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Simulation and analysis of phase-sensitive surface plasmon resonance sensor based on enhanced optical transmission through arrays of nanoholes in silver films

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Abstract: A high spatial resolution, phase-sensitive Surface Plasmon Resonance (SPR) sensor based on Extraordinary Optical Transmission (EOT) is proposed to monitor the binding of organic and biological molecules to the silver surface. The 2D nanohole-array configuration is well suited for dense integration in a sensor chip. The optical geometry is collinear, which simplifies the alignment with respect to the traditional Kretschmann arrangement for SPR sensing. Various design parameters of the device have been studied by simulation. The heterodyne technique is used to improve the sensitivity. The optimization results indicate that the sensor has the advantages of achieving high resolution and a wide dynamic range simultaneously.

Key words: surface plasmon resonance sensor; extraordinary optical transmission; phase interrogation; nanohole; silver film

1　Introduction

In recent years, SPR technology has been commercialized and SPR sensors have become central tools for characterizing and quantifying biomolecular interactions both in the life sciences and pharmaceutical researches due to their capability for real-time measurement with high detection sensitivity[1-3]. Current research attention in SPR sensing has shifted to measure the SPR phase shift[4,5], as the resonant phase behaviour offers the potential in achieving an extremely high detection sensitivity. The traditional phase sensors couple light into the surface plasmon using a prism in the Kretschmann configuration, also known as the Attenuated Total Reflection (ATR) configuration. However, this method suffers from the disadvantages of large size and narrow dynamic range.

In this paper, we report a compact and wide dynamic range biosensor based on detecting the phase change of EOT light from a hole array. A periodic square array of sub-wavelength holes on a silver film is utilized as the SPR phase sensor. Compared with the typical Kretschmann configuration, the hole array configuration operates in the transmission geometry. This allows for a simpler collinear optical arrangement, thus providing a smaller pro-

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bing area and high throughput sensing[6-9]. In addition, the heterodyne technique is used for phase detection so as to realize an even high sensitivity. The unique advantage of the sensors may enhance the preference of using such devices in a number of applications especially for the integration with lab-on-chip platforms.

2 Principle of phase sensor

The structure of the silver nanohole structure is deposited on a silicon dioxide substrate as shown in Fig. 1. The period of the arrays is d, the width of the square holes is a, and the thickness of the silver film is h. The arrays of sub-wavelength holes can be fabricated by focused ion beam (FIB), electron beam lithography (EBL) or UV interference techniques. For the real-time bio-molecule sensing applications, the nanohole device should be immersed in an ethanoic solution of 11-mercaptoundecanoic acid (MUA) prior to the detection procedures[3]. When the population of bio-molecules immobilized on the sensor surface increases, the effective refractive index of the thin surface layer (also containing MUA and protein) increases accordingly. The change of refractive index will then results in a phase shift in the transmitted beam in the far field, thus leading to the possibility of quantitative detection of the density of the immobilized bio-molecular material without using any fluorescence tag.

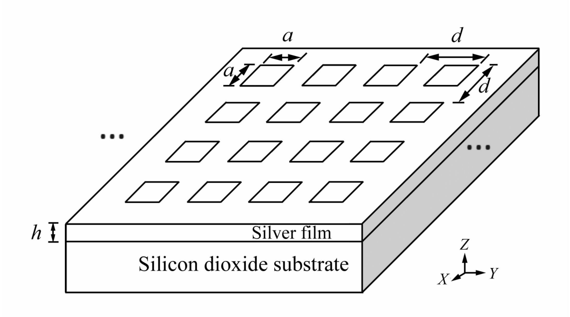

Fig. 1 Schematic of proposed nanohole array SPR phase sensor (a = hole size, d = hole period, h = hole depth).

Fig. 2 shows an experimental phase interrogation scheme based on a heterodyne interferometric system, which offers the benefits of detecting only time-varying signals and a high noise rejection capability by using the long integration time[10-13]. A typical heterodyne interferometer uses an acousto-optic modulator to impose a frequency shift on the input laser beam. When the frequency-shifted reference beam interferes with the signal beam, the phase of the beat frequency, which may be readily measured with the lock-in technique, will provide the phase reading introduced by the sensor device.

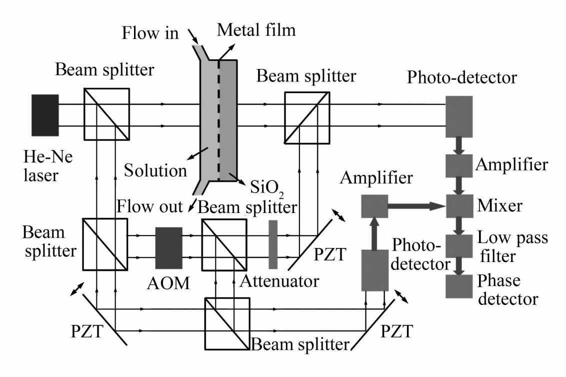

Fig. 2 Optical setup for SPR phase shift measurement.

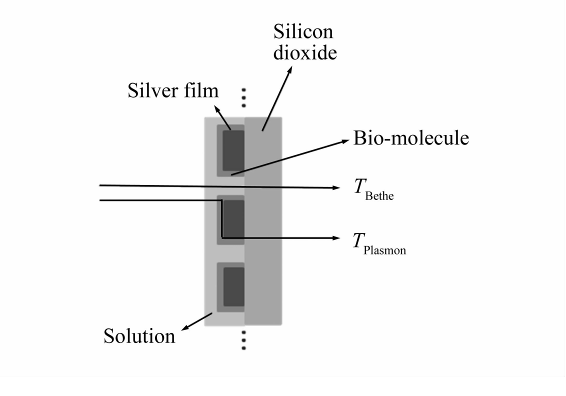

Fig. 3 Cross-section of proposed phase-sensitive SPR sensor.

In order to appreciate the underlying physics of the phase sensor, we use a simple model. As shown schematically in the enlarged area of Fig. 3, the extraordinary optical transmission effect may be separated into two contributions. The first one corresponds to the directive transmission of the incoming field through the holes, i. e. the Bethe-type diffraction regime with the transmission coefficient represented by T_{Bethe}, which is wavelength dependent and proportional to the identity matrix. The second contribution, with a transmission coefficient described by T_{Plasmon}, corresponds to the resonant part of the transmission matrix and is related to the plasmonic effect. In the present case, T_{Plasmon} is the main contribution which provides the phase change in the device.

The resonant transmission process may be described by a four-step process[14-17]: (i) the incident plane wave is converted into a surface wave at a given point scatterer; (ii) the surface wave propagates on the surface of the hole array and builds up several constructive interference modes according to the propagation distance; (iii) the surface wave is coupled into one of the holes and is reflected backwards and forwards several times in the hole, thus resulting in a constructive interference effect; and (iv) the surface waves within the hole array also produce constructive interference between them, and some energy will be re-emitted from the system as a plane wave. The four-step process indicates that when the effective refractive index of the dielectric in the vicinity of the holes experiences a change, the resonance parameters will also shift accordingly, thus leading to a sharp change in the phase of the radiation which drives the resonance. Therefore, the maximum change of the phase should occur in the region which is close to the resonant point, while the resonance parameters are dependent on the choices of hole periodicity, shape and size of the holes and thickness of the metal.

3　Simulation results and discussion

We have performed a series of simulation experiments by varying the hole period, hole shape and silver film thickness, respectively, in order to find the optimized device parameters. FDTD Solutions (Lumerical Solutions, Inc) with a minimum 1 nm mesh size is used to study the device structure. In order to shorten the simulation time, an area with just one square hole is meshed and the periodic boundary conditions are used around the hole. We use a plane wave with a wavelength of 632. 8 nm and the propagation direction is normal to the structure. Perfectly matched layer (PML) boundary conditions with 200 layers are used in the source directions. We assume the total thickness of the receptor layer and the immobilized molecules is 5 nm, and the refractive index of the surrounding aqueous medium is taken as 1. 33 (water). If the effective refractive index of this composite layer is changed from 1. 33 to 1. 50, the phase variation of the transmission beam in the far field, which passes passing through a hole array (typically 100 × 100) can be calculated accordingly. Because the hole period and the metal thickness are the two main parameters affecting the resonance on the film surface and the resonance in the holes, respectively, we can fix one parameter and change the other to obtain the optimal values of these two parameters for the largest phase change when the effective refractive index is varied from 1. 33 to 1. 50. In the present case, we first fix the silver film thickness at 108 nm and find the relationship between the hole period and the phase change as shown in Fig. 4. Here we can see from the sharp

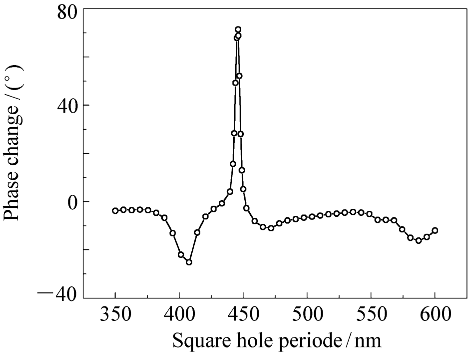

Fig. 4　Square hole period versus phase with paratemers as $n = 1.33 \sim 1.50$, $h = 108$ nm, and $d = 446$ nm.

resonance peaks that the optimal value of the hole period is 446 nm.

From the theory of the EOT, we can explain why the phase change is the largest when the hole period is 446 nm. As we have demonstrated, the maximum phase change should occur near the resonant region. The SPR resonance condition for the normally incident light is given by Ref. [14]

$$(i^2 + j^2)^{1/2}\lambda_{\mathrm{spp}} = d\mathrm{Re}\left[\left(\frac{\varepsilon_{\mathrm{eff}}\varepsilon_{\mathrm{m}}}{\varepsilon_{\mathrm{eff}} + \varepsilon_{\mathrm{m}}}\right)^{1/2}\right], \quad (1)$$

where i and j are integers, λ_{spp} is the wavelength of the incident light, d is the period of the array, ε_{m} is the effective dielectric constant of the silver film, and $\varepsilon_{\mathrm{eff}}$ is the effective dielectric constant at the metal-dielectric interface. The effective dielectric constant can be estimated by performing a weighted average within the extension l of the evanescent SP mode into the dielectric (z direction), according to Ref. [15]

$$\varepsilon_{\mathrm{eff}} = \frac{2}{l}\int_0^{\infty} \varepsilon(z)\exp\left(-\frac{2z}{l}\right)\mathrm{d}z. \quad (2)$$

According to Eq. (1), when $\lambda_{\mathrm{spp}} = 632.8$ nm, a resonant peak occurs when the hole period is 429.7 nm assuming $\varepsilon_{\mathrm{eff}} = 1.33$ and $\varepsilon_{\mathrm{m}} = -17.65 + 0.50\mathrm{i}$. Considering the effective refractive index, $\varepsilon_{\mathrm{eff}}$ must be greater than 1.33 due to the existence of the MUA and protein according to Eq. (2). However, the effective permittivity of the metal ε_{m} must be a little smaller than the permittivity of the pure silver $-17.65 + 0.50\mathrm{i}$ due to the existences of the holes. Therefore, the resonant period should be close to 451.3 nm. Our simulation results show that the maximum in the phase change occurs when the period of the hole is 446 nm which is reasonable considering the uncertainties in the model.

The silver layer thickness also plays an important role in the calculation of phase change. We first fix the period at the optimized value, i. e. 446 nm, and then we vary the silver layer thickness and monitor the phase change. The results are shown in Fig. 5 where we can see that the largest phase changes occur at 110 nm thickness. The physical mechanism for such a resonant peak to occur at 110 nm is due to the surface plasmon trapped and oscillating inside the hole, thereby building up constructive interference[17]. If we assume that the surface plasmon in the hole propagates along an interface between an infinite insulator and a metal despite the fact that it is a metal/insulator/metal (MIM) heterostructure, then the propagation length of a surface plasmon with 2π phase change is calculated as 451.3 nm. Since four times 110 nm equals 440 nm which is close to 451.3 nm, we can conclude that the plasmon in the hole should experience constructive interference if the silver thickness is 110 nm. This also means that when the constructive interference is no longer possible due to the change of refractive index in the surrounding medium, the phase changes rapidly. In addition, we also explored the effect of varying the width of the holes, which has the obvious effect of changing the effective permittivity of the MIM heterostructure. The phase change in the range of refractive index between 1.33 and 1.50 is shown in Fig. 6.

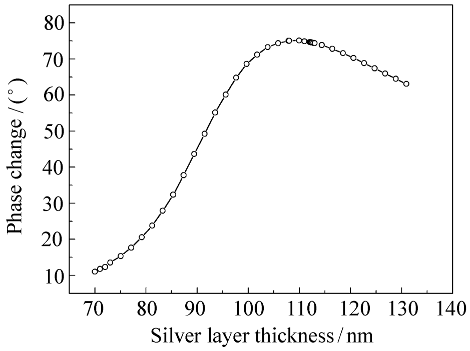

Fig. 5 Silver layer thickness versus phase change with parameters as $n = 1.33 \sim 1.50$, $h = 446$ nm, and $d = 153$ nm.

Above simulation results indicate that if we use 153 nm as the hole width, 446 nm as the hole period and 110 nm as the silver film thickness, these parameter values correspond to the largest value of the phase change which can be achieved. For a maxi-

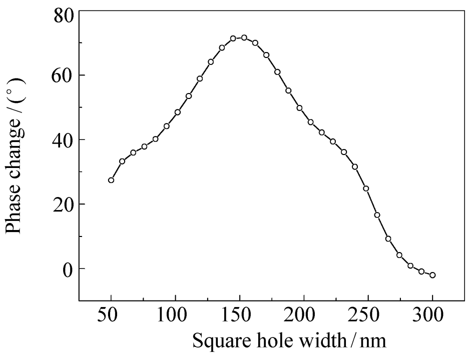

Fig. 6 Square hole width versus phase with parameters as $n = 1.33 \sim 1.55$, $h = 110$ nm, and $d = 446$ nm.

mum refractive index shift of 0.17 in the range of 1.33 and 1.50, the phase change is 75.1° as shown in Fig. 7. A feature of Fig. 7 is that the phase change is quite linear over the entire refractive index range of 0.17, whereas the lack of phase linearly is a known problem for the common Kretschmann configuration.

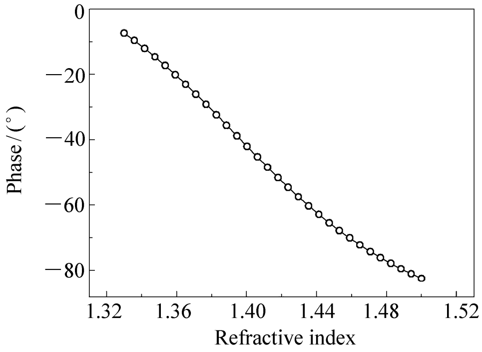

Fig. 7 Refractive index versus phase change with parameters as $a = 153$ nm, $h = 110$ nm, and $d = 446$ nm, which are optimized.

As the phase detection using a heterodyne technique and assuming that the interferometer is operating at its most sensitive point (which corresponds to zero output for the balanced detector arrangement), the photon noise equivalent displacement δx_N can be calculated by the following formula[18]

$$\delta x_N = \frac{\lambda}{4\pi}\left(\frac{h\nu\Delta f}{\eta W_s}\right)^2, \qquad (3)$$

where we assume the wavelength $\lambda = 632.8$ nm and the quantum efficiency η is 70%. For a signal power $W_s = 2.5$ W and a bandwidth $\Delta f = 1$ Hz, we find $\delta x_N = 0.0213$ pm, so that the detection resolution is $(1.93 \times 10^{-6})°$. The calculated sensitivity limit of an optimal silver device is 4.37×10^{-9} RIU.

4 Conclusions

In summary, we have demonstrated the simulation results of arrays of nanoholes in silver films as SP-based phase sensors for the adsorption of biomolecules. The sensitivity of this substrate is comparable to other SP systems and the dynamic range is also quite wide. The arrays of sub-wavelength holes investigated here are only a few micrometers in length, and the detection is made in the transmission mode. These features present that this substrate is ideal for miniaturization and integration as detection systems in microfluidic architectures and lab-on-chip devices.

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