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Optical constants of evaporated gold films measured by surface plasmon resonance at telecommunication wavelengths

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We report the first measurement of the optical constants of evaporated gold films by using the surface plasmon resonance curve fitting method with an attenuated total reflection device from 16 to 70 nm thickness at telecommunication wavelengths. The results that were obtained by surface plasmon resonance measurement are in good agreement with those obtained by ellipsometry. Until now, optical constants of thin metal films are known to change according to the thickness due to the variation of the electrical resistivity. This phenomenon is also verified in this study by a simple surface plasmon resonance measurement. It is observed that for the gold films of thicknesses of less than 20 nm, the real part of the refractive index increases and the imaginary part decreases with decreasing film thickness. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2902395]

I. INTRODUCTION

Surface plasmon polaritons (SPPs), also abbreviated as surface plasmons (SPs), are collective electron charge oscillations localized at the interface between two different media whose dielectric constants have real parts of opposite signs. They can propagate along the interface and the transmitted power of the SPP mode exponentially decreases due to the intrinsic damping caused by the electron oscillation and also by the radiation damping, which results from the light scattering at the metal interface. The first application of SPPs was to measure changes in the index of refraction and thickness of metal films, which is called the surface plasmon resonance (SPR) technique. Kretschmann[1] obtained the index of refraction of metal films of various thicknesses in the visible range, and Chen and Chen[2] showed that the dielectric constant of a metal film can be determined by comparing thickness values measured at different frequencies. Innes and Sambles[3] measured the optical constants of a thin gold film of about 45 nm thickness over the visible region of the spectrum. There have also been reports of experimental investigations on the optical behavior of ultrathin metal films of thicknesses below 20 nm that use the attenuated total reflection (ATR) technique with an He–Ne laser.[4,5] Recently, SPPs have also been applied, for example, to biomolecular sensing and subwavelength optics[6–8] because SPPs exhibit sensitive responses to the change in the refractive index of bounding media and assist extraordinary transmission of light by field enhancement, respectively. In these applications, the film thicknesses chosen were such that the values of their corresponding optical constants were known, i.e., the ones given in literature.[9]

SPP waveguides have not been considered important because the propagation length of SPPs is limited to the order of several tens or hundred micrometers mainly due to Joule heat losses (intrinsic damping). Since long-range SPPs (LRSPPs), however, were theoretically predicted by Sarid,[10] LRSPP waveguides have motivated great research interests. Research interests in thin metal film SPP waveguides have increased due to the possibility of a long-range transfer of light along the metal strip.[11–13] Many kinds of SPP waveguides, such as a metal slab covered with a dielectric medium, a metal clad, and multiple parallel thin metal films, have been proposed.[14] It has been experimentally demonstrated that LRSPP waveguides typically have propagation losses at telecommunication wavelengths of a few dB/cm with tens of nanometers of thickness.[11] LRSPP waveguides for ultrathin films have been studied because of their low propagation loss, which is attributable to the coupling of LRSPP modes with both sides of a metal layer.

As many researchers are paying a lot of attention to LRSPP sensors and waveguides, the optical constants of thin films become very important because the LRSPP mode is dominantly influenced by the index of refraction of a metal film. The indices of refraction of metal films have been commonly quoted from previous works related to the optical properties of metal films as it is difficult to measure the complex optical constants of thin metal films.[9,16] Either one single value of the refractive index regardless of the thickness of employed films[12] or the values estimated from the ones measured at 2 μm wavelength[15] are adopted. Thèye interpreted that the different optical properties exhibited by gold films of thicknesses of 14.6–21 nm are due to the differences in the crystalline structures of the corresponding films.[16] Yano *et al.*[5] measured the effective optical constants of ultrathin metal films as a function of their thickness by using the He–Ne laser and the ATR technique. Although there have been several studies that reported that the index of refraction changes below a certain thickness owing to the

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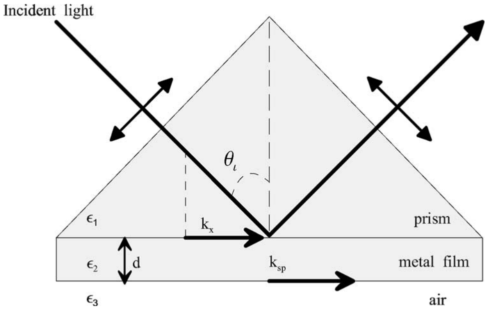

FIG. 1. Configuration of the ATR method.

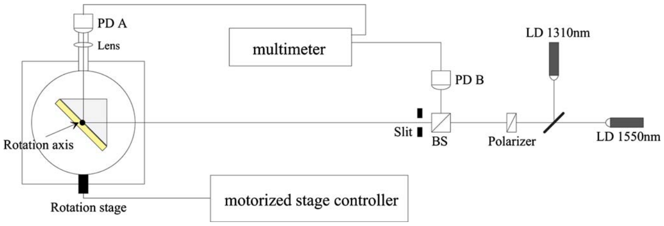

FIG. 2. (Color online) Schematics of experimental setup. LD: laser diode module; PD: photodiode; BS: beam splitter.

variation in the film density and resistivity,[5,17,18] no work on the thickness dependence of the complex optical constants at telecommunication wavelengths has been performed yet. In this study, we show that the optical constants of evaporated gold films can be easily and precisely measured as a function of thicknesses from 16 to 70 nm through the SPR curve fitting method at telecommunication wavelengths.

Section II describes the SPR technique, Sec. III provides the experimental details, Sec. IV gives the results and the discussion, and Sec. V ends the present paper with a conclusion.

II. SURFACE PLASMON RESONANCE

SPR occurs in a metal (ϵ_m)-dielectric (ϵ_d) interface since the real part of the dielectric constant of a metal has a negative value at the optical wavelength range and is detected as a sharp minimum in the totally reflected intensity of light. The dispersion relation for SPs propagating on the metal-dielectric interface can be given by

$$k_{\mathrm{SP}} = \frac{\omega}{c}\sqrt{\frac{\epsilon_m \epsilon_d}{\epsilon_m + \epsilon_d}}, \tag{1}$$

where ω and k_{SP} are the SP frequency and the wave vector, respectively. A SP cannot be directly excited at the metal-dielectric interface due to the dispersion relation of SPs. Therefore, the SPs should be excited by increasing the wave vector of the incident light by using the ATR method. When the incident light enters the prism, the evanescent wave has an increased wave vector and excites SPs at the metal-air interface because of phase matching with SPs, i.e., there is a resonance between the wave vectors on the prism-metal and on the metal-air interfaces. The wave vector can be obtained from the incident angle θ_i and the reflectance can be measured as a function of θ_i. The condition for the excitation of SPs depends on the configuration that is depicted in Fig. 1. The intensity of the reflected light can be described by Eq. (2), which are called Fresnel's equations, for the *p*-polarized incident light on a three-layer system as a function of θ_i.[19] The dielectric constant of a metal can be represented as $\epsilon_2 = \epsilon_{2\mathrm{real}} + i\epsilon_{2\mathrm{imag}}$ and the reflectivity R is given by

$$R = |r_{123}|^2 = \left| \frac{r_{12} + r_{23}\exp(2ik_{z2}d)}{1 + r_{12}r_{23}\exp(2ik_{z2}d)} \right|^2, \tag{2}$$

where d is the thickness of the metal film,

$$r_{ij} = \frac{\epsilon_j k_{zi} - \epsilon_i k_{zj}}{\epsilon_j k_{zi} + \epsilon_i k_{zj}} \quad \text{for} \quad i,j = 1,2,3$$

and

$$k_x = \frac{\omega}{c}\sqrt{\epsilon_1}\sin\theta_i, \quad k_{zj} = \sqrt{\epsilon_j\left(\frac{\omega}{c}\right)^2 - k_x^2}$$

$$\text{for} \quad j = 1,2,3.$$

According to these equations describing the phenomenon of SP excitation, the refractive index and the thickness of a metal film can be obtained from the measured reflectivity through a fitting process with Eq. (2). The complex index of refraction instead of the complex dielectric constant is used. It is obtained from the relations $\epsilon_{\mathrm{real}} = n^2 - k^2$ and $\epsilon_{\mathrm{imag}} = 2nk$, where n and k are the index of refraction (the real part of the refractive index) and the extinction coefficient (the imaginary part of the refractive index), respectively.

III. EXPERIMENTAL DETAILS

For the convenience of experiment, the three-layer Kretschmann configuration consisting of a prism-gold film-air is considered. In our experiment, gold (99.999%) thin films were deposited on BK7 slide (18×32 mm^2, 1 T) substrates through thermal evaporation at a pressure of between 4×10^{-7} and 1×10^{-6} Torr. A thin chromium (with thickness ≤ 1 nm) underlayer could not be used to enhance the adhesion of gold atoms because there remains the question of whether the index of refraction of such a thin chromium is of a bulk or of a thin film character. The deposition rate was 0.5 Å/s at room temperature and the thickness of prepared films was measured to be between 16 and 70 nm. A right angle prism coupled to the BK7 slide using the index-matching fluid was mounted on a motorized rotation stage (Physik Instrumente C-844). The rotation angle step of the prism is 0.01°. The reflection plane of the prism is placed on the rotation axis of the stage. A lens was employed because the beam spot of the incident light was larger than the active area of the photodiode (PD). The experimental arrangement is shown in Fig. 2 and all the experiments were performed at room temperature.

The incident light (Lasermax diode laser modules) was TM (*H* field is perpendicular to the plane of incidence) polarized. The intensity of the reflected light with respect to the normal plane at the air-prism interface was measured by PD B (Newport 818IR) and the reference angle of the prism (45°) was obtained. Then, the intensity of the reflected light (Newport 2835-C) from the gold film was measured by PD A

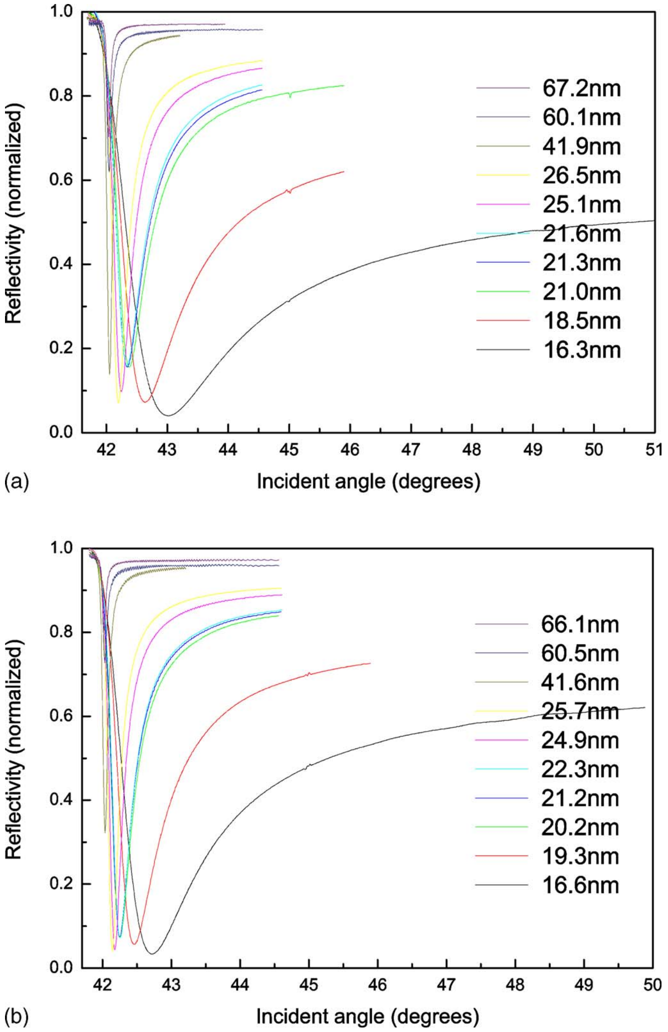

FIG. 3. (Color online) Reflectivity of gold films for various thicknesses as a function of the incident angle measured with a ATR device: (a) $\lambda = 1310$ nm and (b) $\lambda = 1550$ nm.

and normalized with the intensity of reflected light at the bare BK7 slide. In Fig. 3, the measured reflectivity versus incident angle θ_i for films of various thicknesses at the incident wavelengths of 1310 and 1550 nm is shown. We leave out the reflectivities for the angles below the critical angle for the total internal reflection since they are greater than unity. Small fluctuations in the reflectivity in the vicinity of 45° are detected due to the multiple reflections between the prism and the PD.

IV. RESULTS AND DISCUSSION

The thicknesses of gold films were also measured by ellipsometry (J. A. Woollam M-2000V) for the purpose of confirming the validity of the SPR technique, and the thickness values obtained by the two methods are compared to each other in Table I. The measured thicknesses of the gold films by the two methods have maximum differences of about 5.5% at the incident wavelengths of 1310 and 1550 nm. It stands to reason that the optical constants of gold films measured by using the SPR method are precise for the film thicknesses of 16–70 nm.

TABLE I. Thicknesses of gold films measured by the SPR technique and by ellipsometry.

$\lambda = 1310$ nm (nm)	$\lambda = 1550$ nm (nm)	Ellipsometry (nm)
16.3	16.6	16.1
18.5	19.3	18.3
21.0	20.2	20.5
21.3	21.2	21.1
21.6	22.3	21.7
25.1	24.9	24.7
26.5	25.7	26.2
41.9	41.6	41.2
60.1	60.5	61.8
67.2	66.1	66.6

In Figs. 4(a) and 4(b), the measured ATR curves and their fits for gold films of thicknesses of 16.3, 41.9, and 67.2 nm at $\lambda = 1310$ nm and 16.6, 41.6, and 66.1 nm at $\lambda = 1550$ nm are depicted. The optical constants and thicknesses of the gold films can be determined by a least-squares

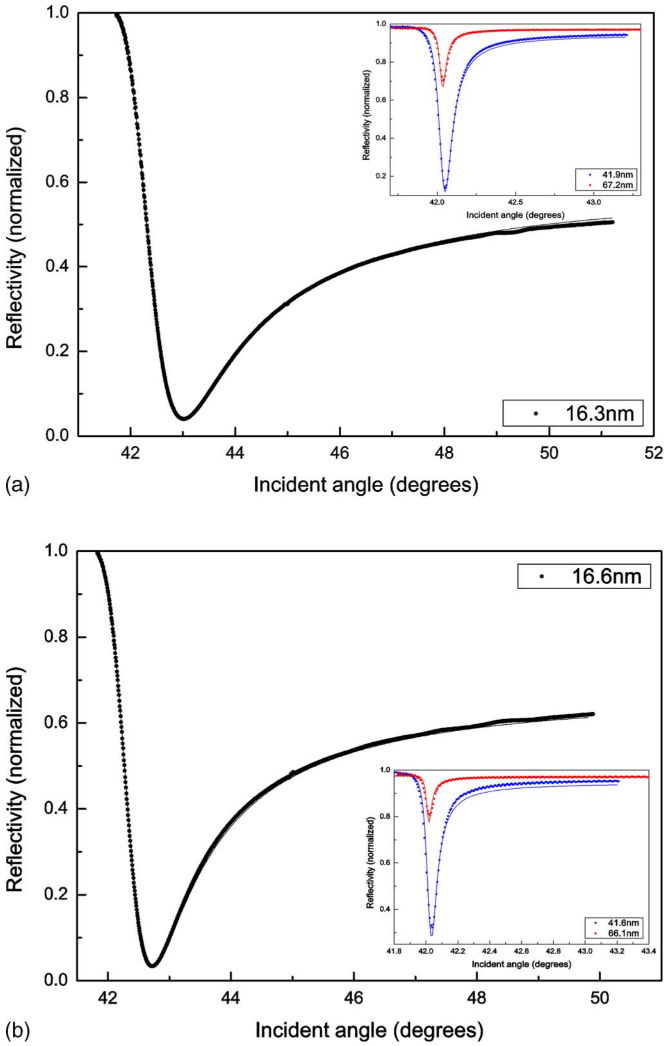

FIG. 4. (Color online) Measured ATR data (dots) and their fitted curves (solid line) for the films with thicknesses of (a) 16.3, 41.9 (in blue in the inset), and 67.2 nm (in red in the inset) at $\lambda = 1310$ nm and (b) 16.6, 41.6 (in blue in the inset), and 66.1 nm (in red in the inset) at $\lambda = 1550$ nm.

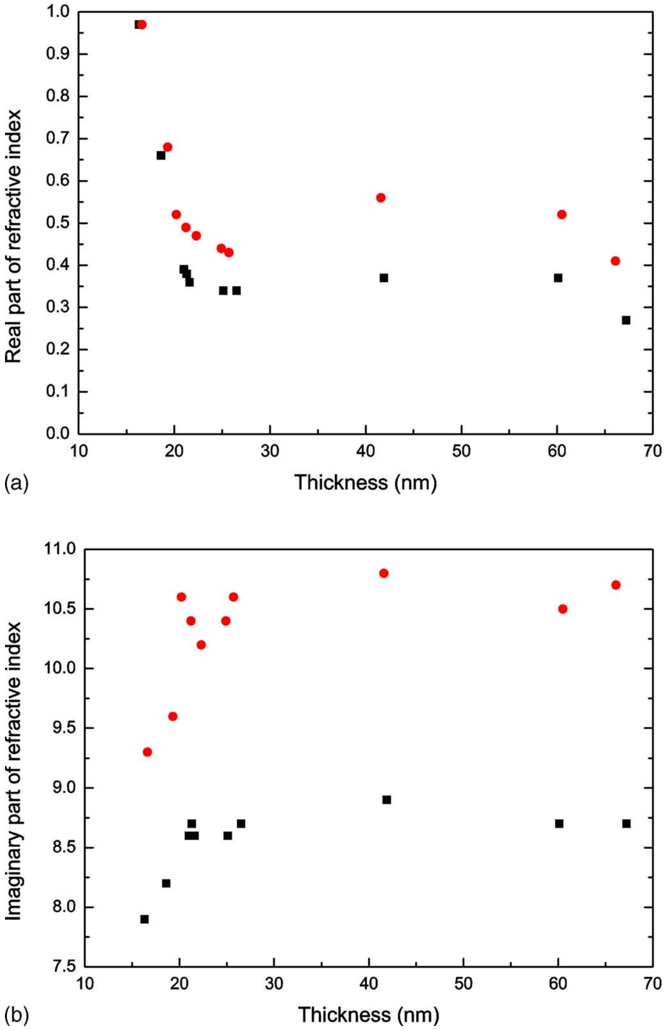

FIG. 5. (Color online) Refractive indices of gold films for the incident lights with wavelengths of 1310 (filled black square) and 1550 nm (filled red circle): (a) the real part of the refractive index and (b) the imaginary part of the refractive index.

fit with adjustable parameters, and in this analysis, the grid-search method is employed as the fitting program.[20]

The refractive indices of the gold films obtained by fitting the data displayed in Fig. 3 are shown in Figs. 5(a) and 5(b). The measured values for the film of 40 nm thickness, $0.37-i8.9$ at $\lambda=1310$ nm and $0.56-i10.8$ at $\lambda=1550$ nm, are in good agreement with those given in the literature. However, the fitted values of the optical constants for the film thicknesses in the vicinity of 66 nm, $0.27-i8.7$ at $\lambda=1310$ nm and $0.41-i10.7$ at $\lambda=1550$ nm, deviate from the values given in Refs. 21 and 22, which are $0.31-i9.0$ at $\lambda=1310$ nm, $0.37-i11.0$ at $\lambda=1550$ nm, and $0.42-i8.4$ at $\lambda=1310$ nm, $0.56-i9.8$ at $\lambda=1550$ nm, respectively. This discrepancy may have its origin in the fewer number of data points taken for thicker films due to the fixed value of the relatively large rotation angle step (0.01°) of the prism. A strange behavior of the imaginary part of the refractive index is observed in the thickness range from 20 to 25 nm, which may have resulted from the fitting errors as the absoprtion of light by the gold films at the reflectivity minima is reduced due to the impurities present in the films.

Figures 5(a) and 5(b) illustrate the general behavior of the optical constants as a function of film thickness. The optical constants reported in the literature, when compared to the present measured values, have the upper[22] and lower limits,[21] which indicate the film and the bulk properties, respectively. It is clear that for thicknesses larger than 20 nm, there is good or reasonable agreement between our experimental data and the values given in the literature; however, below 20 nm, they differ from each other. The values of optical constants are affected very much by the measuring conditions; even so, the optical constants for thicknesses below 20 nm totally escape from those listed in the literature. Therefore, it is obvious that for these thicknesses, the real and imaginary parts of optical constants exhibit sudden increment and decrement, respectively, with decreasing film thickness. This phenomenon was reported by Reale[18] and Ohring[23] and explained by the fact that the film density gets lower below a certain thickness relative to that of the bulk due to many vacancies and micropores. Thus, for these film thicknesses, one is convinced that the optical constants measured by the SPR technique at telecommunication wavelengths are in good agreement with the values listed in the literature.

V. CONCLUSION

For the first time, we have exactly measured the variation of the refractive index of gold film as a function of film thickness from 16 to 70 nm by using the SPR method at telecommunication wavelengths. The thicknesses that were measured by using the SPR technique are in good agreement with those measured by ellipsometry and with the values listed in the literature. Moreover, our results show that the optical constants of thin gold films with thicknesses below 20 nm deviate from the values given in the literature. The real part of the optical constant increases with decreasing thickness, while the imaginary part decreases. Thus, it is quite clear that the SPR technique with a ATR device is confirmed to be a simple method for the measurement of the complex optical constants and film thicknesses from 16 to 70 nm.

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