not influence the character of the curve for s-polarized light. For p-polarized light, however, a minimum in T, corresponding to an increase in absorption, is developed at 3.78 eV as the angle of incidence is increased. As described previously, this corresponds to the excitation of a plasma mode. The value obtained for the plasma frequency is in good agreement with previous results obtained using different

experimental methods.[12–15,28–36] The large shift of the plasmon energy for silver from the free-electron value of 9.2 to 3.78 eV is a consequence of the hybrid nature of the oscillation caused by the interband transitions.[37]

According to Eq. (4) ($\theta<45°$) and Fig. 1, the value of ω_p for a free-electron gas is dependent on the angle of incidence. For the small film thicknesses which had to be used in this experiment, the angular variation is small and was undetectable. The fact that we have no free-electron oscillation may also contribute to a small angular dependence.

Silver-Gold Alloys

The influence on the plasma resonance of introducing gold into the silver matrix is depicted in Fig. 3. As can be seen from these curves, the effect is to reduce the strength of the resonance and also to increase the width until it disappears completely at about 40-at.% Au content. Although it becomes somewhat ambiguous where to locate the true minimum in the transmittance as it gets less pronounced, the energy of the plasma resonance can be seen to be shifted downwards in energy from 3.78 to about 3.70 eV.

The increase in width with increasing Au content can be explained by strong internal damping [Eq. (6)], mainly caused by a decreasing dielectric dispersion $d\epsilon_1/d\omega$ but also by an increase in ϵ_2.

An estimate of the values of τ_d^{-1} and τ_r^{-1} for pure silver can be made using Eqs. (5) and (6). For $\theta=60°$, we obtain $\tau_d^{-1}=0.11$ eV and $\tau_r^{-1}=0.15$ eV. Using these values, one obtains $\epsilon_2=0.21$ and $d\epsilon_1/d\omega=7.7$ eV^{-1}, which is in fair agreement with reported values of the optical constants.[37,38] Even if ϵ_2 is assumed to be frequency-dependent, the above results are influenced to a negligible degree. The internal damping of 0.11 eV obtained in this experiment can be compared with the corresponding value obtained from electron energy-loss experiments, which is 0.075 eV.[39] The discrepancy can be explained by the fact that the dielectric loss is larger in optical experiments because of the smaller k values of the plasmon. The damping in the optical case is therefore increased by the presence of imperfections in the film such as dislocations and grain boundaries. This effect can also explain the wide spread in reported values on optical constants.

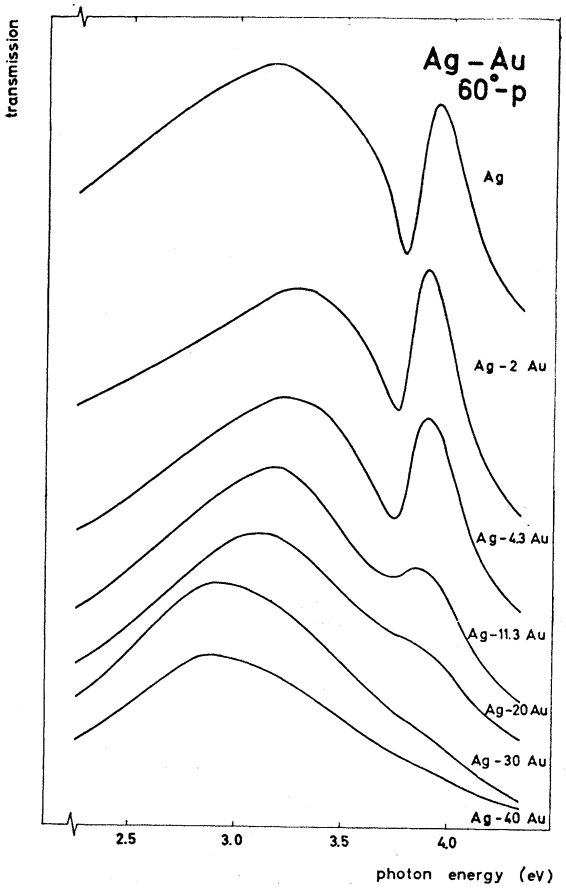

FIG. 3. Transmittance curves for Ag-Au alloys. p-polarized light at 60° angle of incidence was used. The plasma resonance broadens and disappears at about 40 at.% Au.

The collective electronic oscillations in the Ag-Au system have previously been investigated by electron energy-loss experiments[40] and by optical measurements of the reflectance and transmittance.[41] The results from the different methods are compared with the present results in Fig. 4.

In the case of the electron energy-loss experiments, both bulk and surface plasmons are excited. Because of the difficulty in obtaining high resolution in this type of experiment, the observed peak is a mixture of the two plasmons. In the optical experiments, the relevant quantity is the loss function $-\mathrm{Im}(1/\epsilon)$, which exhibits a peak at the plasmon frequency. Both electron energy-loss experiments[40] and optical measurements[41] show a continuous change in the energy of the loss function peak on going from Ag to Au (Fig. 4). In the optical

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[29] W. R. Brown, P. Wessel, and E. P. Trounson, Phys. Rev. Letters **5**, 472 (1960).
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[39] J. Daniels, Z. Physik **203**, 235 (1967).
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[41] H. Fukutani and O. Sueoka, in *Optical Properties and Electronic Structure of Metals and Alloys*, edited by F. Abelès (North-Holland Publishing Co., Amsterdam, 1966), p. 565.

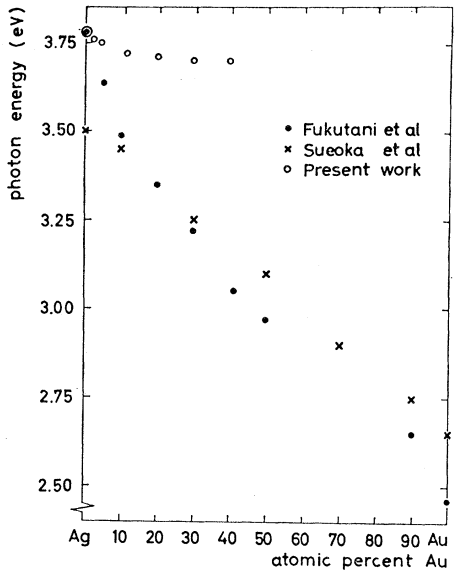

FIG. 4. Energy of the plasmon in *Ag*-Au alloys determined from electron energy-loss experiments (×), from optical constants (●), and from optical plasma-resonance experiments (○, present work).

work, the conclusion is reached that plasma oscillations only exist up to about 30 at.% Au, because the condition $\epsilon_1=0$ is not fulfilled above this limit. The general tendency for a decrease in plasmon energy with increasing Au content is in agreement with our results. However, there is a discrepancy in the absolute values of the plasmon energies. This can probably be explained as follows: The peak in the loss function above 30 at.% Au should be associated with the interband transition, which changes continuously from the Ag value (3.8 eV) to the Au value (2.4 eV). The loss function for concentrations below 30 at.% Au therefore corresponds both to the interband transition and the plasmon excitation. The peak in the loss function obtained from electron energy-loss experiments or optical constants is in this case, therefore, not a correct measure of the plasmon frequency because of the interference of the interband transition, while the optical-resonance absorption experiments yield the true plasma oscillation frequency. (For pure Ag and for alloys with low Au content, the interband transition occurs at higher energy than the plasma excitation energy and no interference occurs.)

The peak in transmittance at normal incidence for *p*-polarized light and at all angles for *s*-polarized light correspond to the onset of the interband absorption. As noted above, this interband threshold value is lowered with increasing amount of Au. One therefore expects the plasmon energy to decrease along similar lines. Experimentally, however, a much smaller shift is observed. This is due to the fact that broadening of the interband edge is also reflected as a "broadening" in ϵ_1, thus shifting $\epsilon_1=0$ to higher values.

Silver-Cadmium and Silver-Indium Alloys

Experimental results for the *Ag*-Cd and *Ag*-In systems are shown in Figs. 5–8. The optical constants for the *Ag*-In system have been determined by Morgan and Lynch[42] and for the *Ag*-Cd system by Green.[43] The energies obtained from the calculated loss function peak agree with our results. The most remarkable feature of the present results is the rapidness of the disappearance of the plasma resonance. This occurs at a concentration as small as 3 at.% Cd or In. At the same time, a new structure appears in the absorption as an additional hump at about 3.5 eV. This feature is also present for the normally incident light and is independent of the polarization of the light for non-normal incidence, indicating that this structure is a genuine band-structure effect. It is probably due to a splitting of the

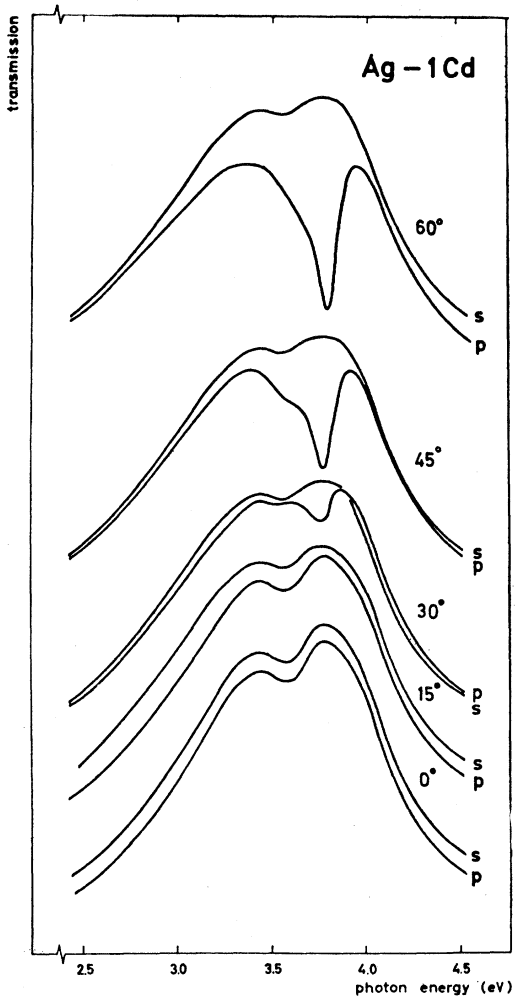

FIG. 5. Transmittance of Ag alloyed with 1 at.% Cd for *s*- and *p*-polarized light at different angles of incidence. An extra interband transition around 3.6 eV is observed.

[42] R. M. Morgan and D. W. Lynch, Phys. Rev. **172**, 628 (1968).
[43] E. L. Green, thesis, Temple University, 1965 (unpublished).

degenerate absorption threshold in pure Ag caused by the perturbation of the impurities on the silver matrix. Because the new absorption feature appears at a lower energy than the plasmon energy, it offers the plasmon a possible decay channel. This is already of such a strength at an impurity concentration of 3 at.% that the internal damping will completely prevent the occurrence of the plasma oscillations even though the condition $\epsilon_1=0$ is probably still valid.

The energy for which $\epsilon_1=0$ in *Ag*-Cd and *Ag*-In should increase with increasing impurity concentration (as a first approximation), because of the following two effects: Alloying with multivalent atoms Cd and In increases the electron concentration. Following the reasoning of the rigid-band model, this increases the interband transition energy from the *d* bands to the Fermi level, and the zero of ϵ_1 ought to increase correspondingly. According to the Friedel model, on the other hand, this energy difference should be invariant on alloying. Experimentally, a slight increase is observed. The increased free-electron concentration also gives a higher value of the free-electron plasmon energy, which should tend to make ϵ_1 become zero at higher energy.

Experimentally, however, we find that the plasma-resonance absorption changes very little with concentration. This can probably be explained by the fact that the interband edge broadens because of the extra hump in ϵ_2, causing a shift of $\epsilon_1=0$ towards lower energies. The increase in the value of ϵ_2 also has the tendency to shift the plasma frequence towards lower values. The total effect is, therefore, to leave the plasma energy almost constant on alloying.

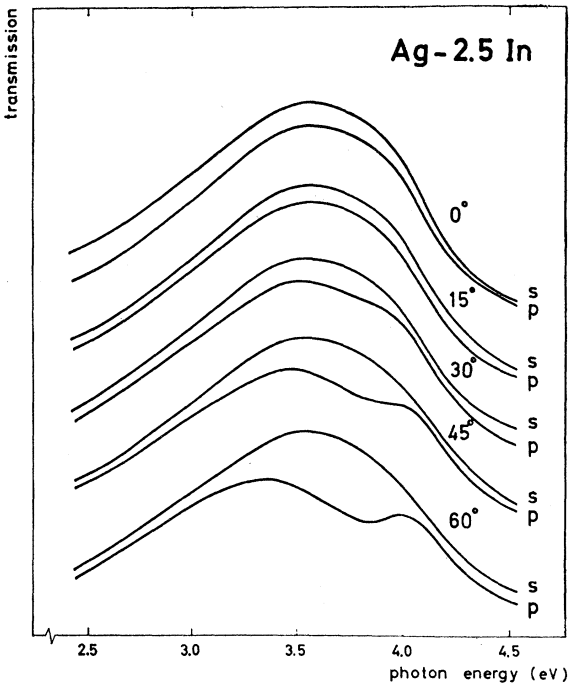

FIG. 7. Transmittance of Ag alloyed with 2.5 at.% In for *s*- and *p*-polarized light at different angles of incidence. The plasma resonance can be seen to occur at about 3.8 eV.

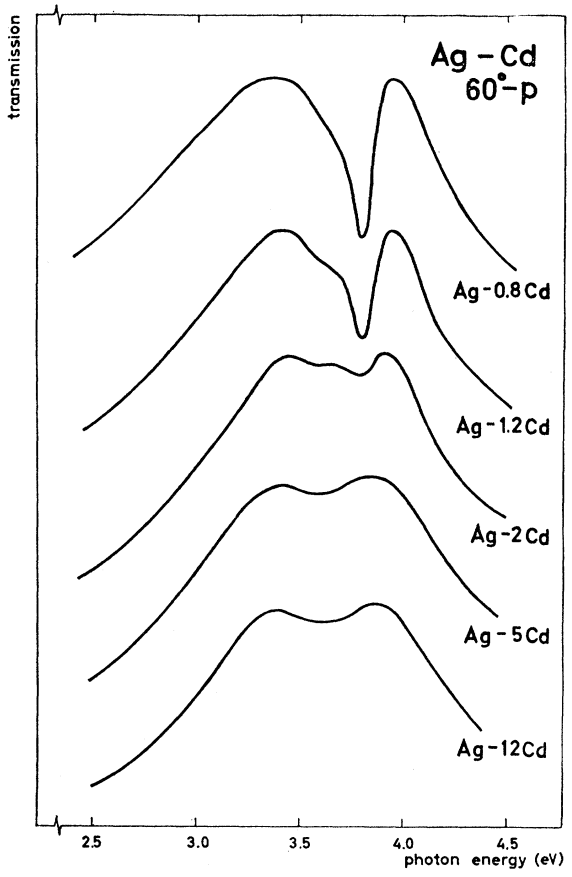

FIG. 6. Transmittance of *Ag*-Cd alloys for *p*-polarized light at 60° angle of incidence. The plasma resonance can be seen to disappear at very low Cd concentration, and at the same time an interband transition develops at 3.6 eV.

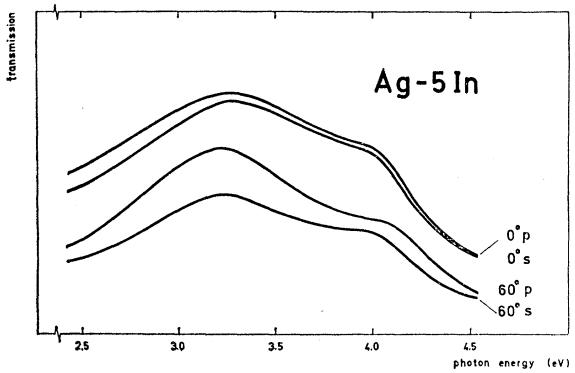

FIG. 8. Transmittance of Ag alloyed with 5 at.% In. No plasma resonance is observed, but an interband transition has developed at 3.6 eV.

V. OBSERVATION OF PLASMA RESONANCE IN PHOTOEMISSION YIELD

The plasmon cannot only decay into a photon. It can also be involved in a single electron excitation. If the plasmon energy is larger than the work function of the specimen, the excited electron may escape through the sample surface. This is observed as an increase in the photoelectric yield. Such observations were made in the 1930's on alkali metals, although the correct interpretation could not be given at that time.[44–46] Photoelectric plasma-resonance experiments have been made on Al films where the resonance occurs around 15 eV.[47–49,18]

For a clean Ag surface, the work function is prohibitively large (4.6 eV) compared with the plasmon energy (3.8 eV) to allow the observation of any photoelectric plasma effect. However, covering the surface with cesium lowers the work function to about 1.8 eV, whereby the escape of plasmon-excited electrons is energetically possible.

Because of the sensitivity of photoelectric measurements on the condition of the surface, all experiments were performed in an ultrahigh vacuum chamber with a working pressure in the 10^{-10}-Torr range. The photoelectric yield as a function of incident photon energy, polarization, and angle of incidence was recorded. Figure 9 shows the experimental results. For s-polarized light, no effect was observed, while p-polarized light gave a distinct increase in the photoelectric yield for 3.78-eV (3280-Å) photon energy and non-normal incidence. Thus, this result is in excellent agreement with the previous results. As can be seen in the figure, there is also a structure appearing around 3.6 eV (3450 Å) close

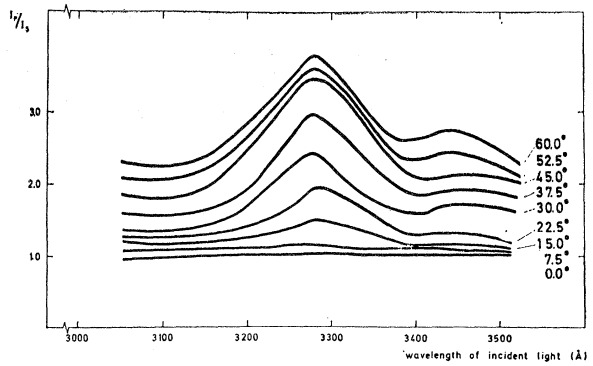

FIG. 9. Quotient of photoemissive yield of a cesiated Ag film for p- and s-polarized light as a function of wavelength. The plasma resonance is observed at 3280 Å corresponding to 3.78 eV. The hump at 3450 Å, corresponding to 3.6 eV, may be due to the decay of the surface plasmon.

to the surface plasmon energy.[39] In principle, the surface modes are nonradiative. However, as previously mentioned, a surface containing irregularities provides momentum-preserving conditions such that optical excitation of surface plasmons is feasible. We therefore tentatively attribute the extra weak structure in the photoelectric yield to the decay of optically excited surface plasmons. A corresponding feature has also been observed in reflectance measurements.[50–52]

A comparison between the optical transmission and the photoelectric results (other than results concerning the plasmon energy) proved to be difficult owing to the strong influence of the Cs layer. Thus, fairly poor agreement was obtained for the damping rates and the angular dependence of the yield.

VI. CONCLUSIONS

The optical-absorption method of studying the plasma resonance offers some advantages compared with other methods, provided that the plasma frequency falls in an easily accessible region of the electromagnetic spectrum. First, the energy determination can be made very accurately. Second, the true plasma oscillation frequency can be obtained by subtracting the contributions from other loss mechanisms such as interband transitions.

The optical plasma-resonance absorption is very pronounced for Ag and occurs at an energy of 3.78 eV. On alloying with Au, the energy is slightly shifted downwards in energy, and at the same time the resonance broadens until it disappears at about 40% Au. This is caused by the shift and broadening of the interband threshold. On alloying with Cd and In, the plasma resonance disappears at a concentration as low as a few atomic percent. This is interpreted as being due to a splitting of the degenerate absorption threshold in pure Ag.

ACKNOWLEDGMENTS

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APPENDIX

According to Fresnel's law,[13] the transmittance of p-polarized light through a slab is given by the formula

$$T=4\epsilon_s^{3/2}\Big/\left|\left(\epsilon_s+\frac{k_2}{\cos\theta}\right)\cos\beta kd - i\left(\frac{k_2\epsilon}{k}+\frac{k\epsilon_2}{\epsilon\cos\theta}\right)\sin\beta kd\right|^2,$$

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where the notations are as follows: $\epsilon=\epsilon_1+i\epsilon_2$ is the dielectric constant of the film; ϵ_s is the dielectric constant of the substrate (infinite thickness); θ is the angle of incidence of the light; λ is the wavelength of incidence light; d is the thickness of the film; $\beta=2\pi/\lambda$, $k_2=(\epsilon_s-\sin^2\theta)^{1/2}$, $k=(\epsilon-\sin^2\theta)^{1/2}$, $\mathrm{Re}k>0$, $\mathrm{Im}k>0$. For a self-supporting film with vacuum on both sides we have $\epsilon_s=1$. If we further assume that $|\epsilon|\ll\sin^2\theta$, we get

$$k\approx i\sin\theta, \qquad k_2\approx\cos\theta,$$
$$\sin\beta kd\approx\beta kd, \qquad \cos\beta kd\approx 1,$$

and the transmittance is given by

$$T=4\Big/\left|2-i\left(\frac{\epsilon\cos\theta}{i\sin\theta}+\frac{i\sin\theta}{\epsilon\cos\theta}\right)i\beta d\sin\theta\right|^2.$$

The assumption $|\epsilon|\ll\sin^2\theta$ leads to

$$\epsilon\cos\theta/\sin\theta\ll\sin\theta/\epsilon\cos\theta,$$

and we get

$$T=\frac{4}{|2+i\beta d\sin^2\theta/\epsilon\cos\theta|^2},$$

or with $\epsilon=\epsilon_1+i\epsilon_2$,

$$T=4\Big/\left[\left(2+\beta d\frac{\sin^2\theta}{\cos\theta}\frac{\epsilon_2}{\epsilon_1^2+\epsilon_2^2}\right)^2+\left(\frac{\beta d\cdot\sin^2\theta}{\cos\theta}\frac{\epsilon_1}{\epsilon_1^2+\epsilon_2^2}\right)^2\right].$$

For a thin film, $d/\lambda\ll 1$, the second term in the denominator can be neglected in comparison with the first one, giving

$$T=4(\epsilon_1^2+\epsilon_2^2)\Big/\left[4(\epsilon_1^2+\epsilon_2^2)+4\beta d\frac{\sin^2\theta}{\cos\theta}\epsilon_2+\frac{\beta^2d^2\sin^4\theta}{\cos^2\theta}\right],$$

where in the last term in the denominator we have put

$$\epsilon_2^2/(\epsilon_1^2+\epsilon_2^2)=1, \quad \text{assuming} \quad \epsilon_1^2\ll\epsilon_2^2.$$

Making the approximation $\epsilon_1=\epsilon_1'(\omega-\omega_p)$ where $\epsilon_1'=(d\epsilon_1/d\omega)_{\omega=\omega_p}$ and assuming that ϵ_2 is constant over the range of interest ($\omega\approx\omega_p$), the transmittance is given by

$$T=\left[4(\omega-\omega_p)^2+\left(\frac{2\epsilon_2}{\epsilon_1'}\right)^2\right]\Big/\left[4(\omega-\omega_p)^2+\left(\frac{2\epsilon_2}{\epsilon_1'}\right)^2+\frac{4\beta d}{\epsilon_1'^2}\frac{\sin^2\theta}{\cos\theta}\epsilon_2+\frac{\beta^2d^2}{\epsilon_1'^2}\frac{\sin^4\theta}{\cos^2\theta}\right].$$

With the notation

$\tau_d^{-1}=2\epsilon_2/\epsilon_1'$ (internal damping constant),

$\tau_r^{-1}=(\beta d/\epsilon_1')(\sin^2\theta/\cos\theta)$ (radiation damping constant),

we finally get

$$T=\frac{4(\omega-\omega_p)^2+\tau_d^{-2}}{4(\omega-\omega_p)^2+(\tau_d^{-1}+\tau_r^{-1})^2}.$$

For a film on a substrate ($\epsilon_s\neq 1$) and with the same approximations made as above, the expression for the transmittance is multiplied with a correction factor B, where

$$B=\frac{4}{\epsilon_s^{1/2}(1+(\epsilon_s-1+\cos^2\theta)^{1/2}/\epsilon_s\cos\theta)^2},$$

and where τ_d^{-1} and τ_r^{-1} now are defined as

$$\tau_d^{-1}=2\epsilon_2/\epsilon_1',$$
$$\tau_r^{-1}=c(\beta d/\epsilon_1')(\sin^2\theta/\cos\theta),$$

where

$$c=\frac{2}{1+(\epsilon_s-1+\cos^2\theta)^{1/2}/\epsilon_s\cos\theta}.$$

If we finally correct for multiple reflections,[53] the transmittance can be written as

$$T=C\frac{4(\omega-\omega_p)^2+\tau_d^{-2}}{4(\omega-\omega_p)^2+(\tau_d^{-1}+\tau_r^{-1})^2},$$

where $C=BA$ and $A\approx 0.996$.

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