Coupled surface plasmons in periodically corrugated thin silver films

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We investigated experimentally the coupled modes of surface plasmons in periodically corrugated thin silver films in a thickness range from 26 to 121 nm. The films studied were self-supporting and corrugated with a period of 1888 nm and amplitudes varying from 6 to 12 nm, depending on the film thickness. Propagation and damping constants of the coupled modes in these films were determined from angle-scan photoacoustic measurements of the resonance absorption of 633-nm photons. Evidence was obtained which indicated that the presence of periodic corrugations was not the primary source of the observed difference between these constants and those in the planar films. The coupling efficiency of an incident photon to the coupled modes evaluated for a constant corrugation amplitude was found to be strongly dependent on film thickness.

I. INTRODUCTION

Resonant coupling of an incident photon to surface plasmons on a periodically corrugated metal surface and subsequent interaction of surface plasmons with surface corrugation structure have been the subject of many recent investigations.[1-6] Most of these works have dealt with a corrugated single surface of an infinitely thick metal and, hence, surface plasmons as a single-surface mode. It is well known[7-10] that in a thin metal film, surface plasmons on the two surfaces interfere with one another and set up coupled two-surface modes if the film is thin enough and bounded on both sides by media with the same or nearly the same refractive index. The mode with a higher frequency (the ω^+ mode) produces a surface charge asymmetric with respect to the midplane of the film with the electric field predominantly normal to the film. The other mode with a lower frequency (the ω^- mode) produces a symmetric surface charge with the electric field essentially parallel to the film. Recently, we reported[11] briefly the first experimental observation of resonant excitation of coupled surface plasmons by photons in a periodically corrugated thin silver film. This observation was made with a 44-nm-thick self-supporting film using a photoacoustic technique.[12] In the present paper, we report experimental results obtained with similar films with thickness varying from 26 to 121 nm. The experimental results are analyzed in terms of the propagation and damping constants of the two coupled modes and the coupling efficiency of incident photons to these modes as a function of film thickness and corrugation amplitude.

Coupled surface plasmons in a planar metal film have been studied rather extensively[13] for the last two decades using electron-energy-loss spectroscopy. However, due to the limited instrumental resolution inherent to this technique, these earlier studies have been restricted to the large wave-number region (the retarded region). In recent years, there has been considerable renewed interest[14-17] in experimental studies of the coupled modes in the retarded region near the light line. The interest arose after it was discovered[18,19] theoretically that the lifetime and, hence, the range of propagation of the ω^+ mode (the ω^- mode) increases (decreases) as the film thickness decreases. Accordingly, these modes have been called in recent studies the long- and short-range surface plasmons (LRSP) and (SRSP), respectively. This interesting property of the coupled modes stems from the fact that the fraction of the electric field associated with the ω^+ mode (the ω^- mode) inside the film becomes small (large) as the film thickness decreases, and hence the rate of internal dissipation also becomes small (large). This theoretical prediction has stimulated several attempts to excite the coupled modes by photons[14-16] and to use the long-range mode to enhance nonlinear optical interactions.[17,20-22] Kuwamura *et al.*,[14] Quail *et al.*,[15] and Craig *et al.*[16] have observed an extremely narrow resonance of this mode in thin silver and aluminum films, exhibiting its long-range propagation, with the resonance width smaller by a factor of 10 than that of the ordinary single-surface mode. These observations have been made by angle-scan attenuated-total-reflection (ATR) measurements using prism coupling. The present experimental study employing grating coupling complements these earlier studies and is aimed at exploring the effects of periodic corrugations on the dispersion and damping of these modes.

II. EXPERIMENT

The self-supporting thin silver films with shallow periodic corrugations were made by following a procedure which was improved somewhat from that used earlier.[11] First, a thin collodion (cellulose nitrate) film was prepared by spreading a drop of collodion solution on the surface of a water pool and allowing the solvent to evaporate. Next, a piece of plastic sheet grating was coated with the collodion film by using the grating to pick up the floating film. After vacuum-evaporating silver on the top of the collodion film, the silvered side of the grating was glued onto a (2×10)-mm^2 aperture of an aluminum frame and then immersed into a pool of amylacetate, a solvent for collodion. The collodion film dissolved away within a few hours, leaving a corrugated silver film separated from the grating mold. The silver film on the frame was carefully lifted from the pool of amylacetate and immediately kept in a small glass container to help the film to dry slowly. We noted that the thinner the film, the slower the speed needed to obtain a rupture-free film.

The best results were obtained with collodion films whose thicknesses were 40±5 nm. It was found that thinner collodion films tended to give gratings exhibiting large resonance half-widths, indicating defects in the gratings. Thicker films, on the other hand, tended to produce gratings whose corrugations were too shallow for detectable plasma resonances. In order to obtain silver films thinner than ~80 nm, a two-step procedure was necessary. First, only the unsilvered side of the grating was exposed to the pool of amylacetate. After a few hours, when the collodion film had dissolved away, the grating was immersed by pouring additional amylacetate into the pool. The first step was necessary to prevent the solvent from attacking the collodion film from the silvered side through pin holes in the silver film and causing serious damage to the silver film prior to separation from the plastic grating. By following these fabrication procedures, we were able to produce periodically corrugated, self-supporting silver films with thicknesses from 26 to 121 nm. Judging from the resonance half-widths, these films were relatively more free from defects than those used in our previous study.[11]

The thicknesses of the silver films were measured by a quartz crystal thickness monitor during the vacuum evaporation. Their spatial periods of corrugation were determined by measuring the diffraction angles of 633-nm photons from a He-Ne laser. The groove separation for all films with different thicknesses fell in the range 1888±3 nm, and no significant difference beyond experimental uncertainty was found among them. The corrugation amplitudes h were determined from the first-order diffraction intensity at normal incidence measured on the incident side of the films with 633-nm photons whose electric field vector was polarized parallel to the grooves. The calculation of h was made by using a theoretical expression[23] for diffraction efficiency of an ordinary shallow metal grating, taking into account the effect of the exit surface of the film on the observed first-order intensity. As shown in Fig. 1, the values of h were found to vary linearly from 5.7 to 11.9 nm with film thickness. An exception was the 76-nm-thick film whose corrugation amplitude was 18.8 nm.

As in our previous study,[11] we have employed the photoacoustic (PA) method[12] for observation of resonant excitation of coupled surface plasmons. We have already demonstrated[24–26] that this relatively new experimental technique, which measures absorption of photons by probing heat generation occurring inside the sample under study, is a useful, convenient experimental one for investigation of plasma resonances in corrugated metal surfaces.

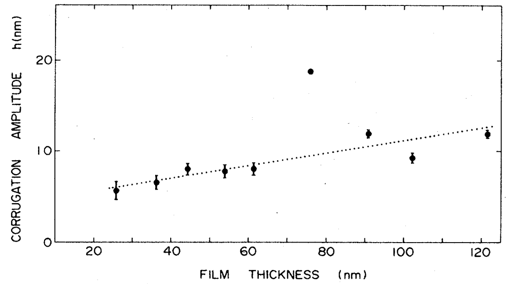

FIG. 1. Corrugation amplitudes h of self-supporting thin silver films as a function of film thickness. Dotted line was drawn as a guide to the eyes.

The PA cell used was of the same type as described before[27] and equipped with an electret condenser microphone and two parallel glass windows. The self-supporting thin film sample was set in the cell with the grooves perpendicular to the plane of photon incidence. Distances between the film and the entrance and the exit windows were 1.5 mm, and the total volume of the cell cavity, filled with air, was 0.19 cm^3, including the space above the microphone diaphragm.

A parallel beam of 633-nm photons from a 1-mW He-Ne laser was directed to the film sample inside the cell through the entrance window after being chopped at 300 Hz by a mechanical light chopper. The PA signal from the film sample was detected as a function of the angle of incidence using a lock-in amplifier. The silver films were thinner than the thermal diffusion length of silver,[12] 0.043 cm at 300 Hz, by a factor of 10^4. They were bounded on both sides by air, which is a good thermal insulator. The PA signals from such thin film samples have been shown[28] to be proportional to the absorptance, the fraction of the incident photon energy dissipated in the film. The PA signals depend also on the film thickness as[29]

$$|Q| = CAF(t), \quad F(t) = (t^2/\mu_s{}^2 + 2gt/\mu_s + 2g^2)^{-1/2}, \quad (1)$$

where $|Q|$ is the PA signal (amplitude), C the instrumental constant depending on thermal and dimensional properties of the cell gas, A the absorptance, t the film thickness, μ_s the thermal diffusion length of the sample material, and g the thermal mass parameter of the cell gas relative to the sample material. In order to compare the PA signal intensities between the films with different thicknesses, it was necessary to take this dependency into account.

III. RESULTS AND ANALYSIS

The PA signals observed with p-polarized incident photons for the films with thicknesses from 26 to 121 nm are presented in Fig. 2 as a function of the angle of incidence. The thickest, 121-nm-thick film exhibits sharp peaks at two different angles, 44.8° and 21.7°. These peaks are due to corrugation-mediated plasma resonances and correspond to the $m = +1$ and $+2$ resonances, respectively, given by the resonance condition $k_{SP} = (\omega/c)\sin\phi + m(2\pi/d)$, where k_{SP} is the surface plasmon wave number, ϕ the angle of photon incidence, d the corrugation

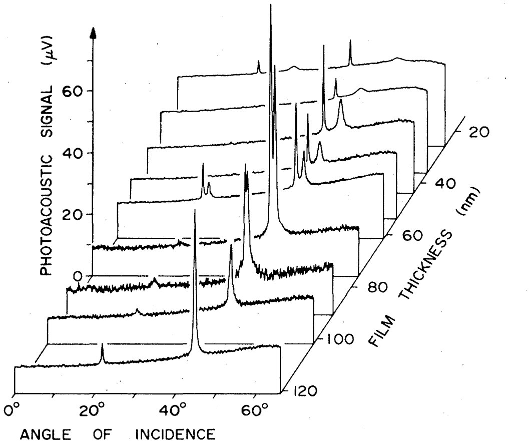

FIG. 2. Photoacoustic signals from periodically corrugated, self-supporting thin silver films observed as a function of the angle of incidence of a p-polarized 633-nm photon. Groove separation $=1888$ nm.

period, ω the angular frequency of the incident photon, and c the velocity of light. These thick film data are similar to our previous results[24,25] obtained for single-surface shallow diffraction gratings of silver.

The thinner films exhibit resonance structures in the same angular regions as exhibited by the 121-nm-thick film. It is seen, however, that each peak for the films thinner than 91 nm splits into two peaks, one at a higher angle and the other at a lower angle. This splitting is due to excitation of coupled surface plasmons resulting from the interaction of plasmons on the two surfaces of the films.

The peaks at the lower and the higher resonance angles correspond to the asymmetric and symmetric modes, respectively, and may be designated as the k^- and k^+ modes according to their wave numbers relative to the single-surface mode. As is seen in Fig. 2, the angular difference between these two modes increases gradually as the film thickness decreases. Also, the resonance widths of the k^- and k^+ modes become narrower and broader, respectively, with decrease of the film thickness, exhibiting their long- and short-range propagation properties. In addition, the intensities of the $m=+1$ resonance near 44° weaken as the film becomes thin, whereas the intrinsic absorption in the off-resonance angular regions becomes considerably stronger. The intensities of the $m=+2$ resonance near 22°, on the other hand, vary drastically from one film to the next and are undetectable in some of the films, reflecting a slight difference in their corrugation profiles. In what follows, we analyze these results with primary interest on the effect of periodic corrugations.

A. Propagation constant and the resonance width

The propagation constants k_{SP} of the coupled modes were determined from the observed resonance angles using the resonance condition given above. From the angular half-widths $\Delta\phi$ at the half-maximum, the resonance half-widths Δk were determined by using the relationship

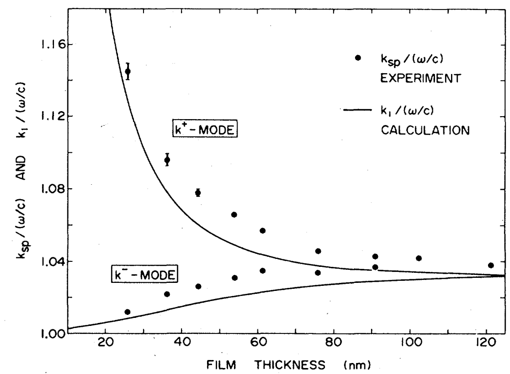

FIG. 3. The propagation constants k_{SP} observed for the k^+ and k^- modes of coupled surface plasmons in periodically corrugated, self-supporting thin silver films as a function of film thickness. The two solid curves represent the real part of the complex wave number k $(=k_1+ik_2)$ for these modes in planar silver films calculated from their dispersion relations.

$$\Delta k=(\omega/c)(\cos\phi_r)\Delta\phi , \tag{2}$$

where ϕ_r was the resonance angle. k_{SP} and Δk thus determined from the angle-scan measurements of the plasma resonances at a fixed photon frequency may be compared with k_1 and $2k_2$, where k_1 and k_2 are the real and imaginary parts of the complex surface plasmon wave number k $(=k_1+ik_2)$ calculated from the dispersion relation with the angular frequency ω as a real number. In the comparison of Δk with $2k_2$, we assume that the plasma resonance line shape is Lorentzian.

In Figs. 3 and 4 we present the results of k_{SP} and Δk, respectively, obtained for the two coupled modes as a function of film thickness. Also plotted in these figures for comparison are k_1 and $2k_2$ calculated from the dispersion relations of these modes in planar films[9] using the literature value[30] of the optical constant of silver $(\epsilon=-16.3+i0.53)$ at 633 nm. It is seen in Fig. 3 that the general behavior of k_{SP} for both the k^+ and k^-

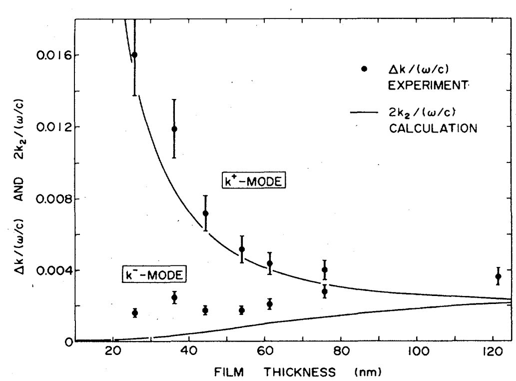

FIG. 4. The resonance half-width Δk observed for the k^+ and k^- modes of coupled surface plasmons in periodically corrugated, self-supporting thin silver films as a function of film thickness. The two solid curves represent twice the imaginary part of the complex wave number k $(=k_1+ik_2)$ for these modes in planar silver films calculated from their dispersion relations.

modes as a function of film thickness agree well with the calculated results of k_1. However, the values of k_{SP} found for both modes are consistently larger than the calculated values of k_1 at all film thicknesses. Results similar to this are found in Fig. 4, where the experimental results of Δk are compared with the calculated results of $2k_2$.[31] Of particular importance in the results in Fig. 4 is that the values of Δk obtained for the k^- mode (the long-range mode) are considerably larger than the calculated values of $2k_2$. This implies that the ranges of propagation of this mode in our corrugated films are considerably shorter than those expected for the planar films. For example, the experimental propagation distance ($=1/\Delta k$) of this mode obtained for our 26-nm-thick film is 63 μm, while the calculated distance ($=1/2k_2$) in the planar film of the same thickness is 840 μm.

For the single-surface mode on an infinitely thick metal surface, it is well known that the presence of periodic corrugations on the surface changes the propagation and the damping of such a mode from what they are on a planar surface. Pockrand and Raether[1,32,33] have found experimentally that k_{SP} and Δk of the single-surface mode on shallow corrugated silver surfaces both increase as the corrugation amplitude h increases and may be represented by

$$k_{SP}=k_{SP,0}+h^2A \ , \tag{3}$$

$$\Delta k=\Delta k_0+h^2B \ , \tag{4}$$

respectively, for $h<60$ nm, where $k_{SP,0}$ and Δk_0 are the propagation constant and the resonance half-width, respectively, on a planar surface, and A and B are positive constants. This experimental observation is explained by the perturbation theory due to Kröger and Kretschmann[34] of the effect of corrugations on surface plasmon dispersion. The results obtained in Figs. 3 and 4 appear to be similar to the single-surface mode results. However, evidence in the present data makes it difficult to ascribe the discrepancies found between the experimental and calculated results in Figs. 3 and 4 to the effect of periodic corrugations in our silver films.

The first evidence is found in the results of our thickest, 121-nm-thick film, in which a single-surface mode was excited. According to Kröger and Kretschmann's expression,[34] the effect of periodic corrugations on both k_1 and $2k_2$ for this film are calculated to be in the order of $0.0001\omega/c$. The differences seen in Figs. 3 and 4 are an order of magnitude larger than these predicted values. The second evidence is found in the experimental results of the 76-nm-thick film, whose corrugation amplitude was nearly twice that of the other films (see Fig. 1). If the discrepancies are due to the effect of periodic corrugations as described by expressions (3) and (4), we would expect this film to exhibit discrepancies nearly four times larger than those exhibited by the othe films. However, such exceptionally large discrepancies are not found in the results in Figs. 3 and 4 for the 76-nm-thick film. These experimental facts suggest strongly that the presence of periodic corrugations is not the major source of the observed discrepancies.

To confirm this inference further, we compare in Fig. 5

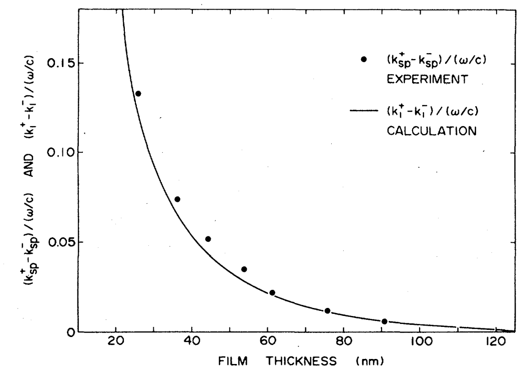

FIG. 5. Observed difference between the propagation constants k_{SP}^+ and k_{SP}^- for the k^+ and k^- modes as a function of film thickness. The solid curve represents the calculated difference between the real parts of the complex wave number k ($=k_1+ik_2$) calculated for the two modes in a planar film.

$k_{SP}^+-k_{SP}^-$, the difference between the two modes, as a function of film thickness, with the calculated values $k_1^+-k_1^-$ for the planar film. If we denote the contribution of periodic corrugations to k_{SP} for the two modes by δ^+ and δ^-, respectively, i.e.,

$$k_{SP}^+=k_{SP,0}^++\delta^+ \ , \tag{5}$$

$$k_{SP}^-=k_{SP,0}^-+\delta^- \ , \tag{6}$$

then

$$k_{SP}^+-k_{SP}^-=(k_{SP,0}^+-k_{SP,0}^-)+(\delta^+-\delta^-) \ . \tag{7}$$

$k_{SP,0}^+-k_{SP,0}^-$ is the difference in k_{SP} between the two modes in a planar film and, therefore, corresponds directly to $k_1^+-k_1^-$. As seen in Fig. 5, the agreement between $k_{SP}^+-k_{SP}^-$ and $k_1^+-k_1^-$ plotted as a function of film thickness is remarkably good, indicating that $\delta^+=\delta^-$ or δ^+ and $\delta^-\simeq 0$. The former case, $\delta^+=\delta^-$, implies that the effect of surface corrugations is the same for the two coupled modes for all film thicknesses. This does not seem plausible because, as we noted before and will show later, the electric fields associated with these two coupled modes are considerably different for different film thicknesses. Hence, the latter special case, δ^+ and $\delta^-\simeq 0$, is inferred, which supports our conjecture made above that periodic corrugations had no appreciable effect on the experimental results in Figs. 3 and 4. A conclusion similar to this is obtained from Fig. 6, where $\Delta k^+-\Delta k^-$, the difference in half-widths between the two coupled modes, is plotted as a function of film thickness and compared with the calculated values $2k_2^+-2k_2^-$ for the planar film.

As mentioned above, we have used the literature values[30] of the optical constants of silver in our calculation of k_1 and $2k_2$. These values were found to give normal incidence transmittance values of planar silver films which were consistent with those measured for our corrugated films (the zero-order transmittance). In order to check if this could be a possible source of the discrepancies in Figs. 3 and 4, we have tried to find values of the optical constants which gave k_1 and $2k_2$ values agreeing

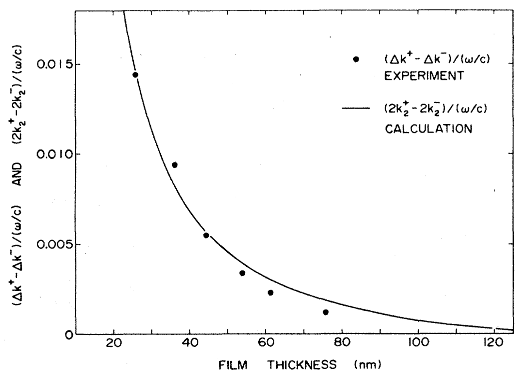

FIG. 6. $\Delta k^+ - \Delta k^-$, the difference in half-widths between the two coupled modes, is plotted as a function of film thickness and compared with the calculated values $2k_2^+ - 2k_2^-$ for the planar film.

with the experimental values of k_{SP} and Δk, respectively. For the single-surface mode observed for the 121-nm-thick film, we found that $\epsilon = 13.2 + i\,0.61$ reproduced the observed k_{SP} and Δk values. This value of ϵ, however, resulted in k_1 and $2k_2$ values for the k^+ mode in the 26-nm-thick film which were nearly twice the observed k_{SP} and Δk values, respectively. For the two coupled modes in thinner films, it was not possible to find optical constants which yielded k_1 and $2k_2$ values consistent, respectively, with the observed k_{SP} and Δk for both modes simultaneously.

These results imply that there exist some other sources which modified the propagation and damping properties of the coupled surface plasmons in our corrugated films from those in planar films. In our films fabricated by the method described above, microscopic random roughness was difficult to eliminate, since most of this existed originally on the surface of the replica grating used as a mold. Inspection of the silver films at various stages of the fabrication process by a scanning electron microscope indicated that the collodion film used as a parting agent diminished some of the random roughness of the grating mold. However, the roughness with relatively long correlation lengths was found remaining in the films, together with some irregularities in the corrugation profile, even after being formed as self-supporting films. These imperfections could be a source of the observed modification of k_{SP} and Δk.

B. Intensity of the resonance absorption

As mentioned before, the PA signals from the silver films shown in Fig. 2 as a function of the angle of photon incidence are proportional to the absorptance A which is defined by $A = 1 - R - T$ in the case of a planar film, where R and T are the reflectance and the transmittance, respectively. The PA signals depend also on the thickness of the film in a manner given by Eq. (1). Before analyzing the experimental results of the resonance intensity, let us verify this latter dependence of the PA signal in our experimental results presented in Fig. 2.

In the off-resonance angular region, the photon absorption in our corrugated films may be supposed to be not too different from those in planar films. Therefore, using the literature values[30] of the optical constants of silver, we first calculated R and T at normal incidence for a self-supporting planar silver film as a function of film thickness. Using the absorptance values A deduced from them, the PA signal $|Q|$ at normal incidence was calculated from Eq. (1) as a function of film thickness. The results of $|Q|$ are plotted in Fig. 7 and compared with the experimental results of Fig. 2 at normal incidence. The calculated results were fitted to the experimental results at the film thickness 121 nm by adjusting the value of the constant C in Eq. (1). As is seen, agreement between the two results over the range of the present film thicknesses is satisfactory. Plotted also in this figure is the calculated absorptance A. It is clearly seen that the observed increase of the PA signal with decreasing film thickness results from a combination of two distinctive effects. One is the increase of the absorptance with decrease of the film thickness, and the other is the thermal effect described by the factor $F(t)$ in Eq. (1). The former, unexpected effect stems from the decrease of R which exceeds the increase of T with decreasing film thickness in the range of present interest. The results in Fig. 7 show that the absorptance observed as the PA signal for the films with different thicknesses may be compared quantitatively with each other if they are corrected for the thermal factor $F(t)$.

To our knowledge, no theoretical study has been made on photoabsorption in periodically corrugated thin metal films due to plasma resonance. For a single-surface system of an infinitely thick metal, the perturbation theory[35] of corrugation-induced photoabsorption has been adapted to interpret the experimental results[24,25] of the plasma resonance absorption of the single-surface mode. This theory, treating the effect of periodic corrugations as a first-order perturbation, predicts that the plasma resonance absorption in an infinitely thick, corrugated metal surface is proportional to h^2, the square of the corrugation amplitude. Owing to the corrugation-induced radia-

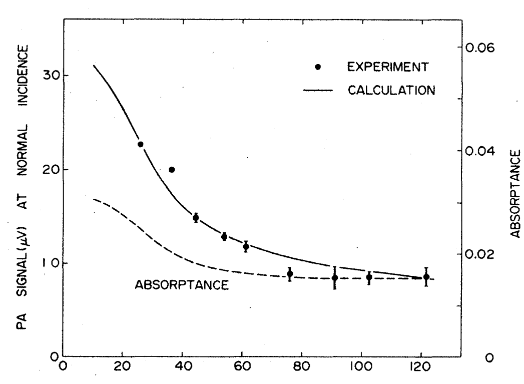

FIG. 7. Photoacoustic signal observed for periodically corrugated, self-supporting thin silver films at normal incidence as a function of film thickness. The dashed curve represents the absorptance A calculated for planar silver films, and the solid curve represents the photoacoustic signal calculated therefrom.

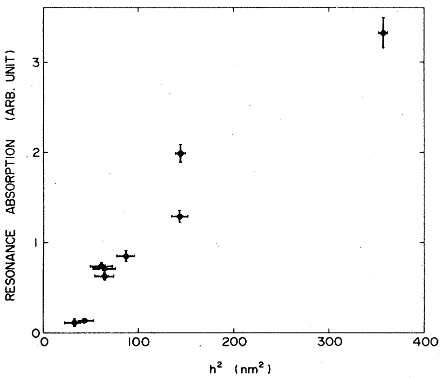

FIG. 8. Integrated, total intensities of the $m=+1$ resonance absorption in periodically corrugated, self-supporting thin silver films as a function of h^2, the square of the corrugation amplitude of the film.

tive damping of the surface plasmon excited on such a surface (the second-order process), there are experimental indications[24] that this theory breaks down even for very shallow corrugations. However, without having any other theoretical guide to the present problem of the coupled two-surface modes, it is of interest to test this simple prediction.

The absorption intensities of the $m=+1$ resonance exhibited by each film near 44° in Fig. 2 were obtained by measuring the sum of the areas under the resonance curves for the k^+ and k^- modes and then correcting for the thermal factor $F(t)$ according to their film thickness. These integrated, total intensities obtained are plotted in Fig. 8 as a function of h^2. It is seen that the resonance intensities exhibit a linear dependence on h^2. However, this dependence is only approximate, and significant deviations from a linear dependence are found in this plot. We assumed that the deviations arose from a dependence of the resonance intensity on film thickness. To test this hypothesis, we plotted in Fig. 9 the resonance intensities divided by h^2 as a function of the film thickness. It is found that these normalized resonance intensities increase with increase of film thickness and begin to decrease after reaching the maximum near 90 nm. This decrease for films thicker than ~90 nm occurs because surface plasmons are excited only on the incidence side of the films. The initial rise of the resonance intensity with increasing film thickness indicates clearly the importance of film thickness as a crucial parameter in the coupling efficiency of an incident photon to coupled surface plasmons.

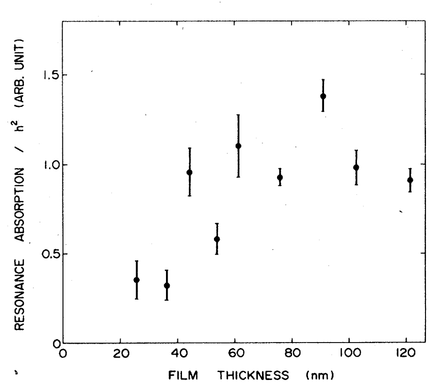

FIG. 9. Integrated, total intensities of the $m=+1$ resonance absorption divided by h^2, the square of the corrugation amplitude of the film.

As seen in Fig. 2, the absorption intensities of the $m=+2$ resonance near 22° are appreciably larger for the films with thicknesses 26, 62, and 121 nm. This indicates that the second harmonic component of the corrugation profile function is larger in these films compared with the other films. This would have affected the observed resonance intensities for the $m=+1$ resonance in these films. It is to be noted, however, that even if the data from these three films are omitted from the results in Fig. 9, the finding made above on the thickness dependence of the resonance intensity is not altered.

IV. DISCUSSION AND SUMMARY

The thicknesses of the silver films used in the present study were of the order of the decay length of the plasmon field inside the film along the film normal. The amplitudes of the periodic corrugations of these films, necessary for the photon-plasmon coupling, were a significant fraction of this decay length and the film thickness. As was found above, characteristics of the coupled modes and their excitations were hence strongly affected by both of these film parameters. Under these circumstances, it was an intriguing exercise to explore the interaction of these modes with the periodic corrugations. However, the corrugation amplitudes attained in the present study were found too small to yield a significant deviation of their dispersion and damping properties from what they were in a planar film. Observed instead were modifications of these properties due to, possibly, microscopic imperfection in the films which were difficult to control in the present experiment.

In contrast to the case of the single-surface mode, little has been studied theoretically on the properties of coupled modes and their excitation by photons in periodically corrugated metal films. Farias *et al.*[5] have recently studied the dispersion of these modes by the method of reduced Rayleigh equations. Their treatment was made neglecting retardation and, hence, is not applicable to the present experimental results obtained in the retarded region. More recently, Agarwal[36] has examined theoretically a multilayered structure in which the coupled surface plasmons may be excited by photons in a planar metal film through grating coupling. In contrast to the case of the present self-supporting films, periodic corrugations were introduced on a dielectric layer which coated a thin planar metal film supported on a dielectric substrate, whose refractive index was similar to that of the dielectric overlayer. He has shown that with such a multilayered structure, efficient coupling of external photons to coupled modes may be achieved. This interesting system may be studied experimentally with the present photoacoustic method as the thermal effects of the dielectric overlayer and substrate on the PA signal generation are well understood.[12,37]

The self-supporting thin metal films with periodic corrugations used in the present study may be applied to many other studies of surface plasmon-related phenomena. For example, the k^- mode (the long-range mode) excited in such a film would be useful in exploring the minigap region[6] of plasmon dispersion because of its narrow-resonance width. Such a film would also be useful for investigation of surface-plasmon-mediated enhancement of nonlinear optical phenomena such as Raman scattering[38] and second harmonic generation.[39] The electric field distributions associated with the two coupled modes in such a film are quite different if the film is sufficiently thin. This is illustrated in Fig. 10, where we depict the time-averaged distribution[8] of the electric field strengh, $\overline{E}$, calculated for 30-, 60-, and 100-nm-thick planar silver films. In this figure, $\overline{E}$ just outside the films are normalized to a constant value. It is seen that, in contrast to the case of the 100-nm-thick film, the $\overline{E}$, fields are quite different for the k^- and k^+ modes both inside and outside the 30-nm-thick film. It would be of interest to investigate the effects of this difference on surface-enhanced optical phenomena involving excitation of these coupled modes. Corrugated films may be fabricated in the form of multilayered structures such as metal-oxide-metal junction. Such self-supporting structures have been used as light-emitting tunnel junctions[40] utilizing corrugation-assisted conversion of surface plasmons to external photons.

In summary, using the photoacoustic method, we have observed coupled surface plasmons in periodically corrugated thin silver films in the thickness range from 26 to 121 nm. The observed variation of their propagation and damping constants with film thickness generally agrees with predictions made for planar silver films. However, significant discrepancies were found which were neither simply explainable by the presence of periodic corrugations in the films nor by the values of the optical constants used in the analysis. The coupling efficiency of incident photons to coupled surface plasmons evaluated for a constant corrugation amplitude was found to be strongly dependent on film thickness and reached a maximum near 90 nm.

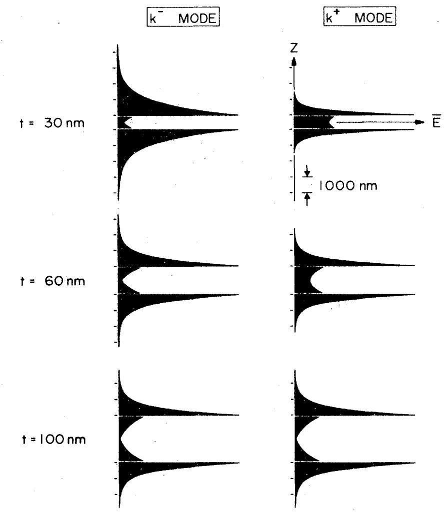

FIG. 10. Time-averaged distributions of the electric field strength $\overline{E}$ associated with the k^- and k^+ modes of coupled surface plasmons in planar silver films with three different thicknesses t. The z axis is in the direction normal to the film surface. Calculations are for the plasmon frequency corresponding to the photon wavelength 633 nm in free space. Note that the scale inside the film is expanded in this figure by a factor 33 over the region outside the film.

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