Complex refractive-index measurement based on Fresnel's equations and the uses of heterodyne interferometry

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The phase difference between *s* and *p* polarization of the light reflected from a material is used for measuring the material's complex refractive index. First, two phase differences that correspond to two different incidence angles are measured by heterodyne interferometry. Then these two phase differences are substituted into Fresnel's equations, and a set of simultaneous equations is obtained. Finally, the equations are solved by use of a personal computer by a numerical analysis technique, and the complex refractive index of the material can be estimated. © 1999 Optical Society of America

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1. Introduction

A complex refractive index $N(n, k)$ is an important characteristic constant of thin-film materials, where n is the refractive index and k is the extinction coefficient. There are several methods for measuring the complex refractive index of a material, e.g., R-versus-θ (reflectance versus incidence angle) methods[1–6] and ellipsometry.[7,8] In those methods, the reflectances of s and p polarization at several different incidence angles or polarization conditions must be measured. Consequently, most of them are related to the measurement of light-intensity variations. However, the stability of the light source, the scattering light, the internal reflection, and other factors influence the accuracy of the measurement results. Recently, Feke *et al.*[9] proposed a novel method to measure the complex refractive index. The phases that correspond to two orthogonal polarizations at different incidence angles are measured with a phase-shifting interferometric technique. These data are substituted into the special equations derived from Fresnel's equations, and the complex refractive index is obtained.

In this paper a simple method for measuring a complex refractive index is proposed. A light beam of either s or p polarization reflected from a material with a complex refractive index has a phase shift. The phase difference between these two kinds of polarization is a function of n, k, and incidence angle θ. The phase differences that correspond to two different incidence angles are measured with the heterodyne interferometry proposed by Chiu *et al.*[10] These data are substituted into Fresnel's equations,[11] and a set of simultaneous equations is obtained. Then these equations are solved by use of a personal computer by a numerical analysis technique, and the complex refractive index of the material can be estimated. The technique has several merits, including a simple optical setup, easy operation, high stability, high measurement accuracy, and rapid measurement. We demonstrate its feasibility.

2. Principles

A. Phase Difference Resulting from Reflection from an Absorbing Material

A ray of light in air is incident at θ onto an absorbing material with a complex refractive index $N(n, k)$, as shown in Fig. 1. According to Fresnel's equa-

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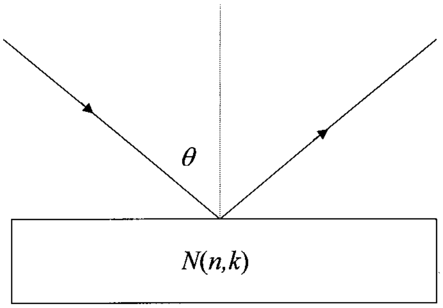


Fig. 1. Reflection at the surface of an absorbing material.

tions,[11] the amplitude reflection coefficients of s and p polarization can be expressed as

$$r_s = \frac{\cos\theta - (u + iv)}{\cos\theta + (u + iv)} = |r_s|\exp(i\delta_s), \tag{1}$$

$$r_p = \frac{N^2\cos\theta - (u + iv)}{N^2\cos\theta + (u + iv)} = |r_p|\exp(i\delta_p), \tag{2}$$

respectively, where

$$u^2 = \tfrac{1}{2}\{(n^2 - k^2 - \sin^2\theta) + [(n^2 - k^2 - \sin^2\theta)^2 + 4n^2k^2]^{1/2}\}, \tag{3}$$

$$v^2 = \tfrac{1}{2}\{-(n^2 - k^2 - \sin^2\theta) + [(n^2 - k^2 - \sin^2\theta)^2 + 4n^2k^2]^{1/2}\}, \tag{4}$$

and δ_s and δ_p are the phase shifts of s and p polarizations and can be expressed as

$$\delta_s = \tan^{-1}\left(\frac{2v\cos\theta}{u^2 + v^2 - \cos^2\theta}\right), \tag{5}$$

$$\delta_p = \tan^{-1}\left[\frac{2v\cos\theta(n^2 - k^2 - 2u^2)}{u^2 + v^2 - (n^2 + k^2)^2\cos^2\theta}\right], \tag{6}$$

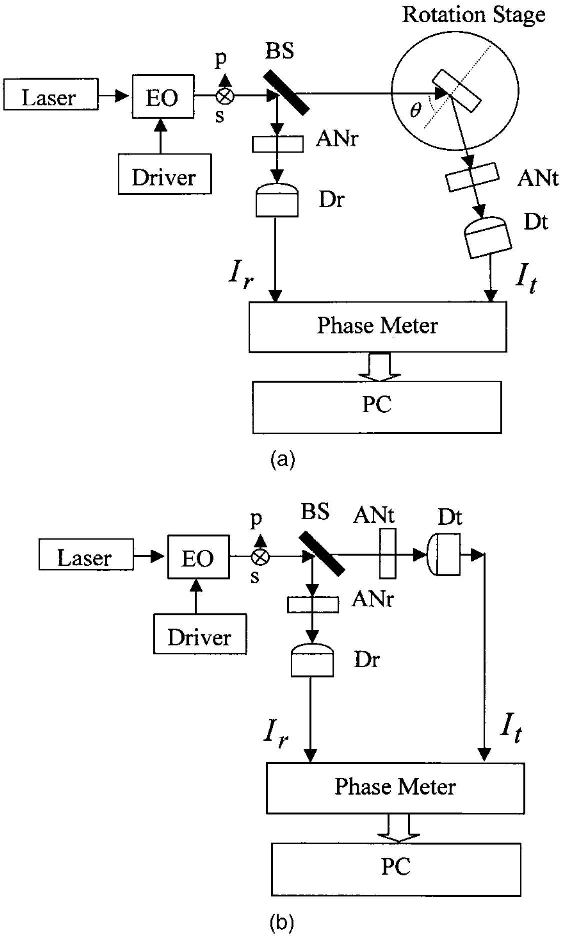
respectively. Hence the phase difference of s polarization relative to p polarization is

$$\phi = \delta_s - \delta_p = \tan^{-1}\left(\frac{ad - bc}{ac + bd}\right), \tag{7}$$

where

$$\begin{aligned} a &= 2v\cos\theta, \\ b &= u^2 + v^2 - \cos^2\theta, \\ c &= 2v\cos\theta(n^2 - k^2 - 2u^2), \\ d &= u^2 + v^2 - (n^2 + k^2)^2\cos^2\theta. \end{aligned} \tag{8}$$

From Eqs. (7) and (8) it is obvious that phase difference ϕ is a function of n, k, and θ, and ϕ can be experimentally measured for each given θ. To evaluate the values of n and k we require two phase differences ϕ_1 and ϕ_2 that correspond to two incidence angles, θ_1 and θ_2. Hence a set of simultaneous equations



(a)

(b)

Fig. 2. Schematic diagram of measurement of the phase differences owing to reflection at (a) an absorbing material and (b) a beam splitter. Other abbreviations are defined in text.

$$\phi_1 = \phi_1(n, k, \theta_1), \tag{9}$$

$$\phi_2 = \phi_2(n, k, \theta_2) \tag{10}$$

is obtained. If these simultaneous equations are solved, the complex refractive index of the material can be estimated.

B. Phase-Difference Measurements with Heterodyne Interferometry

Chiu *et al.*[10] proposed a method for measuring the refractive index of a transparent material by using total-internal-reflection heterodyne interferometry. A schematic diagram of the optical arrangement of our method, which is based on similar considerations, was designed and is shown in Fig. 2(a). Linearly polarized light passing through an electro-optic modulator (EO) is incident upon a beam splitter (BS) and divided into two parts, a reference beam and a test beam. The reference beam passes through analyzer AN*r*, and enters photodetector D*r*. If the amplitude of light detected by D*r* is E_r, then the intensity mea-

Table 1. Experimental Conditions and Measurement Results[a]

Material	θ_1 (°)	θ_2 (°)	ϕ_1 (°)	ϕ_2 (°)	n	k	Δn	Δk
Ni	60	80	144.96	74.15	2.007	3.781	0.01	0.006
	70	80	121.32	74.15	2.006	3.782	0.01	0.006
Cu	60	80	134.22	65.22	0.27	3.430	0.05	0.004
	70	80	108.25	65.22	0.31	3.428	0.07	0.005

[a] Reference values from Ref. 14: $N(\text{Ni}) = 1.98 + i3.74$ and $N(\text{Cu}) = 0.272 + i3.42$.

sured by Dr is the $I_r = |E_r|^2$. Here I_r is the reference signal. The test beam is incident at θ upon a test material. Finally, the reflected light passes through analyzer ANt and is detected by another photodetector, Dt. If the amplitude of the test beam detected by Dt is E_t, then Dt measures the intensity $I_t = |E_t|^2$, and I_t is the test signal.

For convenience the $+z$ axis is in the propagation direction and the y axis is in the vertical direction. Let the incident light be linearly polarized at 45° with respect to the x axis, the fast axis of EO under an applied electric field be in the horizontal direction, and the transmission axes of both ANr and ANt be at 45° with respect to the x axis. If a sawtoothed signal of frequency f and amplitude $V_{\lambda/2}$, the half-wave voltage of the EO, is applied to the EO, then by using Jones calculus we get[12,13]

$$I_r = \tfrac{1}{2}[1 + \cos(2\pi ft - \phi_{\text{BS}})], \tag{11}$$

$$I_t = \tfrac{1}{2}\left[\frac{|r_s|^2}{2} + \frac{|r_p|^2}{2} + \left|r_s\right|\left|r_p\right|\cos(2\pi f - \phi)\right], \tag{12}$$

where ϕ_{BS} is the phase difference between s and p polarization that is due to the reflection at the beam splitter. These two sinusoidal signals are sent to the phasemeter, and their phase difference

$$\phi' = \phi - \phi_{\text{BS}} \tag{13}$$

can be obtained. Next, let the test beam enter photodetector D_t directly without the reflection from the test material, as shown in Fig. 2(b). The test signal still has the form of Eq. (12) but this time with $\phi = 0$. Therefore the phasemeter in Fig. 2(b) represents $-\phi_{\text{BS}}$. Substituting $-\phi_{\text{BS}}$ into Eq. (13), we obtain the phase difference ϕ.

3. Experiments and Results

To show the feasibility of this technique, we measured the complex refractive indices of nickel (Ni) and cupper (Cu). A He–Ne laser with 632.8-nm wavelength and an EO (Model PC200/2, England Electro-Optics Developments, Ltd.) with a half-wave voltage of 170 V were used in this test. The frequency of the sawtoothed signal that was applied to the EO was 800 Hz. We used a high-precision rotation stage (PS–θ–90) with an angular resolution of 0.005° (Japan Chuo Precision Industrial Company, Ltd.) to mount and rotate the test material and a high-resolution phasemeter with an angular resolution of 0.01° to measure the phase difference. In addition, we used a personal computer to record and analyze the data. The experimental conditions and results are summarized in Table 1. The average experimental values of the complex refractive indices of nickel and copper are $N(\text{Ni}) = 2.01 + i3.78$ and $N(\text{Cu}) = 0.29 + i3.43$, respectively.

4. Discussion

From Eqs. (9) and (10) we get

$$\Delta n \cong \frac{\left|\frac{\partial\phi_2}{\partial k}\right||\Delta\phi_1| + \left|\frac{\partial\phi_1}{\partial k}\right||\Delta\phi_2|}{\left|\frac{\partial\phi_1}{\partial n}\frac{\partial\phi_2}{\partial k} - \frac{\partial\phi_2}{\partial n}\frac{\partial\phi_1}{\partial k}\right|}, \tag{14}$$

$$\Delta k \cong \frac{\left|\frac{\partial\phi_1}{\partial n}\right||\Delta\phi_2| + \left|\frac{\partial\phi_2}{\partial n}\right||\Delta\phi_1|}{\left|\frac{\partial\phi_1}{\partial n}\frac{\partial\phi_2}{\partial k} - \frac{\partial\phi_2}{\partial n}\frac{\partial\phi_1}{\partial k}\right|}, \tag{15}$$

where Δn and Δk are the errors in n and k and $\Delta\phi_1$ and $\Delta\phi_2$ are the errors in the phase differences at two different incidence angles θ_1 and θ_2, respectively. The errors in the phase differences in this technique may be influenced by the following factors:

(*1*) *Angular resolution of the phasemeter*

The angular resolution $\Delta\phi_p$ of the phasemeter can be expressed as[15]

$$\Delta\phi_p = \frac{f}{f_c} \times 360°, \tag{16}$$

where f and f_c are the frequencies of the input waves and the reference clock in the phasemeter, respectively. In our experiments we used $f = 800$ Hz and $f_c = 32$ MHz, so the angular resolution of the phase meter is better than 0.01°.

(*2*) *Second-harmonic error*

The second-harmonic error comes from the deviation angle θ_r between the polarization directions of p polarization of the incident beam and the incidence plane. It introduces an error in the phase difference[16]:

$$\Delta\phi_r = \frac{\tan\phi(\sec 2\theta_r - 1)}{1 + \sec 2\theta_r \tan^2\phi} \tag{17}$$

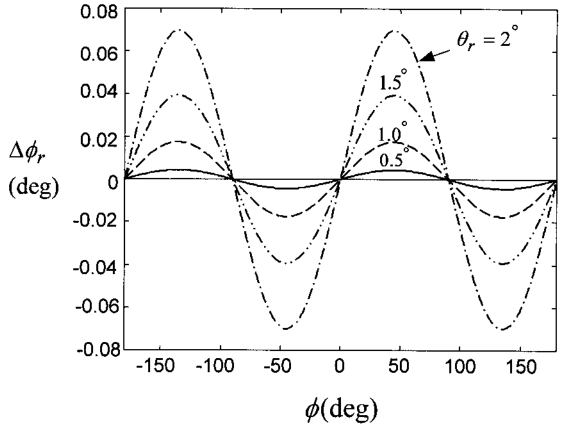


Fig. 3. $\Delta\phi_r$ versus ϕ for several values of θ_r.

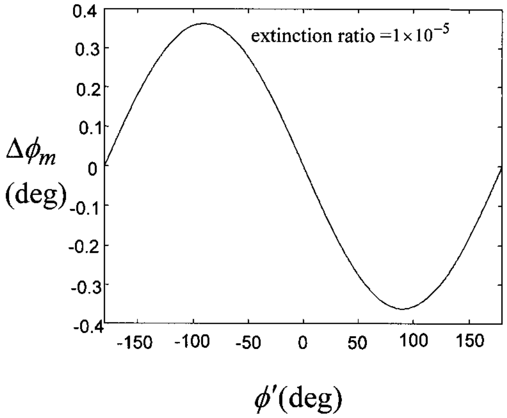


Fig. 4. $\Delta\phi_m$ versus ϕ' for a polarizer with the extinction ratio shown.

into ϕ. The relation curves of $\Delta\phi_r$ versus ϕ for $\theta_r = 0.5°, 1.0°, 1.5°, 2.0°$ are depicted in Fig. 3, and their maximum errors are 0.004°, 0.02°, 0.04°, and 0.07°, respectively. It is obvious that $\Delta\phi_r$ equals zero as $\phi = -180°, -90°, 90°, 180°$. This error can be made nearly zero by accurate modification of the azimuthal angle of every polarization component, as was done by Chiu *et al.*[13]

(*3*) *Polarization-mixing error*

Owing to the extinction ratio effect of a polarizer, mixing of light polarization occurs. After a ray light passes through a polarizer, its Jones vector should have the following form:

$$\begin{bmatrix} E_x \\ E_y \end{bmatrix} = \begin{bmatrix} \mathbf{A}\exp(i2\pi ft/2) + \boldsymbol{\beta}\exp(-i2\pi ft/2) \\ \mathbf{B}\exp(-i2\pi ft/2) + \boldsymbol{\alpha}\exp(i2\pi ft/2) \end{bmatrix} = \begin{bmatrix} |\mathbf{A}|\exp(i2\pi ft/2 + i\phi_A) + |\boldsymbol{\beta}|\exp(-i2\pi ft/2 + i\phi_\beta) \\ |\mathbf{B}|\exp(-i2\pi ft/2 + i\phi_B) + |\boldsymbol{\alpha}|\exp(i2\pi ft/2 + i\phi_\alpha) \end{bmatrix}, \tag{18}$$

where $\boldsymbol{\alpha}$ and $\boldsymbol{\beta}$ are the complex amplitudes of components with vibration directions along the x and y axes, and they are mixed into E_y and E_x, respectively; f is the frequency difference produced by the EO. For clarity the coefficients related to the reference signal and the test signal are expressed by the subscripts r and t, respectively. Based on the derivations of Jones calculus, the intensity of the reference signal can be written as[17,18]

$$\begin{aligned} I_r = {} & |\mathbf{A}_r|^2 + |\mathbf{B}_r|^2 + |\boldsymbol{\alpha}_r|^2 + |\boldsymbol{\beta}_r|^2 + 2|\mathbf{A}_r||\boldsymbol{\alpha}_r|\cos(\phi_{Ar} - \phi_{\alpha r}) \\ & + 2|\mathbf{B}_r||\boldsymbol{\beta}_r|\cos(\phi_{Br} - \phi_{\beta r}) + 2|\mathbf{A}_r||\mathbf{B}_r|\cos(2\pi ft \\ & + \phi_{Ar} - \phi_{Br}) + 2|\mathbf{A}_r||\boldsymbol{\beta}_r|\cos(2\pi ft + \phi_{Ar} - \phi_{\beta \mathbf{r}}) \\ & + 2|\boldsymbol{\alpha}_r||\mathbf{B}_r|\cos(2\pi ft + \phi_{\alpha r} - \phi_{Br}) \\ & + 2|\boldsymbol{\alpha}_r||\boldsymbol{\beta}_r|\cos(2\pi ft + \phi_{\alpha r} - \phi_{\beta r}). \end{aligned} \tag{19}$$

Because both interfering beams (s and p polarization) propagate along the same optical path and they are influenced by the reflection from the beam splitter, we have $|\mathbf{A}_r| \neq |\mathbf{B}_r|$, $\phi_{Ar} - \phi_{Br} = \phi_{\alpha r} - \phi_{\beta r} = \phi_{\mathrm{BS}}$, $\phi_{Ar} = \phi_{\alpha r}$, $\phi_{Br} = \phi_{\beta r}$, and $|\mathbf{A}_r|/|\mathbf{B}_r| = |\boldsymbol{\alpha}_r|/|\boldsymbol{\beta}_r|$. Consequently Eq. (19) can be rewritten as

$$\begin{aligned} I_r = {} & (|\mathbf{A}_r| + |\boldsymbol{\alpha}_r|)^2 + (|\mathbf{B}_r| + |\boldsymbol{\beta}_r|)^2 + 2(|\mathbf{A}_r||\mathbf{B}_r| + |\mathbf{A}_r||\boldsymbol{\beta}_r| \\ & + |\boldsymbol{\alpha}_r||\mathbf{B}_r| + |\boldsymbol{\alpha}_\mathbf{r}||\boldsymbol{\beta}_r|)\cos(2\pi ft + \phi_{\mathrm{BS}}). \end{aligned} \tag{20}$$

Similarly, the intensity of the test signal is given as

$$\begin{aligned} I_t = {} & |\mathbf{A}_t|^2 + |\mathbf{B}_t|^2 + |\boldsymbol{\alpha}_t|^2 + |\boldsymbol{\beta}_t|^2 \\ & + 2(|\mathbf{A}_t||\boldsymbol{\alpha}_t| + |\mathbf{B}_t||\boldsymbol{\beta}_t|)\cos\phi \\ & + 2\{[(|A_t||\mathbf{B}_t| + |\boldsymbol{\alpha}_t||\boldsymbol{\beta}_t|)\cos\phi + |\mathbf{A}_t||\boldsymbol{\beta}_t| + |\mathbf{B}_t||\boldsymbol{\alpha}_t|]^2 \\ & + (|\mathbf{A}_t||\mathbf{B}_t| - |\boldsymbol{\alpha}_t||\boldsymbol{\beta}_t|)^2\sin^2\phi\}^{1/2}\cos(2\pi ft - \phi'), \end{aligned} \tag{21}$$

where

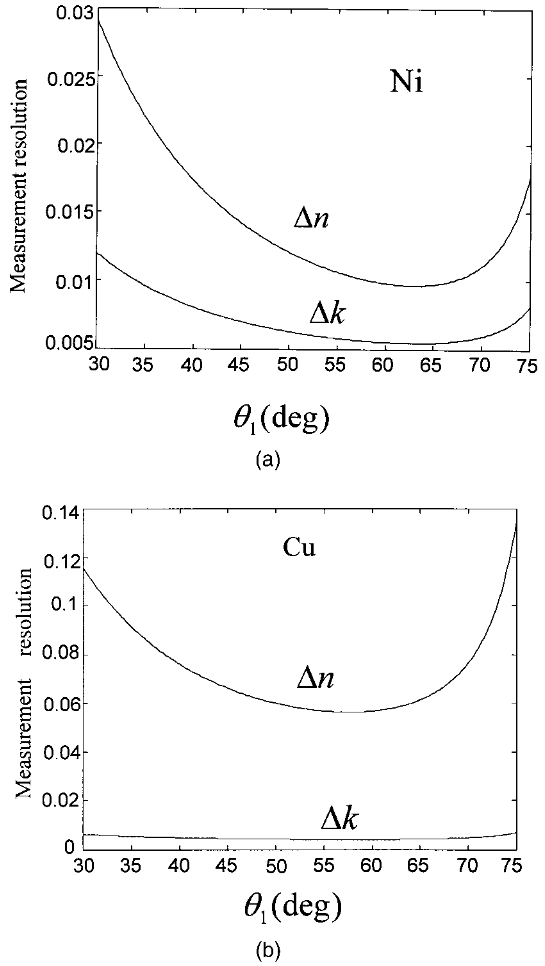
$$\phi' = \tan^{-1}\left[\frac{(|\mathbf{A}_t||\mathbf{B}_t| - |\boldsymbol{\alpha}_t||\boldsymbol{\beta}_t|)\sin\phi}{|\mathbf{A}_t||\boldsymbol{\beta}_t| + |\mathbf{B}_t||\boldsymbol{\alpha}_t| + (|\mathbf{A}_t||\mathbf{B}_t| + |\boldsymbol{\alpha}_t||\boldsymbol{\beta}_t|)\cos\phi}\right], \tag{22}$$

and we obtain it by comparing the test signal [Eq. (21)] with the reference signal [Eq. (22)]. Hence the polarization mixing error is

$$\Delta\phi_m = \phi' - \phi. \tag{23}$$

In our experiments, the extinction ratio of the polarizer (Japan Sigma Koki, Ltd.) is 1×10^{-5}, that is, $|\boldsymbol{\alpha}|/|\mathbf{A}| = |\boldsymbol{\beta}|/|\mathbf{B}| = 0.0032$. Introducing these data into Eqs. (22) and (23) results in the relation curve of $\Delta\phi_m$ versus ϕ' depicted in Fig. 4. We obtain the values of ϕ_1 and ϕ_2 in Table 1 by modifying the corresponding values of ϕ' with the relation curves shown in Fig. 4. With this modification, the total error of the phase difference can be decreased to 0.03°.

Consequently we can obtain the curves of measure-



(a)

(b)

Fig. 5. Measurement resolution versus θ_1 for Δn and Δk of (a) nickel and (b) copper at $\theta_2 = 80°$.

ment resolution versus θ_1 for Δn and Δk of nickel and copper at $\theta_2 = 80°$ by substituting $|\Delta\phi_1| = |\Delta\phi_2| = 0.03°$ into relations (14) and (15). The results are shown in Figs. 5(a) and 5(b), respectively. Obviously, the best resolution can be obtained when θ_1 is close to 60°. Δn and Δk corresponding to our experimental conditions were calculated and are included in Table 1.

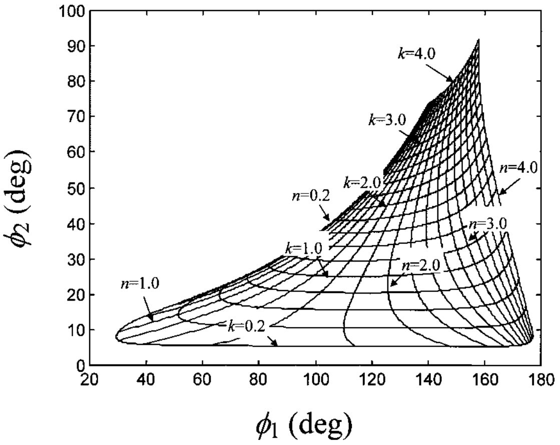


Fig. 6. Constant n and k as functions of ϕ at $\theta_1 = 60°$ and $\theta_2 = 80°$, where the values of n and k are 0.2 to 4 in steps of 0.2.

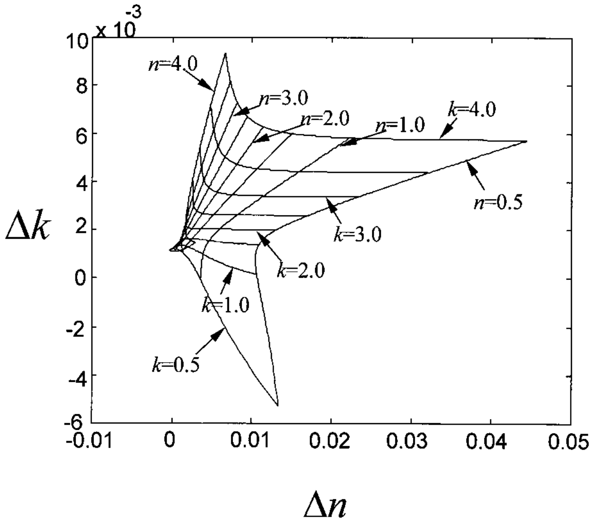


Fig. 7. Constant n and k as functions of Δn and Δk at $\theta_1 = 60°$ and $\theta_2 = 80°$, where the values of n and k are 0.5 to 4 in steps of 0.5.

To investigate the effects of experimental conditions on the measurements we depict in Fig. 6 the curves of constant n and k as functions of phase difference ϕ for $\theta_1 = 60°$ and $\theta_2 = 80°$. In the figure the values of n and k are shown from 0.2 to 4.0 in steps of 0.2. According to Humphreys-Owen,[4] the sensitivity to the experimental conditions is indicated by the spacing between contours: If the spacing is large, the sensitivity is good and vice versa. It is obvious that our experimental conditions are useful if n and k are small. For good sensitivity, it is better to choose optimum incidence angles with the method proposed by Logofatu *et al.*[19] Moreover, curves of constant n and k as functions of Δn and Δk are shown in Fig. 7. We obtained them by substituting the experimental conditions $\theta_1 = 60°$, $\theta_2 = 80°$, and $|\Delta\phi_1| = |\Delta\phi_2| = 0.03°$ into relations (14) and (15). In Fig. 7 the values of n and k are from 0.5 to 4 in steps of 0.5. It can be seen that both Δn and Δk are smaller than 1×10^{-2} for the test materials with $n > 1$ and $k < 2$. Furthermore, this method is highly stable against air turbulence because of its common path configuration.

5. Conclusion

Based on Fresnel's equations and the use of heterodyne interferometry, we have developed a new method for measuring a complex refractive index. The phase difference between s and p polarization of the reflected light from an absorbing material is measured with a heterodyne interferometer. Two phase differences, corresponding to two different incidence angles, are measured. These two phase differences are substituted into Fresnel's equations to yield a set of simultaneous equations. Then the equations are solved by use of a personal computer by a numerical analysis technique, and the complex refractive index of the material can be estimated. The method has several merits, including a simple optical setup, easy operation, high stability, high measurement accuracy, and rapid measurement. Its feasibility has been demonstrated.

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