



Measurement of refractive index variation of liquids by surface plasmon resonance and wavelength-modulated heterodyne interferometry

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ABSTRACT

In this study an alternative method based on surface plasmon resonance is proposed for in-situ monitoring of variation in the refractive index of a test sample. A wavelength-modulated light source and an unequal-path-length optical configuration heterodyne interferometer are used to detect the phase difference change, which can then be used to estimate the change in the refractive index of a test sample. The experimental results demonstrate a phase stability of 0.02° . The resolution power of the refractive index is 1.5×10^{-6} RIU. This method has several advantages over previously used methods such as simple optical setup, easier operation in real time, and low cost.

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1. Introduction

The surface plasmon resonance (SPR) method is a highly sensitive technique for evaluating the thickness of the test medium on a metal surface [1]. Numerous real-time biomolecular interaction detection schemes have been developed based on the SPR method. Such schemes have been applied in a variety of fields such as pharmaceutical development and life sciences [2,3]. In addition, since SPR also provides ultra-high sensitivity for detecting tiny changes in the refractive index (RI) or other quantities which can be converted into an equivalent RI, it is also widely applied for the measurement of temperature [4], pressure [5], displacement [6], angle [7], and so on.

SPR is primarily measured by the detection of the resonance angle θ_r , resonance wavelength λ_r or optical phase ϕ . The variation in the optical phase ϕ is much more abrupt than either the resonance angle or wavelength as the SPR is excited [8–10], which is the phase-based techniques have been so widely used for characterizing tiny variations in the samples. Several novel approaches to measuring the phase during SPR excitation have been shown to be effective. For example, the Mach–Zehnder interferometer (MZI) combines phase shift techniques to determine the phase variation of SPR [11,12]. The piezoelectric actuator, the main device providing step phase shifting in the MZI, should be calibrated for precision step phase displacement. Additionally, the non-common path configuration means that the interferometer may suffer from environmental disturbances, so sophisticated control is required to maintain the system stability. The heterodyne interferometer (HI) [9,13,14] detects the SPR phase by using a Zeeman laser or optical modulator, such as an acousto-optic

modulator or electro-optic modulator [3,4,6,7,13–16]. It has been reported that these HI phase detection techniques offer high measurement performance, high sensitivity and high resolution in real time. Moreover, the common path configuration of the HI makes it very stable. However, these systems require complex and expensive electronic equipment, such as modulation and demodulation instruments. Also, because steep phase variation occurs only in a small region within the plasmon resonance dip, the MZI and HI SPR schemes offer a limited dynamic range [17]. In this study, a SPR-based method for the measurement of the RI change of liquids with wavelength-modulated heterodyne interferometry [18] is proposed. We demonstrate that we can achieve heterodyning with wavelength modulation of a laser diode source by means of injection current modulation, combined with an unbalanced optical path difference configuration. The variation in the signal's phase difference corresponding to the SPR's phase change can be measured by our heterodyne phase detection method. The RI change of liquids can then be estimated from the measured phase difference variation. There is no acousto-optical, electro-optical modulator or Zeeman laser in our system, making it easy to operate in real time, and also lowering the cost. In addition, the dynamic range of our method is larger than that of older methods. The experimental results show the feasibility of this proposed technique. With it we can achieve a resolution of 1.5×10^{-6} RIU.

2. Principles

2.1. Wavelength-modulated heterodyne light source

As shown in Fig. 1, when the linear polarized light of the laser diode (LD) passes through a Michelson interferometer, with the

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optical path difference l , the Jones vector of the light can be written as

$$E_o = \begin{pmatrix} \exp(ikl/2) \\ \exp(-ikl/2) \end{pmatrix}. \quad (1)$$

Here $k = 2\pi/\lambda_o$ is the wave number and λ_o is the center wavelength of the LD. If LD is driven by a sawtooth current signal with frequency f , then the wavelength is modulated in sawtooth form $\lambda(t) = \lambda_o + \Delta\lambda \cdot f \cdot t$, because the wavelength is proportional to the driving current [18]. $\Delta\lambda (\ll \lambda_o)$ is the modulation depth of the wavelength. In this way, the phases of the p and s polarization states in Eq. (1) can thus be modulated and written as

$$\frac{kl}{2} = \frac{\pi l}{(\lambda_o + \Delta\lambda f t)} \approx \frac{\pi l}{\lambda_o} - \frac{\pi l \Delta\lambda f t}{\lambda_o^2} = \frac{\phi_o}{2} - \frac{\omega t}{2}. \quad (2)$$

Here $\phi_o = 2\pi l/\lambda_o$ is the initial optical phase and $\omega = 2\pi l \Delta\lambda f/\lambda_o^2$ is the optical frequency difference between the p and s polarization states. Therefore, from Eqs. (1) and (2), we obtain the heterodyne light source E_h which can be written as

$$E_h = \begin{pmatrix} \exp[i(\phi_o - \omega t)/2] \\ \exp[i(-\phi_o + \omega t)/2] \end{pmatrix}. \quad (3)$$

2.2. Phase difference between s and p polarized light resulting from SPR

As shown in Fig. 1, the reflection coefficient of the p and s polarization in Kretschmann configuration can be expressed as [13]

$$r_q = \frac{r_{12}^q + r_{23}^q \exp(i2k_{z2}d)}{1 + r_{12}^q \cdot r_{23}^q \exp(i2k_{z2}d)} = |r_q| \exp(i\phi_q), \quad (4)$$

where q ($=s$ or p) represents the s or p polarization state; k_{z2} is the wave vector in the thin gold film; and d is the thickness of the thin gold film. Hence, $r_{m,m+1}^q$ is the Fresnel reflection coefficient at the interface between the m^{th} and the $m+1^{\text{th}}$ media ($m=1, 2$). The subscripts 1, 2 and 3 represent the prism, thin gold film and test sample, respectively. Their refractive indices are n_1, n_2 and n_3 . It is well known that the reflection coefficients r_p and r_s are both complex numbers, and that ϕ_p and ϕ_s indicate the optical phase of the p and s

polarizations coming from the reflection at the boundary surface under the conditions of SPR. The optical phase difference $\phi = \phi_p - \phi_s$ between the p and s polarizations is strongly dependent on the refractive index n_3 of the test sample and the incident angle θ_i . The Jones matrix of the SPR sensing area for reflection light can be written as

$$J_{\text{SPR}} = \begin{bmatrix} |r_p| \exp(i\phi_p) & 0 \\ 0 & |r_s| \exp(i\phi_s) \end{bmatrix}. \quad (5)$$

The optical phase difference ϕ between the p and s polarizations can be measured by heterodyne or phase shifting interferometry to determine the refractive index of the sample or the bio-interaction on the surface of the gold film.

2.3. Circular heterodyne interferometer for phase difference measurements

For convenience, the $+z$ -axis is chosen to follow the direction of light propagation and the $+x$ -axis is along the horizontal plane. The heterodyne light E_h (Eq. (3)) passes through a quarter-wave plate Q_3 whose fast axis is set to be at 45° with respect to the x -axis, so that the light becomes a circular heterodyne light. Then light reflected from the SPR sensing area is divided into two paths by a beam splitter BS. These two light beams pass through linear polarizers P_1 and P_2 , with transmittance axes at 45° and -45° , respectively, before being received by photodetectors D_1 and D_2 . The amplitude of the light detected by photodetector D_1 is

$$\begin{aligned} E_1 &= P(45^\circ) \cdot J_{\text{SPR}} \cdot J_Q(45^\circ) \cdot E_h \\ &= \frac{1}{2} \begin{pmatrix} 1 & 1 \\ 1 & 1 \end{pmatrix} \begin{pmatrix} |r_p| e^{i\phi_p} & 0 \\ 0 & |r_s| e^{i\phi_s} \end{pmatrix} \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & i \\ i & 1 \end{pmatrix} \begin{pmatrix} e^{i(\phi_o - \omega t)/2} \\ e^{i(-\phi_o + \omega t)/2} \end{pmatrix} \\ &= \frac{1}{2\sqrt{2}} \left[(|r_p| e^{i\phi_p} + i|r_s| e^{i\phi_s}) e^{i(\phi_o - \omega t)/2} \right. \\ &\quad \left. + (i|r_p| e^{i\phi_p} + |r_s| e^{i\phi_s}) e^{i(-\phi_o + \omega t)/2} \right] \cdot \begin{pmatrix} 1 \\ 1 \end{pmatrix}. \end{aligned} \quad (6)$$

Its intensity is

$$I_1 \propto |E_1|^2 = |r_p|^2 + |r_s|^2 + \sqrt{A^2 + B^2} \cos[\phi_o - \omega t - \tan^{-1}(B/A)]. \quad (6a)$$

Here $A = 2|r_p||r_s|\cos\phi$, $B = |r_p|^2 - |r_s|^2$, where ϕ is the SPR's phase difference between the p and s polarizations. The last term $-\tan^{-1}(B/A)$ in Eq. (6a) is the signal phase which results from the complex reflective coefficient of SPR. Similarly, the amplitude and intensity of the light beam on the photodetector D_2 are

$$\begin{aligned} E_2 &= P(-45^\circ) \cdot J_{\text{SPR}} \cdot J_Q(45^\circ) \cdot E_h \\ &= \frac{1}{2\sqrt{2}} \left[(|r_p| e^{i\phi_p} - i|r_s| e^{i\phi_s}) e^{i(\phi_o - \omega t)/2} \right. \\ &\quad \left. + (i|r_p| e^{i\phi_p} - |r_s| e^{i\phi_s}) e^{i(-\phi_o + \omega t)/2} \right] \cdot \begin{pmatrix} 1 \\ -1 \end{pmatrix}, \end{aligned} \quad (7)$$

and

$$I_2 \propto |E_2|^2 = |r_p|^2 + |r_s|^2 - \sqrt{A^2 + B^2} \cos[\phi_o - \omega t + \tan^{-1}(B/A)]. \quad (7a)$$

It is obvious that these two signals (Eqs. (6a) and (7a)) have the same initial phase ϕ_o , which results from the optical path difference in the Michelson interferometer. In addition, their last terms $[\pm \tan^{-1}(B/A)]$

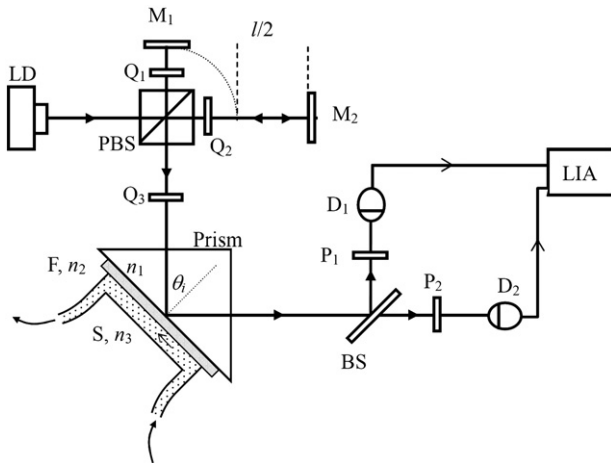


Fig. 1. Schematic diagram of the wavelength-modulated circular heterodyne interferometer for SPR phase difference measurements: LD, laser diode; M, mirror; Q, quarter-wave plate; PBS, polarizing beam splitter; F, gold film; S, sample; BS, beam splitter; P, polarizer; D, photodetector; LIA, lock-in amplifier.

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