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Determination of Optical Constants of Anisotropic Thin Film from Attenuated-total-reflection

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Abstract

The broadening of the half-width and the change of the reflection minimum of the attenuated-total-reflection (ATR) curve which is due to the anisotropic thin film are demonstrated. The optical constants and the thickness of the anisotropic film are calculated.

Keywords: Anisotropy, Surface plasma, Attenuated-total-reflection

1. Theory

In recent years, thin films with anisotropy characteristic have been applied to many optical devices[1,2]. By various incident angles of vapor flux in deposition, thin films reveal anisotropy with one of the principal axes in the direction of columnar growth. In this paper, we offer a sensitive and convenient method by ATR device to obtain the optical property of the anisotroic thin films including optical constants, column growth angle and thickness.

Optical excitation of surface plasma wave, SPW, in the attenuated-total-reflection, ATR, of the Kretschmann configuration is a very powerful device to obtain optical constants of thin metal film[5,6,7] .Considering a prism-metal film-vacuum configuration as shown in Figure 1, SPW will be

excited when the incident angle of p-polarized light of frequency ω is larger than the critical angle. The reflected intensity R can be given as follows by Fresnel's equations for the three layer system 0/1/2 (for 0: prism; 1: metal film of thickness d; 2: air)

$$R=\left|\frac{r_{01}+r_{12}\exp(2ik_{z1}d)}{1+r_{01}r_{12}\exp(2ik_{z1}d)}\right|^2 \quad (1) \quad \text{with} \quad r_{ik}=\frac{\dfrac{k_{zi}}{\varepsilon_i}-\dfrac{k_{zk}}{\varepsilon_k}}{\dfrac{k_{zi}}{\varepsilon_i}+\dfrac{k_{zk}}{\varepsilon_k}} \quad (2)$$

where ε_i and k_{zi} are the dielectric constants and the wave-vector components perpendicular to the interface in medium i.

The dielectric constant of the metal can be represented further as a complex form: $\varepsilon_1=\varepsilon_{1r}+i\varepsilon_{1i}$. Under the conditions $|\varepsilon_{1r}|\rangle\rangle 1$ and $|\varepsilon_{1i}|\langle\langle|\varepsilon_{1r}|$, the reflection R has an approximately inverted Lorentzian type that is given as[7]:

$$R=1-\frac{4\Gamma_i\Gamma_{rad}}{\left[k_x-\mathrm{Re}\left(k_{sp}^0\right)\right]^2+(\Gamma_i+\Gamma_{rad})^2} \quad (3)$$

where $k_{sp}^0=k_{x0}+\Delta k_x$ and $k_x=n\dfrac{\omega}{c}\sin\theta$. k_x is the parallel component of the incident wave vector through the prism, and the term Δk_x can be approximated as[7]

$$\Delta k_x=\left[\frac{\omega}{c}\frac{2}{1+|\varepsilon_{1r}|}\left(\frac{|\varepsilon_{1r}|}{|\varepsilon_{1r}|-1}\right)^{3/2}\right]\exp\left(-2|k_{x0}|d\right)r_{01}(k_{x0}) \quad (4)$$

In Eq. (4), k_{xo} is the complex wave vector of SPW on the plane surface of a semi-infinite metal with the dielectric function ($\varepsilon_1 = \varepsilon_{1r} + i\varepsilon_{1i}$) adjacent to a medium ε_2 as air or vacuum and which can be expressed as

$$k_{xo} = (\frac{\varepsilon_1\varepsilon_2}{\varepsilon_1 + \varepsilon_2})^{1/2}\frac{\omega}{c} \tag{5}$$

The intrinsic damping in Eq. (3), Γ_i, represents the Joule loss in the metal. The radiation damping, Γ_{rad}, represents the energy lost by back-coupled radiation. These two quantities can be expressed as $\Gamma_i = \mathrm{Im}(k_{x0})$ and $\Gamma_{rad} = \mathrm{Im}(\Delta k_x)$.

From Eq. (3), the half-width of the ATR curve is equal to $2(\Gamma_i + \Gamma_{rad})$, and the reflection minimum occurs at the incident angle θ which satisfies

$$k_x = n\frac{\omega}{c}\sin(\theta) = \mathrm{Re}(k_{x0} + \Delta k_x) \tag{6}$$

and the reflection minimum is depend on the match condition of Γ_i and Γ_{rad}

$$R_{\min} = 1 - \frac{4\Gamma_i\Gamma_{rad}}{(\Gamma_i + \Gamma_{rad})^2} \tag{7}$$

Thus by measuring the half-width and the reflection minimum of the ATR curve, we are able to calculate the optical constants of a metallic film. In this paper, we use the method developed by W.P.Chen and J.M.Chen[7] to determined the n, k, d (n, the index of refraction; k, the index of extinction) of thin silver films. From Eq. (3), the half-width of the ATR curve is equal to $2(\Gamma_i + \Gamma_{rad})$, and the reflection minimum occurs at the incident angle θ which satisfies:

$$k_x = n\frac{\omega}{c} sin\theta = Re(k_{x0} + \Delta k_x) \tag{8}$$

and the reflection minimum is depend on the match condition of Γ_i and Γ_{rad}:

$$R_{min} = 1 - \frac{4\Gamma_i\Gamma_{rad}}{(\Gamma_i' + \Gamma_{rad})^2} \tag{9}$$

With Eq. (1) -(7), we can use the iteration process[7] to determine the optical constants of thin silver film. Although there are two sets of solutions obtained, one of them can be easily eliminated by judging the thickness we have coated.

In order to measure the anisotropy thin film character by ATR device, we coat the anisotropic SiO_2 thin film on the silver film.. If we change the incident angle of vapor flux, the column structure (principal axes) will make an angle with the normal of the substrate. The theory will show that the ATR curve is shifted and broaden by the tilt angle sensitively. By curve fitting method, the index of refraction in principal axes n'_x, n'_y, n'_z, the tilt angle ϕ and the thickness of the SiO_2 thin film will be derived.

In Kretschmann configuration, we coat SiO_2 on silver thin film. It becomes a four-layer system. The reflection can be derived from Fresnel's equations for the four layer system 0/1/2/3. The Eq. (1) is modified by substituting r_{12} to r_{13} which represents the reflection coefficient of layer system 1/2/3.

$$R = \left| \frac{r_{01} + r_{13}\, exp(2ik_{z1}d)}{1 + r_{01}r_{13}\, exp(2ik_{z1}d)} \right|^2 \tag{10}$$

The reflection coefficient can be derived as follows. The three layer system 1/2/3 can be consider as depicted in figure 2. The relationship between principal axes of the film (x', y', z') and the film surface coordinates (x, y, z) may be expressed as

$$x = x' \qquad y = y'\cos\phi - z'\sin\phi \qquad z = y'\sin\phi + z'\cos\phi \tag{11}$$

The displacement vector, $\vec{D}$, in the principal axes coordinates and in the film coordinates can be represented as[4]

$$\begin{aligned} \vec{D} &= \hat{e}_x \varepsilon_{x'} E_{x'} + \hat{e}_{y'} \varepsilon_{y'} E_{y'} + \hat{e}_{z'} \varepsilon_{z'} E_{z'} \\ &= \hat{e}_x \varepsilon_x E_x + \hat{e}_y \varepsilon_y E_y + \hat{e}_z \varepsilon_z E_z \end{aligned} \tag{12}$$

The index of refraction in three principal axes n'_x, n'_y, n'_z can be used to represented the linear relationship between the displacement vector and the electric field vector

$$D_x = D_{x'}, \quad D_y = \varepsilon_0 (n_y^2 E_y + n_{yz} E_z), \quad D_z = \varepsilon_0 (n_{yz} E_y + n_z^2 E_z) \tag{13}$$

which the index of refraction in film coordinates can be represented as a tensor factors

$$n_y^2 = n_{y'}^2 \cos^2\phi + n_{z'} \sin^2\phi \tag{14}$$

$$n_z^2 = n_{y'}^2 \sin^2\phi + n_{z'}^2 \cos^2\phi \tag{15}$$

$$n_{yz} = (n_{y'}^2 - n_{z'}^2) \sin\phi \cos\phi \tag{16}$$

thus the reflection coefficient for p-polarized light is

$$r_{12} = \frac{n_{2z} n_{2y} (1 - (n_{2yz} / n_{2y} n_{2z})^2)^{1/2} (n_1^2 - (n_0 \sin\phi)^2) - n_1^2 (n_{2z}^2 - (n_0 \sin\phi)^2)}{n_{2z} n_{2y} (1 - (n_{2yz} / n_{2y} n_{2z})^2)^{1/2} (n_1^2 - (n_0 \sin\phi)^2) + n_1^2 (n_{2z}^2 - (n_0 \sin\phi)^2)} \tag{17}$$

$$r_{23} = \frac{-n_{2z}n_{2y}(1-(n_{2yz}/n_{2y}n_{2z})^2)^{1/2}(n_3^2-(n_0\,sin\phi)^2)+n_3^2(n_{2z}^2-(n_0\,sin\phi)^2)}{n_{2z}n_{2y}(1-(n_{2yz}/n_{2y}n_{2z})^2)^{1/2}(n_3^2-(n_0\,sin\phi)^2)+n_3^2(n_{2z}^2-(n_0\,sin\phi)^2)} \tag{18}$$

$$r_{13} = \frac{r_{12}+r_{23}\,exp(2ik_{z2}d)}{1+r_{12}r_{23}\,exp(2ik_{z2}d)} \tag{19}$$

If we simulate the ATR curve of the four-layer system. The silver film on the prism with index of refraction 0.05, index of extinction 3 and thickness 50nm first, the anisotropic thin SiO_2 film with index of refraction $n'_x = 1.40, n'_y = 1.40, n'_z = 1.30$ and thickness 100nm, the principal axis makes an angle ϕ with the normal of the thin film. The tilt angle ϕ varies from 20 degree to 60 degree, we can find the obvious change in ATR curve (Fig.2~Fig.6). This phenomenon confirms the sensitivity in anisotropy by using ATR device.

2. Experiments

The silver films were evaporated by an electron beam gun and deposited on the prism at an ambient temperature. Two samples were prepared under the same coating condition. One of them was used to calculate the optical constants of silver film. The other one remained on the coater to coat anisotropic thin film. The chamber was cryogenically pumped to a base pressure of 0.1 mPa before each deposition. An electron beam voltage of 7kV was then used to evaporate the silver with an electron beam current ranging from 30 to 60 mA, depending on the evaporation rate. The purity of silver for evaporation was 99.99%. A quartz thickness monitor set next to the substrate was used to measure deposition rate and controlled the thickness of silver film and SiO_2 film to be 50nm and 100nm. The SiO_2 film is evaporated on the silver film with different incident angles of vapour flux $\phi' = 50^0$.

The optical constants of silver film can be calculated by the character of the ATR curve of the sample without SiO_2 film. The ATR curve of the sample with SiO_2 film will be used to derive the optical constants n'_x, n'_y, n'_z, d_2, an the tilt angle ϕ. First we take the SiO_2 film as an isotropic film, and then calculate its optical constants $\hat{n}_2$, $\hat{d}_2$. The curve fitting method is used to get the real values. The initial values of n'_x, n'_y, n'_z are taken to be the same value $\hat{n}_2$, the initial value of thickness is taken to be $\hat{d}_2$. The initial value of ϕ can be theoretically predicted by the relation[9] $tan\phi = 0.5\,tan\phi'$. The measured curve is shown in Fig.6.

The curve fitting results show that the optical constants of silver film are n=0.041, k=3.902, d=50.203nm. The optical constants of SiO_2 film are $n'_y = 1.402$, $n'_z = 1.324$, $d_2 = 100.489nm$, the tilt angle is 34.776 deg.

The optical constant n'_x can be measured by the s-light reflection spectrum with the same ATR equipment. The value of n'_x is 1.399.

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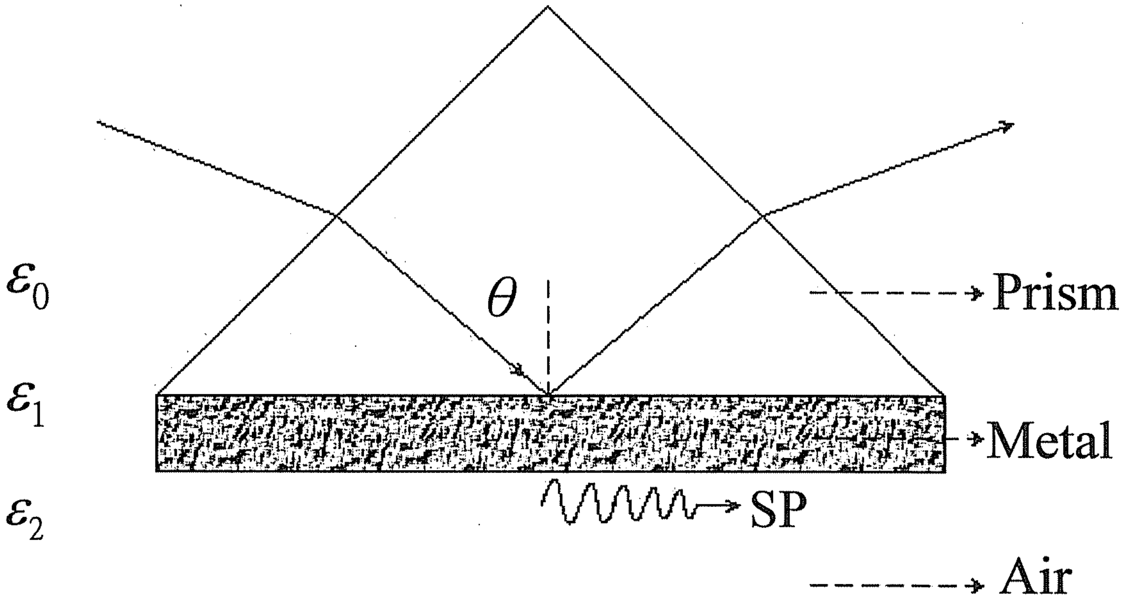

Fig. 1. Scheme of the ATR coupler in the Kretschmann configuration.

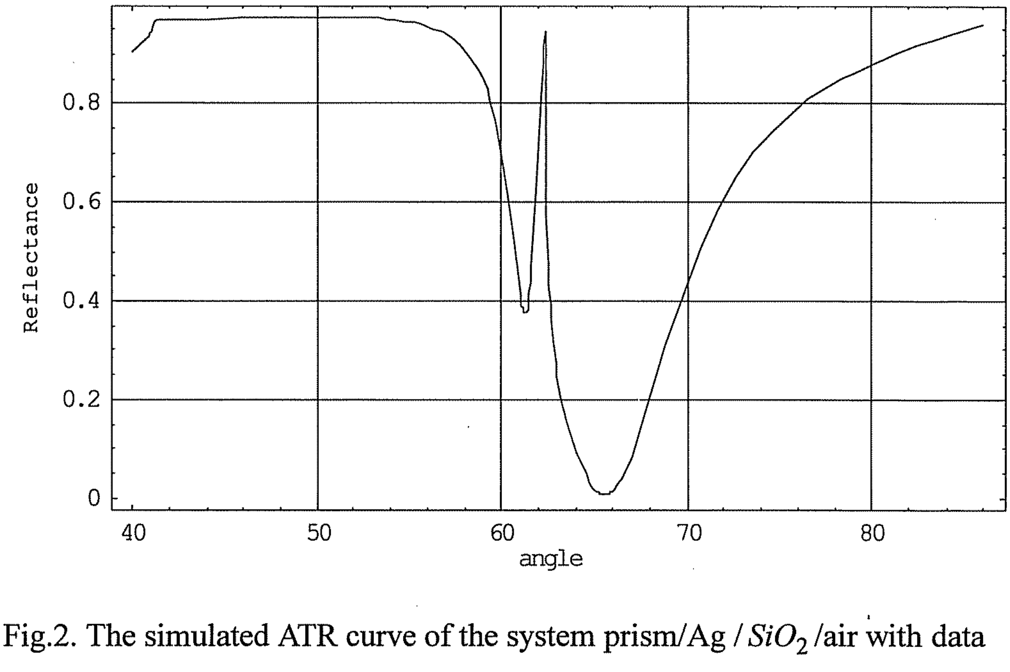

Fig.2. The simulated ATR curve of the system prism/Ag / SiO_2 /air with data

silver film : index of refraction 0.05, index of extinction 3 and thickness 50nm

SiO_2 film : index of refraction $n'_x = 1.40, n'_y = 1.40, n'_z = 1.30$ and thickness 100nm, the angle ϕ =20 deg

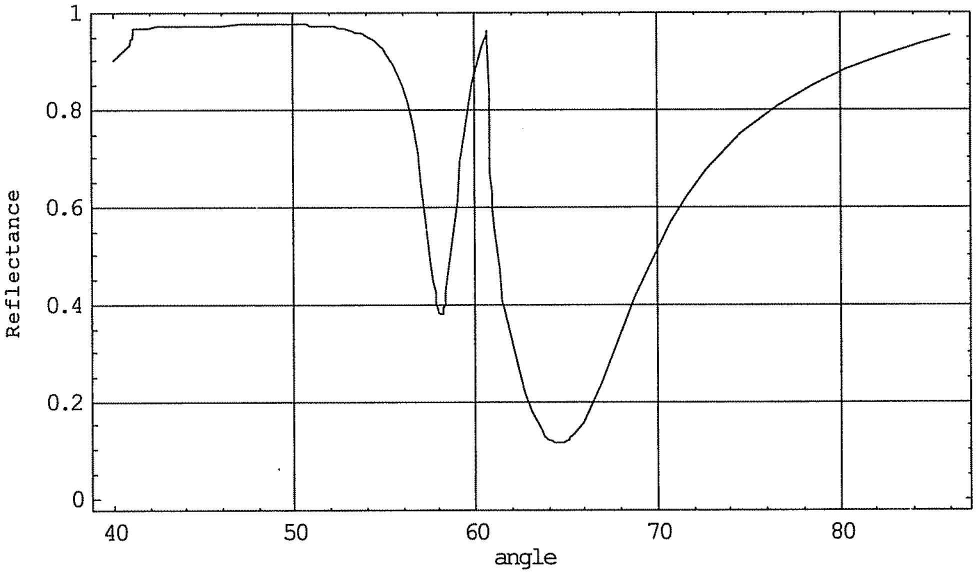

Fig.3. The simulated ATR curve of the system prism/Ag / SiO_2 /air with data

silver film : index of refraction 0.05, index of extinction 3 and thickness 50nm
SiO_2 film : index of refraction $n'_x = 1.40, n'_y = 1.40, n'_z = 1.30$
and thickness 100nm, the angle ϕ =30 deg

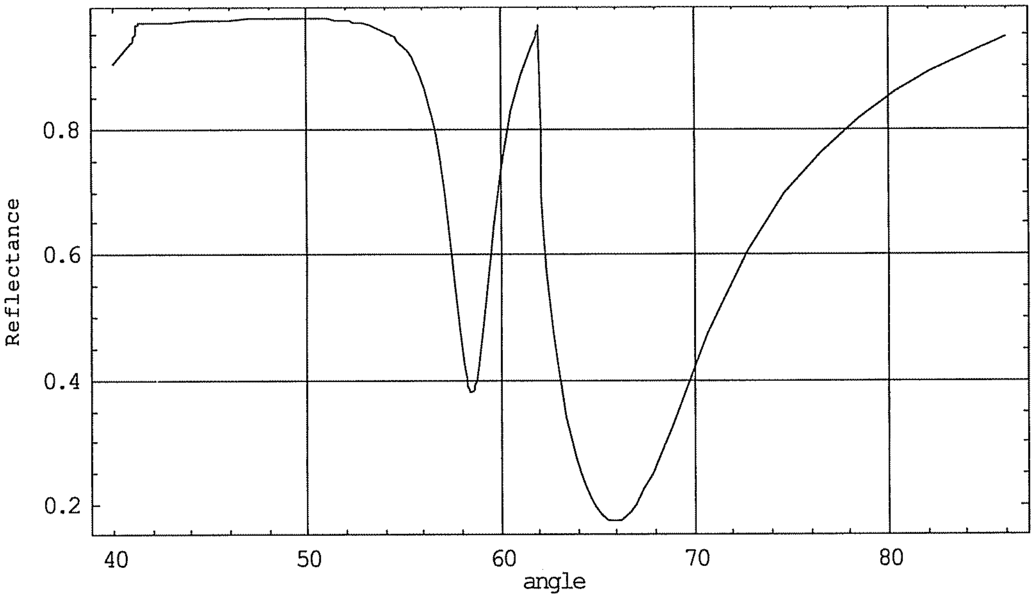

Fig.4. The simulated ATR curve of the system prism/Ag / SiO_2 /air with data

silver film : index of refraction 0.05, index of extinction 3 and thickness 50nm
SiO_2 film : index of refraction $n'_x = 1.40, n'_y = 1.40, n'_z = 1.30$ and thickness
100nm, the angle ϕ =40 deg

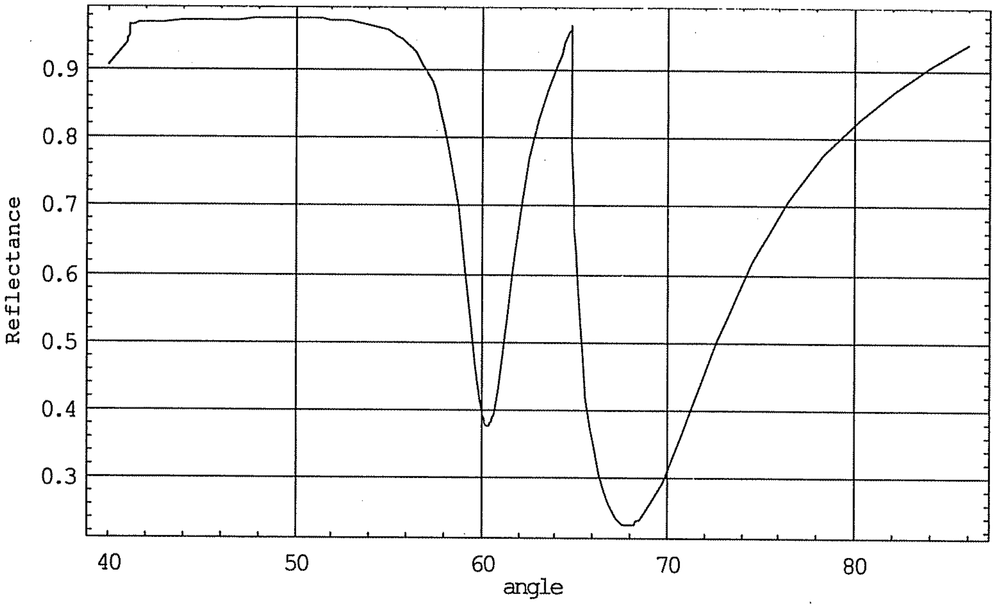

Fig.5. The simulated ATR curve of the system prism/Ag / SiO_2 /air with data silver film : index of refraction 0.05, index of extinction 3 and thickness 50nm SiO_2 film : index of refraction $n'_x = 1.40, n'_y = 1.40, n'_z = 1.30$ and thickness 100nm, the angle ϕ=40 deg

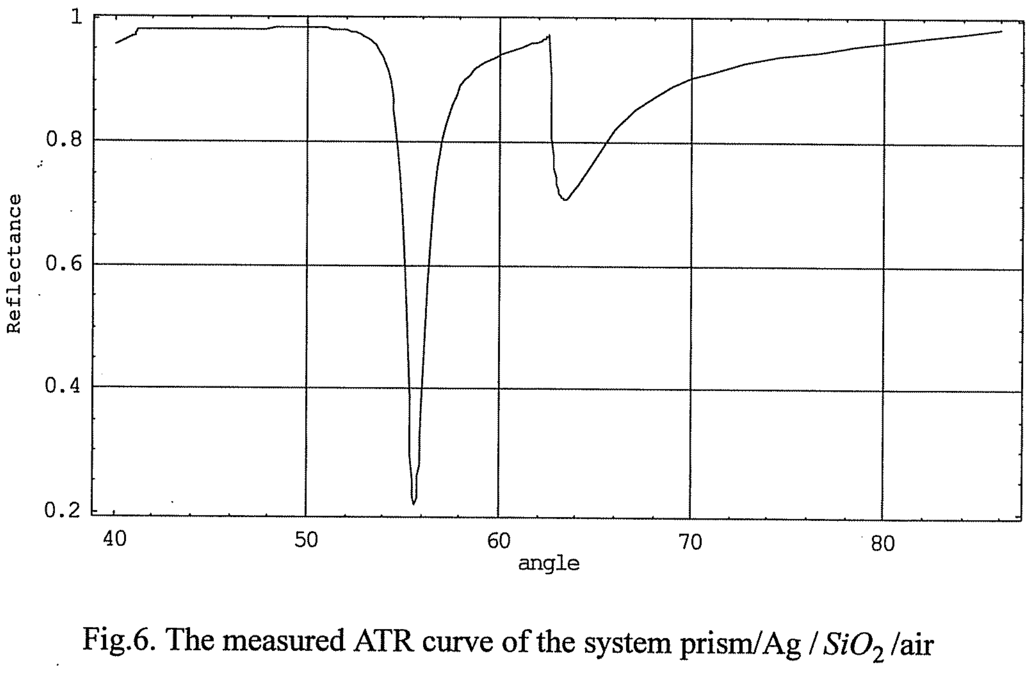

Fig.6. The measured ATR curve of the system prism/Ag / SiO_2 /air

利用全反射衰減法來決定非均向性薄膜的光學常數

任貽均*

摘　要

由於非均向性薄膜的特性顯著地影響全反射衰減法曲線的特性，包括半寬度的改變以及最小反射的角度位移，本篇論文將以全反射衰減法為工具來量測非均向性薄膜的光學特性。

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