Appl. Phys. B 77, 839–842 (2003)
DOI: 10.1007/s00340-003-1292-6

Applied Physics B
Lasers and Optics

K.H. CHEN✉
C.C. HSU
D.C. SU

A method for measuring the complex refractive index and thickness of a thin metal film

Institute of Electro-Optical Engineering, National Chiao Tung University, 1001 Ta-Hsueh Road, Hsin-Chu, Taiwan, R.O.C.

Received: 15 May 2003/Revised version 23 June 2003

ABSTRACT A circularly polarized heterodyne light beam is incident on a thin metal film, causing successive reflections and refractions to occur at the two sides of the thin film. The phase difference between *p*- and *s*-polarizations of the multiple-beam interference signal can be measured accurately with an analyzer and heterodyne interferometry. The phase difference depends on the azimuth angle of the analyzer, the complex refractive index and the thickness of the thin metal film. The measured values of the phase differences under three different azimuth angles of the analyzer can be substituted into the special equations derived from Fresnel's equations and multiple-beam interference. Hence, the complex refractive index and the thickness of the thin metal film can be estimated by using a personal computer with a numerical analysis technique. Because of its common-path optical configuration and its heterodyne interferometric phase measurement, this method has many merits, such as high stability against surrounding vibrations, high resolution and easy operation.

PACS 78.66.Bz; 78.20.Ci; 07.60.Cy

1 Introduction

The determination of the complex refractive index and the thickness of a thin metal film is very important for microelectronics, optoelectronics and optics. Several methods [1–5] have been proposed to measure either of them. Other methods [6–9] have been reported to be able to measure both of them simultaneously. However, most of these measurement processes are related to light intensity variations; hence, the surrounding light and scattered light easily influence the measurement accuracy and resolution. One of the previous methods [5] measures the phase variation instead of the intensity variation; it has better measurement resolution. Nevertheless, it can only estimate the complex refractive index with measured data for different incident angles. Hence, the adjustment of the optical set-up becomes tedious. Furthermore, it cannot measure the thickness.

In this paper, an alternative method is presented for measuring both the complex refractive index and the thickness with only one optical configuration. A circularly polarized heterodyne light beam is incident on a thin metal film, and successive reflections and refractions occur at the two sides of the thin film. Multiply reflected beams interfere with one another, and an interference signal can be obtained. The phase difference between *p*- and *s*-polarizations of the interference signal can be measured accurately with an analyzer and heterodyne interferometry. Its value depends on the azimuth angle of the analyzer, the complex refractive index and the thickness of the thin metal film. By substituting the measured values of the phase differences at three different azimuth angles of the analyzer into the special equations derived from Fresnel's equations and multiple-beam interference, the complex refractive index and the thickness of the thin metal film can be estimated by using a personal computer with a numerical analysis technique. Because of its common-path optical configuration and its heterodyne interferometric phase measurements at three different azimuth angles of the analyzer, this method has many merits, such as high stability against surrounding vibrations, high resolution and easy operation. In addition, five samples were tested to show the validity of this method.

2 Principles

The schematic diagram of this method is shown in Fig. 1. For convenience, the $+z$ axis is chosen to be along the direction of propagation and the x axis is along the horizontal direction. A circularly polarized heterodyne light source with an angular frequency difference ω between left- and right-circular polarizations was used. If the amplitude of the light coming from this light source is unity, then its Jones vector can be written as [10]:

$$E_i = \begin{pmatrix} \cos\left(\frac{\omega t}{2}\right) \\ -\sin\left(\frac{\omega t}{2}\right) \end{pmatrix} = \frac{1}{2}\begin{pmatrix} 1 \\ i \end{pmatrix} \exp\left(i\frac{\omega t}{2}\right) + \frac{1}{2}\begin{pmatrix} 1 \\ -i \end{pmatrix} \exp\left(-i\frac{\omega t}{2}\right) . \quad (1)$$

It is incident at θ_0 onto the sample S, which consists of a thin metal film deposited on a substrate. Multiple reflections and refractions occur at both sides of the thin metal film, as shown in Fig. 2. These multiply reflected light beams (E_{1r}, E_{2r}, E_{3r}, ...) pass through a small aperture AP and an

---

✉ Fax: +886-3/571-6631, E-mail: t7503@faculty.nctu.edu.tw

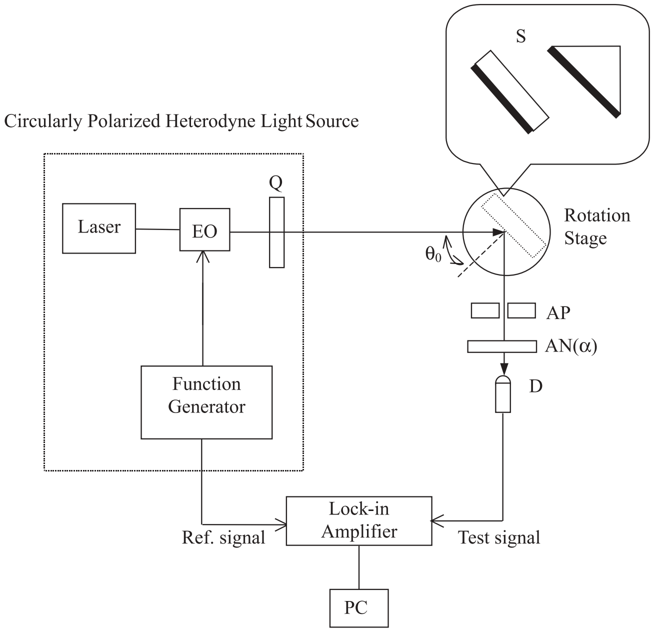

FIGURE 1 Schematic diagram of measurement of the phase difference using multiple-beam reflection in the thin metal film. EO, electro-optic modulator; Q, quarter-wave plate; S, sample; AP, aperture; AN, analyzer; D, photodetector

analyzer AN, and then finally enter a photodetector D. The aperture is used to block the reflected light from the back surface of the substrate. Based on Fresnel's equations and multiple-beam interference, the reflection coefficients of the p- and s-polarizations can be expressed as [11]:

$$r_p = \frac{r_{01,p} + r_{12,p} \mathrm{e}^{i2\beta}}{1 + r_{01,p} r_{12,p} \mathrm{e}^{i2\beta}} , \tag{2a}$$

and

$$r_s = \frac{r_{01,s} + r_{12,s} \mathrm{e}^{i2\beta}}{1 + r_{01,s} r_{12,s} \mathrm{e}^{i2\beta}} ; \tag{2b}$$

where r_{ij} is the amplitude reflection coefficient at the interface as the beam from medium i is incident on medium j. Subscripts i and j are any of 0 (air), 1 (thin metal film) and 2 (substrate), whose refractive indices are n_0, $n_1 = n + ik$, and n_2, respectively. They can be expressed as [11]:

$$r_{01,p} = \frac{n_0 \cos\theta_1 - n_1 \cos\theta_0}{n_0 \cos\theta_1 + n_0 \cos\theta_0}, \quad r_{12,p} = \frac{n_1 \cos\theta_2 - n_2 \cos\theta_1}{n_1 \cos\theta_2 + n_2 \cos\theta_1} , \tag{3a}$$

and

$$r_{01,s} = \frac{n_0 \cos\theta_1 - n_1 \cos\theta_1}{n_0 \cos\theta_0 + n_1 \cos\theta_1}, \quad r_{12,s} = \frac{n_1 \cos\theta_1 - n_2 \cos\theta_2}{n_1 \cos\theta_1 + n_2 \cos\theta_2} , \tag{3b}$$

respectively. In addition,

$$\beta = \frac{2\pi d \sqrt{n_1^2 - n_0^2 \sin^2\theta_0}}{\lambda} , \tag{4}$$

where λ is the wavelength of the light beam, θ_1 and θ_2 are the corresponding refractive angles of air to thin metal film and thin metal film to substrate, respectively, and d is the thickness of the thin metal film, as indicated in Fig. 2.

If the transmission axis of AN is located at α with respect to the x-axis, then the Jones vector of the light arriving at D is:

$$\begin{aligned} E_t &= \begin{pmatrix} \cos^2\alpha & \sin\alpha\cos\alpha \\ \sin\alpha\cos\alpha & \sin^2\alpha \end{pmatrix} \begin{pmatrix} r_p & 0 \\ 0 & r_s \end{pmatrix} \begin{pmatrix} \cos\frac{\omega t}{2} \\ -\sin\frac{\omega t}{2} \end{pmatrix} \\ &= \begin{pmatrix} r_p \cos^2\alpha \cos\frac{\omega t}{2} - r_s \sin\alpha\cos\alpha \sin\frac{\omega t}{2} \\ r_p \sin\alpha\cos\alpha\cos\frac{\omega t}{2} - r_s \sin^2\alpha \sin\frac{\omega t}{2} \end{pmatrix} . \end{aligned} \tag{5}$$

Hence, the intensity measured by D can be written as:

$$I_t = |E_t|^2 = I_0 [1 + \gamma \cos(\omega t + \phi)] , \tag{6}$$

where I_t is the test signal, I_0 and γ are the bias intensity and the visibility of the signal, and ϕ is the phase difference between the p and s polarizations of the beam reflected from the sample. They can be expressed as:

$$I_0 = \frac{1}{2} \left(|r_p|^2 \cos^2\alpha + |r_s|^2 \sin^2\alpha \right) , \tag{7a}$$

$$\gamma = \frac{\sqrt{A^2 + B^2}}{I_0} , \tag{7b}$$

$$A = \frac{1}{2} \left(|r_p|^2 \cos^2\alpha - |r_s|^2 \sin^2\alpha \right) , \tag{7c}$$

$$B = \frac{1}{2} \left(r_p r_s^* + r_s r_p^* \right) \sin\alpha\cos\alpha \tag{7d}$$

and

$$\phi = \tan^{-1}\left(\frac{B}{A}\right) = \tan^{-1}\left[\frac{\left(r_p r_s^* + r_s r_p^* \right) \sin\alpha\cos\alpha}{|r_p|^2 \cos^2\alpha - |r_s|^2 \sin^2\alpha} \right] , \tag{7e}$$

where r_p^* and r_s^* are the conjugates of r_p and r_s, respectively.

In contrast, the modulated electronic signal of the heterodyne light source is filtered and becomes the reference signal. It has the form:

$$I_r = \frac{1}{2} [1 + \cos(\omega t)] . \tag{8}$$

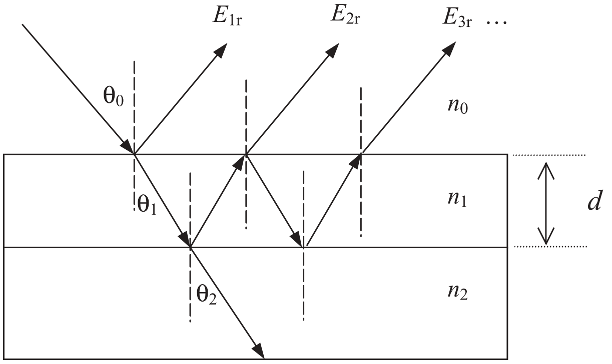

FIGURE 2 Multiple-beam reflection in the thin metal film

Both of these two sinusoidal signals $I_t$and I_r are sent to a lock-in amplifier and ϕ can be measured accurately.

From (2)–(4) and (7), it is obvious that the phase difference ϕ is a function of n, k, d and α. Here, the phase difference ϕ can be experimentally measured for each given α. To evaluate the values of n, k and d, we require three phase differences ϕ_1, ϕ_2 and ϕ_3 that correspond to the three azimuth angles α_1, α_2 and α_3. Hence a set of simultaneous equations are obtained:

$$\phi_1 = f(n, k, d, \alpha_1)\,, \tag{9a}$$

$$\phi_2 = f(n, k, d, \alpha_2) \tag{9b}$$

and

$$\phi_3 = f(n, k, d, \alpha_3)\,. \tag{9c}$$

If these simultaneous equations are solved, the complex refractive index n, k and the thickness d of the thin metal film can be estimated, simultaneously.

3 Experiments and results

To show the feasibility of this method, we measured the complex refractive indices of the thin gold films of five samples, three on BK7 glass substrates (samples 1–3) and the others on the hypotenuse of BK7 prisms (samples 4 and 5). The latter two samples were the surface plasmon resonance probes of the Kretschmann configuration [12, 13]. A circularly polarized heterodyne light source with a 1-kHz difference frequency between the left- and right-circular polarizations was used. It consisted of a He–Ne laser at 632.8 nm, a quarter-wave plate Q and an electro-optic modulator EO (model 4001, New Focus, Inc.) driven by a function generator FG, as shown in Fig. 1. We used a high-precision rotation stage (model PS-θ-90, Japan Chuo Precision Industrial Company Ltd.) with an angular resolution of 0.005° to mount and rotate the test sample, and a lock-in amplifier (model SR-850, Stanford Research Systems) with an angular resolution of 0.001° to measure the phase difference. For convenience, we measured the phase differences under the experimental conditions $\theta_0 = 50°, \alpha_1 = 50°, \alpha_2 = 55°$ and $\alpha_3 = 60°$. The measurement results and estimated results are summarized in Tables 1 and 2. The reference data measured by a commercial ellipsometer (model eta, Steag Inc.) are added in Table 2 for comparison. It is clear that they show good correspondence.

Sample No.	ϕ_1	ϕ_2	ϕ_3
1	75.357°	68.897°	61.947°
2	78.951°	72.621°	65.777°
3	82.406°	76.287°	69.644°
4	82.966°	76.879°	70.274°
5	83.774°	77.746°	71.202°

TABLE 1 Measurement results at $\alpha = 50°$, 55° and 60°

Sample No.	Estimated results			Reference data		
	n	k	d (nm)	n	k	d (nm)
1	0.288	3.536	15.83	0.288	3.543	15.89
2	0.294	3.574	20.99	0.295	3.573	21.11
3	0.201	3.562	31.99	0.199	3.560	32.64
4	0.197	3.563	35.17	0.201	3.561	35.28
5	0.211	3.596	40.24	0.213	3.599	40.12

TABLE 2 Estimated results and the reference data

4 Discussion

Because there are three unknowns (n, k and d), we need at least three equations to solve them. These can be obtained by measuring ϕ at three different azimuth angles of the analyzer, as described in our method. In contrast to other methods [4–6], which operated at three different incident angles, this method can be operated more easily.

The thin gold film of each sample was deposited by a sputtering processes. Owing to the sputtering conditions, each sample may have its corresponding constraints [14]. Thus, the measurement data for n and k of each sample are slightly different. However, they are almost equal to the values obtained with an ellipsometer for the same sample. Therefore, it can be concluded that this method is valid.

From (2)–(4) and (7), we can obtain:

$$|\Delta n| = -\frac{\begin{gathered} B_3C_2\,|\Delta\phi_1| - B_2C_3\,|\Delta\phi_1| - B_3C_1\,|\Delta\phi_2| \\ +B_1C_3\,|\Delta\phi_2| + B_2C_1\,|\Delta\phi_3| - B_1C_2\,|\Delta\phi_3| \end{gathered}}{\begin{gathered} -A_3B_2C_1 + A_2B_3C_1 + A_3B_1C_2 \\ -A_1B_3C_2 - A_2B_1C_3 + A_1B_2C_3 \end{gathered}}\,, \tag{10}$$

$$|\Delta k| = -\frac{\begin{gathered} A_3C_2\,|\Delta\phi_1| - A_2C_3\,|\Delta\phi_1| - A_3C_1\,|\Delta\phi_2| \\ +A_1C_3\,|\Delta\phi_2| + A_2C_1\,|\Delta\phi_3| - A_1C_2\,|\Delta\phi_3| \end{gathered}}{\begin{gathered} A_3B_2C_1 - A_2B_3C_1 - A_3B_1C_2 \\ +A_1B_3C_2 + A_2B_1C_3 - A_1B_2C_3 \end{gathered}}\,, \tag{11}$$

$$|\Delta d| = -\frac{\begin{gathered} A_3B_2\,|\Delta\phi_1| - A_2B_3\,|\Delta\phi_1| - A_3B_1\,|\Delta\phi_2| \\ +A_1B_3\,|\Delta\phi_2| + A_2B_1\,|\Delta\phi_3| - A_1B_2\,|\Delta\phi_3| \end{gathered}}{\begin{gathered} -A_3B_2C_1 + A_2B_3C_1 + A_3B_1C_2 \\ -A_1B_3C_2 - A_2B_1C_3 + A_1B_2C_3 \end{gathered}}\,, \tag{12}$$

where

$$A_1 = \left|\frac{\partial\phi_1}{\partial n}\right|\,,\quad A_2 = \left|\frac{\partial\phi_2}{\partial n}\right|\,,\quad A_3 = \left|\frac{\partial\phi_3}{\partial n}\right|\,; \tag{13a}$$

$$B_1 = \left|\frac{\partial\phi_1}{\partial k}\right|\,,\quad B_2 = \left|\frac{\partial\phi_2}{\partial k}\right|\,,\quad B_3 = \left|\frac{\partial\phi_3}{\partial k}\right|\,; \tag{13b}$$

and

$$C_1 = \left|\frac{\partial\phi_1}{\partial d}\right|\,,\quad C_2 = \left|\frac{\partial\phi_2}{\partial d}\right|\,,\quad C_3 = \left|\frac{\partial\phi_3}{\partial d}\right|\,; \tag{13c}$$

Δn, Δk and Δd are the errors in n, k and d, and $\Delta\phi_1$, $\Delta\phi_2$, and $\Delta\phi_3$ are the errors in the phase differences ϕ_1, ϕ_2 and ϕ_3, respectively. By taking into account the angular resolution of the lock-in amplifier, the second-harmonic error and the polarization-mixing errors, $|\Delta\phi_1| = |\Delta\phi_2| = |\Delta\phi_3| \cong 0.03°$ can be estimated in our experiments [5]. Substituting these data and the experimental conditions into (10)–(13), the corresponding data for $|\Delta n|$, $|\Delta k|$ and $|\Delta d|$ of each sample were calculated with the software "Mathematica", and the results are summarized in Table 3. In addition, the errors in θ_0 and α were 0.005° and 0.08° in our experiments. Their induced errors in ϕ can be calculated similarly: they are 0.006° and 0.005°, respectively. Since these are much smaller than 0.03°, they can be neglected.

Since the thin metal film is absorbing, the amplitude–reflection coefficients at the interfaces are not so high. Consequently, only the first two reflected beams E_{1r} and E_{2r} need to be considered. For better contrast of the interference signal, it is necessary to satisfy the condition:

$$\frac{|E_{2r}|^2}{|E_{1r}|^2} > 0.1 \,. \tag{14}$$

Therefore, we have

$$kd < \frac{\lambda}{8\pi\cos\theta_1} \ln \left| \frac{6.25\times10^{-3}\left(n_1^2\cos^2\theta_0-\cos^2\theta_1\right)^2 \times\left(n_2\cos\theta_1+n_1\cos\theta_2\right)^2}{n_1^2\cos^2\theta_0\cos^2\theta_1(n_2\cos\theta_1-n_1\cos\theta_2)^2} \right| . \tag{15}$$

From (15), it can be seen that the measurable range of thickness depends on the incident angle and the refractive indices of the thin metal film and the substrate. By substituting the experimental conditions and the measured results into (15), we can conclude that this method is suitable for thin gold films of thickness smaller than 55 nm under our experimental conditions.

Sample	$\lvert\Delta n\rvert$	$\lvert\Delta k\rvert$	$\lvert\Delta d\rvert$(nm)
1	1.01×10^{-3}	1.45×10^{-3}	0.05
2	1.81×10^{-3}	2.53×10^{-3}	0.08
3	2.3×10^{-3}	3.09×10^{-3}	0.18
4	1.35×10^{-3}	1.86×10^{-3}	0.23
5	1.43×10^{-3}	3.14×10^{-3}	0.33

TABLE 3 Estimated errors in Δn, Δk and Δd

For practical uses, a broadband light source and interference filters can be used to replace the laser light source. The half-wave voltage of the modulated electronic signal applied to the electro-optic modulator EO should be changed accordingly as the wavelength varies. For convenience, it is better to use a zero-order quarter-wave plate because of its broadband stability.

5 Conclusions

In this paper, we have presented an alternative method for measuring both the complex refractive index and the thickness of a thin metal film by using heterodyne interferometry, Fresnel's equations and multiple-beam interference. These measurements can be obtained with just one optical setup at three different azimuth angles of the analyzer. During the measurement procedure, the incident angle need not be varied as it is in other methods. Hence, measurements are made more easily. In addition, the method has the advantages of both a common-path interferometer and a heterodyne interferometer.

ACKNOWLEDGEMENTS This study was supported in part by the National Science Council, Taiwan, under Contract No. NSC 91-2215-E-009-020.

REFERENCES

1 M. Cathelinaud, F. Lemarquis, C. Amra: Appl. Opt. **41**, 2546 (2002)
2 E. Elizalde, J.M. Frigerio, J. Rivory: Appl. Opt. **25**, 4557 (1986)
3 H.G. Tompkins, T. Zhu, E. Chen: J. Vac. Sci. Technol. A **16**, 1297 (1998)
4 V.A. Lodi, S. Donati, S. Merlo, D. Beltrami: Appl. Opt. **37**, 5298 (1998)
5 M.H. Chiu, J.Y. Lee, D.C. Su: Appl. Opt. **38**, 4047 (1999)
6 H. Kitajima, K. Hiea, Y. Suematsu: J. Opt. Soc. Am. **70**, 1507 (1980)
7 J. Lekner: J. Opt. Soc. Am. A **11**, 2156 (1994)
8 E.W. Rogala, H.H. Barrett: J. Opt. Am. A **15**, 538 (1998)
9 A.B. Djurisic, T. Fritz, K. Leo: Opt. Commun. **166**, 35 (1999)
10 J.Y. Lee, D.C. Su: J. Opt. **29**, 349 (1998)
11 M. Born, E. Wolf: *Principles of Optics*, 7th edn. (Cambridge University Press, Cambridge 1999), Chap. 1
12 K.H. Chen, C.C. Hsu, D.C. Su: Opt. Commun. **209**, 167 (2002)
13 Y.C. Cheng, W.K. Su, J.H. Liou: Opt. Eng. **39**, 311 (2000)
14 S.A. Campbell: *The Science and Engineering of Microelectronic Fabrication*, 1st edn. (Oxford University Press, Oxford 1990) p. 292