SPR sensor by method of electro-optic phase modulation and polarization interferometry

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We propose a surface plasmon resonance (SPR) sensor based on phase modulation and polarization interferometry, both of which provide a refractive index (RI) resolution of the same order as that of SPR sensors of the phase type. And it has a wide dynamic range and insensitivity of RI resolution to the thickness of metal films as that of the intensity type SPR sensors. In this letter, we choose electro-optic (EO) phase modulation instead of the angle modulation. We demonstrate theoretically that with the EO phase modulation, our sensor could provide a better RI resolution.

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Surface plasmon resonance (SPR) has been studied extensively in the bio-optical field since it had been introduced by Nylander $et \ al.^{[1,2]}$ in gas detection and biosensors by virtue of its promising merits: real time measurement, label free, and quantitative biosensing^[3,4]. Otto *et al.*^[5,6], each proposed their own SPR setup which was widely employed in the SPR biosensors, especially the Kretschmann setup in the attenuated total reflection (ATR) configure. As is reported, the SPR sensor with the light intensity as a measuring signal has its refractive index (RI) resolution of the order 10^{-6} due to the fluctuation of the light intensity [1,2,4,7]. A SPR sensor based on angular interrogation in the ATR configure has a better RI resolution of $5 \times 10^{-7[8]}$. A white light source especially halogen lamps is introduced for the SPR sensors with the type of wavelength interrogation by virtue of a range of wavelengths provided. But much attention has been devoted to the phase [9-14] type for a better resolution of RI within the order 10^{-8} [14] reached by the phase detection. The phase type has a narrow dynamic range of measurement and is sensitive to the thickness of the metal film although it has a very high resolution of. Therefore, our group has presented a SPR sensor based on the phase modulation^[15] and polarization interferometry technique^[18-20]. The sensor has a high resolution close to that of the SPR sensor of the phase type, a large dynamic range of measurement. And the RI resolution is insensitive to the thickness of the metal film. A RI resolution of 5×10^{-8} can be achieved by this type of SPR sensors according to our theoretical analysis^[16]. But only a RI resolution of 5.1×10^{-7} in the experiment has been achieved in our group^[17]. The main reason why the RI resolution of the theoretical analysis and that of the experiment are not in the same order is that the noises introduced by the method of angle modulation by the PZT material with the mechanical drift. We choose the electro-optic (EO) phase modulation without the mechanical drift instead of the angle modulation, expecting a high RI resolution in this letter.

Our system is shown in Fig. 1. The linearly polarized light from He-Ne laser is divided into two beams by a polarizing beam splitter (PBS). The beam irradiated onto the detector D1 is for a reference so that one can monitor the fluctuation of the intensity of the light from the laser. The polarizer P1 could be rotated to adjust the light intensity due to the linearly polarized light from the laser so that the detectors work in the linear region. And the other beam is for the phase modulation by the EO phase modulator (EOM) after through the polarizer P2 which is for the EOM requiring the polarization direction of linearly polarized light to be modulated 45° with respect to the crystal axis of the lithium niobate in the EOM. Each diaphragm allows only the light passing through its aperture, stopping the light passage but this. The Kretschmann SPR setupk, the 1/4 wave plate (QWP), and the polarizer P3 together realize the polarization interferometry technique. The detector D2 is used to measure the light signal irradiated on the D2, a signal generator (SG) gives one signal which is divided into two signals with the same frequency and



Fig. 1. Experimental scheme for the SPR sensor by the method of EO phase modulation and polarization interferometry.

amplitude. One triggers the modulation of the input light, the other for an external reference for the lockin amplifier. The lock-in amplifier is used to measure the phase of the signal detected by the detector D2 with a signal from the signal generator as a reference. We provide the theoretical analysis of the sensor as follows.

We calculate the result with the help of Jones matrices $\mathrm{as}^{[16]}$

$$\mathbf{J}_{\rm EOM} = \begin{bmatrix} e^{-iM\sin(\omega t)} & 0\\ 0 & e^{iM\sin(\omega t)} \end{bmatrix}, \qquad (1)$$

$$\mathbf{J}_{\rm SPR} = \begin{bmatrix} r_{\rm p} \mathrm{e}^{\mathrm{i}\,\boldsymbol{\Phi}_{\rm p}} & 0\\ 0 & r_{\rm s} \mathrm{e}^{\mathrm{i}\,\boldsymbol{\Phi}_{\rm s}} \end{bmatrix},\tag{2}$$

$$\mathbf{J}_{\rm QWP} = \frac{1}{\sqrt{2}} \left[\begin{array}{cc} 1 & \mathrm{i} \\ \mathrm{i} & 1 \end{array} \right],\tag{3}$$

$$\mathbf{J}_{P3} = \begin{bmatrix} \cos^2 \beta & \frac{1}{2} \sin(2\beta) \\ \frac{1}{2} \sin(2\beta) & \sin^2 \beta \end{bmatrix}, \quad (4)$$

we can calculate the \mathbf{E} vector easily as

$$\mathbf{E} \propto \mathbf{J}_{P3} \mathbf{J}_{QWP} \mathbf{J}_{SPR} \mathbf{J}_{EOM} \mathbf{E}_{0} = \begin{bmatrix} \cos^{2} \beta & \frac{1}{2} \sin(2\beta) \\ \frac{1}{2} \sin(2\beta) & \sin^{2} \beta \end{bmatrix}$$
$$\cdot \frac{1}{\sqrt{2}} \begin{bmatrix} 1 & \mathbf{i} \\ \mathbf{i} & 1 \end{bmatrix} \begin{bmatrix} r_{p} e^{\mathbf{i} \boldsymbol{\Phi}_{p}} & 0 \\ 0 & r_{s} e^{\mathbf{i} \boldsymbol{\Phi}_{s}} \end{bmatrix}$$
$$\cdot \begin{bmatrix} e^{-\mathbf{i} M \sin(\omega t)} & 0 \\ 0 & e^{\mathbf{i} M \sin(\omega t)} \end{bmatrix} \mathbf{E}_{0}.$$
(5)

The final result after simple manipulation is

 $\mathbf{E} \propto$

$$\begin{bmatrix} r_{\rm p} \cos\beta e^{i[\varPhi_{\rm p}+\beta-M\sin(\omega t)]} + r_{\rm s} \cos\beta e^{i[\varPhi_{\rm s}-\beta+M\sin(\omega t)+\frac{\pi}{2}]} \\ r_{\rm p} \sin\beta e^{i[\varPhi_{\rm p}+\beta-M\sin(\omega t)]} + r_{\rm s} \sin\beta e^{i[\varPhi_{\rm s}-\beta+M\sin(\omega t)+\frac{\pi}{2}]} \end{bmatrix},$$
(6)

As is known, the light intensity is $\mathbf{I} \propto \mathbf{E}^+ \mathbf{E}$, so we can obtain the final result of the light intensity, namely,

$$\mathbf{I} \propto \frac{r_{\rm p}^2 + r_{\rm s}^2}{2} + r_{\rm p} r_{\rm s} \cos\left[\boldsymbol{\varPhi}_{\rm s} - \boldsymbol{\varPhi}_{\rm p} - 2\beta + 2M\sin(\omega t) + \frac{\pi}{2}\right]. \quad (7)$$

Obviously, the polarizer P3 and QWP can be tuned so that in some condition, $\Phi_{\rm s0} - \Phi_{\rm p0} - 2\beta_0 + \frac{\pi}{2} = (2k+1)\pi$, where k is an integer, so we can expand $\cos \left[\Phi_{\rm s} - \Phi_{\rm p} - 2\beta + 2M\sin(\omega t) + \frac{\pi}{2} \right]$ around $(2k+1)\pi$, giving the result near the frequency ω as $I_{\omega} \propto Mr_{\rm p}r_{\rm s}[\Delta\Phi_{\rm s}(t) - \Delta\Phi_{\rm p}(t)]\sin(\omega t)$, where $\left\{ \Delta\Phi_{\rm p}(t) = \Phi_{\rm p}(t) - \Phi_{\rm p0} \right\}$, so the sensitivity of the system to the change of refractive index is given by

$$S = \frac{\partial I_{\omega}}{\partial n},\tag{8}$$

where I_{ω} is alternating current (AC) of the light intensity at the frequency ω . A high frequency can be introduced to reduce the 1/f noise compared to the magneto-optic modulator^[15,16].

So, the resolution of the system is given by $\delta n = \frac{\Delta n}{\Delta v} \delta v$, where δn represents the resolution of the RI units, and δv is the deviation of the lock-in amplifier which is used to measure the phase of the signal. When in a stable state, Δv is the change due to the change of the RI.

The sensitivity of the SPR sensor is represented as

$$S = \frac{\partial I_{\omega}}{\partial n} \propto M r_{\rm s} \left\{ \frac{\partial r_{\rm p}}{\partial n} \Delta \Phi_{\rm p}(t) + r_{\rm p} \frac{\partial [\Delta \Phi_{\rm p}(t)]}{\partial n} \right\}$$
$$\approx M r_{\rm s} \frac{\partial l}{\partial n}, \tag{9}$$

where $r_{\rm s}$ is assumed constant, l is the arc length^[16] and is a function of the film thickness. The scheme of the angle interrogation is shown in the work^[17] in our group. The drift of the amplitude and frequency of the signal generator as well as the mechanical drift has not been discussed in the context. So, we assume the modulation signal is

$$y = A\sin\left(\omega t\right),\tag{10}$$

and the corresponded angle light incident on the metal film is assumed to be

$$\Theta = \alpha y + \beta, \tag{11}$$

where β is the mechanical drift of the PZT, and α is a constant due to the small angle modulation stemmed from the PZT material. The drift of the amplitude and frequency of the signal generator are assumed to be random variables with a very small variance, so the angle difference can be given by

$$\delta \Theta = \frac{\partial \Theta}{\partial A} \delta A + \frac{\partial \Theta}{\partial \omega} \delta \omega + \frac{\partial \Theta}{\partial \beta} \delta \beta$$
$$= \alpha \left[(\delta A) \sin(\omega t) + A \cos(\omega t) (\delta \omega) t \right] + \delta \beta, \qquad (12)$$

where t is assumed to be in the interval $[0, \frac{2\pi}{\omega}]$, or should be changed to the interval by minus the integral number of $\frac{2\pi}{\omega}$. So we can get the result that the incident point on the film changes its behavior due to the drift of the amplitude and frequency of the signal generator as well as the mechanical drift. The uniformity of the thickness of the metal film can be defined as

$$U(x,y) = \frac{\Delta Z(x,y)}{\overline{Z}(x,y)},$$
(13)

where $\overline{Z}(x, y)$ is the mean value of Z(x, y), $\Delta Z(x, y) = Z(x, y) - \overline{Z}(x, y)$. The different values of U(x, y) in the xyz coordinate system means thickness of the metal film in xy distribution.

From the Maxwell's equations, we can obtain the complex reflection coefficient of the p and s lights $as^{[6,21]}$

$$\widetilde{r}_q = \frac{r_{01}^q + r_{12}^q \exp\left(2ik_{1z}d\right)}{1 + r_{01}^q r_{12}^q \exp\left(2ik_{1z}d\right)}, \quad q = p, s, \qquad (14)$$

S12401-2

$$r_{i,i+1}^{q} = \frac{X_{i}^{q} - X_{i+1}^{q}}{X_{i}^{q} + X_{i+1}^{q}}, \quad i = 0, 1,$$
(15)

$$X_i^{\rm p} = \frac{\varepsilon_i}{k_{iz}}, \quad X_i^{\rm s} = k_{iz}, \tag{16}$$

$$k_{iz} = \frac{\omega_0}{c} \sqrt{\varepsilon_i - \varepsilon_0 \sin^2 \theta},$$

where d is the thickness of the metal film. So we can easily conclude that the reflection ratio changes due to the drift of the amplitude and frequency of the signal (denoted by $\delta \Theta$) and the mechanical drift of the PZT (denoted by U(x, y)).

A new method of the phase modulation of the input light can be introduced instead of the angle interrogation. The incident point is fixed on the metal film without the mechanical drift avoiding the noise from the PZT. A lock-in amplifier with an external reference can be used to detect the phase of the signal avoiding the drift of the amplitude and frequency of the signal generator. EOM can be used as a electronically tunable waveplate. An appropriate oscillating signal can be applied to the modulator to obtain a phase modulated light from linearly polarized to elliptically polarized. As is shown in our previous work^[16], the expected resolution of our sensor in this letter is better than 5×10^{-8} .

In conclusion, our new SPR system has a better resolution of RI units, at least in the order of 10^{-8} . A new experiment scheme is presented. A new method of polarization modulation of input light is introduced to enhance the resolution of RI units. A high frequency can be used as a modulation frequency to reduce the random noise.

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