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Dielectric constants and thickness of metallic film using attenuated total reflection technique

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Abstract Gold films of thickness 40 nm to 50 nm are deposited on ultrasonically cleaned glass slides by thermal evaporation under vacuum of 10^{-6} Torr and at the evaporation rate 1 nm/sec. The gold-coated slides are brought into optical contact with the prism using an index matching fluid. For excitation of surface plasmons a He-Ne laser source of *P* polarized mono-chromatic beam (λ = 633 nm) is employed. The prism and the sample are placed on a $\theta-2\theta$ rotation stage. The reflectance minima R_p in gold as a function of the thickness is plotted. It shows that R_p becomes nearly equal to zero at $d \sim 45.75$ nm. The gold film of approximately 45 *nm* thick is taken and subjected to further surface plasmon resonance (SPR) study. The resulting SPR curve was well fitted by Fresnels theory and the value of refractive index (*n*), extinction co-efficient (*k*) and thickness (*d*) are found to be 0.2 ± 0.03, 3.2 ± 0.05 and 45 ± 2 nm respectively. The other observations from this experiment includes : the variations of real and imaginary parts of metal dielectric constants with film thickness and SPR curve at different thickness *etc*. Our results show good agreements with similar results of silver films and agree with the data obtained by ellipsometric measurements

Keywords Dielectric constants and thickness, metallic film, attenuated total reflection

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1. Introduction

Optical dielectric constants and thickness of thin films are two important physical parameters for optical uses of metallic films. These two parameters depend on both the chemistry and the structure of the film. Ellipsometry is a well-established method for the characterization of both nonabsorbing and moderately absorbing dielectric films [1]. However, for highly absorbing films, such as metal films, simple ellipsometry is not normally the appropriate method for a complete film characterization (*n*, *k*, *d*), because the light reflected at the back surface of the metal film and returning to the front surface is very weak compared with the light reflected directly at the metal surface. Surface plasmon Resonance (SPR) with attenuated total reflection (ATR) is a well known technique for optical characterisation of metallic as well as organic overlayers. Their determination involves optical excitations of surface plasmons followed by appropriate curve fitting procedure with the Fresnel's reflection theory. Surface Plasmons (SP) are very elementary excitations of metal-dielectric interfaces. The dispersion relation of the SP is obtained by solving Maxwell's equations with special boundary conditions, such as the finite thickness of the metal and the dielectric constant. To allow the excitation of the surface plasmon oscillations the real part $\varepsilon_r(\omega)$ of the dielectric function $\varepsilon(\omega)$ has to be negative *i.e.* $\varepsilon_r(\omega) < -1$ and the imaginary part $\varepsilon_i(\omega)$ should be very small *i.e.* $\varepsilon_i(\omega) < |\varepsilon_r(\omega)|$, a condition which is fulfilled in metal. Gold, silver, copper, and aluminum are some of the metals which meet these conditions for visible light at least for larger wavelengths. In the neighborhood of so called 'plasma angle' the conditions can be matched and thus the SP can be excited. Surface excitations (surface plasmons) do not couple directly to the external photons because energy and momentum can not be conserved in this transition. SPR technique with attenuated total reflection (ATR) has been widely used to excite the surface plasmons ever since it was introduced in 1968 by Otto [2] and later developed by Kretschmann [3]. The dielectric prism with refractive index *n*, lowers the photon dispersion curve to $\omega = Kc/n$, but does not effect appreciably the surface mode. At total reflection angle θ, $1/n < \sin\theta < 1$, the evanescent photon waves from the prism, with parallel momenta $K_x = n(\omega/c)\sin\theta$, couples to the surface plasmons in the region of K : $\omega/c < K < (\omega/c)n$. Under the optimum coupling, the total incident energy is dissipated through

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diffusion of plasmon wave in metal, the reflection coefficient falls to zero, and the surface plasmon resonance (SPR) appears as a dip in the reflected intensity as a function of the angle of incidence. Reflectivity of glass- metal- air system can display a sharp minimum owing to strong absorption. The reflectivity can even vanish at the plasmon angle for optimum thickness of the film. This decrease in reflectivity is called ATR. The resulting SPR curve of three layered system (prism-metal-air) may be well fitted by Fresnel's theory so as to determine the metal layer parameters -thickness and dielectric constants. In addition to this, SPR effect is often used to construct highly sensitive optical sensors – chemical [4,5] gas [6], bio and temperature [7] sensors. Examples are the monitoring of protein adsorption and immune reactions at a silver surface. Optical methods offer the advantage of being noncontact, nondestructive, fast, precise, sensitive and reproducible

2. Optical constants of metal film (three layered system)

Let us consider an incident TM light of frequency ω in a prism-metal -vacuum (air) configuration as shown in Figure 1.

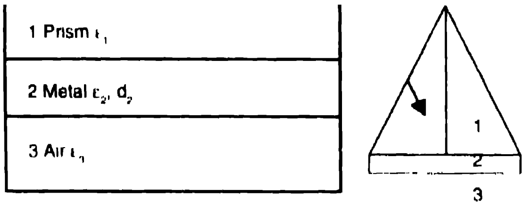

Figure 1. A Schematic diagram showing the Kretchmann configuration of prism-gold air system

When the light is incident at an angle θ_t, it can couple directly with the surface plasmon wave (SPW) of the metal-vacuum interface, here $\theta_t = \sin^{-1}(1/n)$, and n is the index of refraction of the prism. The coupling takes place when ω and $K_x(\theta)$ of the incident light match with the frequency ω and the complex wave vector K of the SPW. Here $K_x(\theta)$ is the component of the incident wave vector parallel to the interface. Since K_x is real and K is complex, only an approximate match can be reached for $K_x(\theta) = \mathrm{Re}(K)$ at

$$\theta = \theta_{ATR} = \sin^{-1}[\mathrm{Re}(K)c/n) = \sin^{-1}[Kxc/n\omega]. \quad (1)$$

At θ_{ATR}, the total reflection of the incident light is attenuated. The coupling can be analyzed by calculating the reflectance R of the light given by

$$R = \frac{\gamma_{12} + \gamma_{23}\exp(i2K_{z2}d)}{1 + \gamma_{12}\gamma_{23}\exp(i2K_{z2}d)} \quad (2)$$

The parameters are well-defined in Ref. [3]. With θ close to θ_{ATR}, the reflectance R can be approximately expressed as a function of θ as

$$R(\theta) = 1 - \frac{4\,\mathrm{Im}(K^0)\,\mathrm{Im}(K^R)}{[K_x - \mathrm{Re}(K)]^2 + \mathrm{Im}(K)^2}, \quad (3)$$

with $K = K^0 + K^R$. The value of K^0 and K^R are defined in th Ref. [8] Here, K is the complex wave vector of the surface plasmo wave (SPW) in the Kretschmann configuration, and K^0 is th complex wave vector of the SPW and K^R is the perturbation t k^0 in the presence of the prism.

The reflectance has a Lorentz dip at θ_{ATR} with a half widt W_0 and minimum reflectance R_{min} given by

$$W_\theta = 2\,\mathrm{Im}(K)\cos(\theta_{ATR})\,c/n\omega, \quad (4$$

$$R_{min} = 1 - 4\eta/(1+\eta)^2, \quad (5$$

where

$$\eta = \mathrm{Im}(K^0)/\mathrm{Im}(K^R). \quad (6$$

With above equations, one can proceed to determine th dielectric constant ε_2 and thickness $d_2$of a metal film from th measured R *versus* θ curve.

3. Experiment (deposition technique and characterisation)

Gold films of thickness 40 nm to 50 nm are deposited o ultrasonically cleaned glass slides by thermal evaporation unde vacuum of 10^{-6} Torr and at the evaporation rate 1 nm/sec. Th vacuum system used is a standard combination of a rotar mechanical pump and an oil diffusion pump. A crystal fil thickness monitor is used to ensure the controlled rate o evaporation. This instrument measures the change in oscillato frequency of a crystal (due to the change in mass) placed nex to the sample. The amount of metal deposited on the crystal i proportional to the change in oscillating frequency. Th instrument can be calibrated to read accurate values for the rat of deposition and the thickness.

The gold coated slides are brought into optical contact wit the prism using an index matching fluid. For excitation of surfa plasmons, a He-Ne laser source of P polarised mono-chromat beam (λ = 633 nm) is employed. The prism and the sample ar placed on a $\theta - 2\theta$ rotation stage which is driven by microprocessor controlled stepping motor. The details of th experimental set up is given in Figure 2.

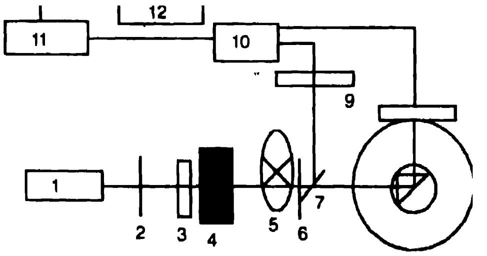

Figure 2. Optical set-up for SPR measurement [1-He-Ne Lase 2-Aperture, 3-Neutral density filter, 4-Polariser, 5-Beam choppe 6-Aperature, 7-Beam Splitter, 8-($\theta - 2\theta$) prism table, 9-photo detecto 10-Lock-in-amplifier, 11-microprocessor, 12($\theta - 2\theta$ drive)].

When optical coupling is optimum, the total incident energ is dissipated through diffusion of the plasmon wave in gold fil and the reflection intensity falls to zero. The reflectance minima R_p in gold as a function of the angle of incidence for different thicknesses are plotted in Figure-3. It shows that R_p becomes nearly equal to zero at $d \sim 45.75$ nm.

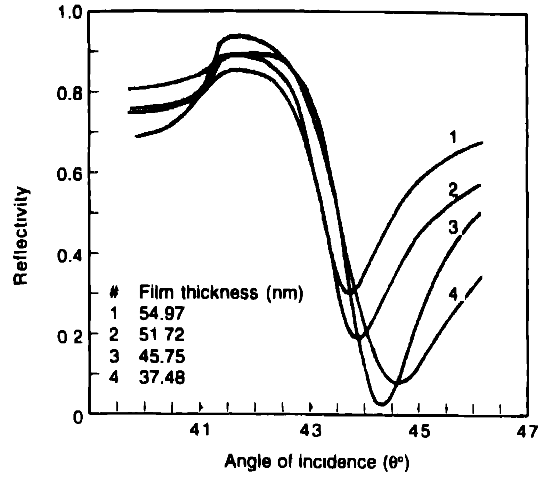

Figure 3. Measured reflectivity of gold films of different thickness as the function of angle of incidence

The gold films of approximately 45 nm thick is taken and subjected to further SPR study. The resulting SPR curve was well-fitted by Fresnel's theory and the values of n, k and d for gold are found to be 0.2 ± 0.03, 3.2 ± 0.05 and 45 ± 2 nm respectively. The thickness dependence resonance position of the surface plasmons at the metal/air interface is shown in Figure 4. A kind of inverse relation exist between them.

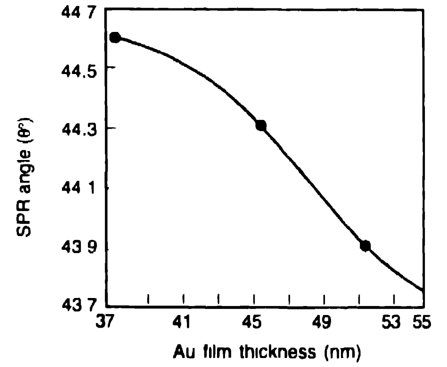

Figure 4. Variation of SPR angle with Au film thickness

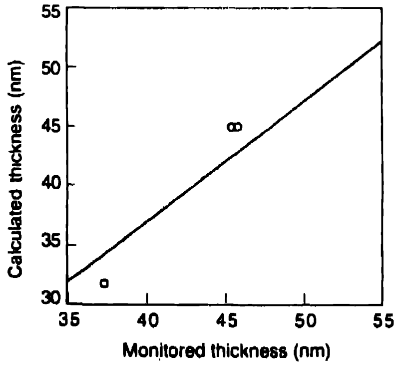

Figure 5. Variation of monitored thickness with calculated thickness.

Thinner films are known to result in more radiation losses and hence shallower and broader resonance curves, where as thicker films result in increased intrinsic losses. The monitored film thickness of the gold films are plotted against the calculated film thickness in Figure 5. The relation is linear with slope approximately unity.

4. Conclusion

In the work, an attempt is made to characterize the optical properties (dielectric constant and thickness) of vacuum deposited thin films of gold. Basically, it is understood that the dielectric constant of metal films is a function of the incident light frequency [9] and that film geometry can be a factor only when the film thickness is below a certain value [10]. It is important to note that the value of gold dielectric constant for example is at variance for the same film thickness among different reports in the literature. This is not unusual since solids exhibit different structural and morphological properties when prepared as thin films. Such variations can be ascribed to several reasons which include the purity of the starting material, the background pressure of the vacuum chamber, the rate of evaporation. All these parameters may lead to variation in the smoothness of the layer, the size of the crystallized particles and the adhesion of the layer to the substrate. Consequently, such variations may result in differing properties of the material when its thickness or preparation conditions are varied.

The fitting method is suitable for determining optical parameters of metal films (Au) for a thickness range 100 to 1000A^0 where the leakage loss and the joule loss of SPW are comparable. If the film is thinner, coupling is increased and the reflection minimum becomes broader and shallower; if the film is thicker the coupling becomes weaker and the reflection minimum decreases. As a consequence, there is an upper and lower thickness limit for the application. However for thickness greater than 1000 A^0, the dielectric constants of metallic thin films can be derived using the expressions given under the Section 2.

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