



Biomolecular binding dynamics in sensors based on metallic photonic crystals

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ABSTRACT

We investigated the evolution dynamics of sensor signal for the binding of the bovine serum albumin (BSA) molecules to the metallic photonic crystals (MPCs). Measurements on the BSA/phosphate solution indicate a sensitivity as high as 1 ng/ml of the MPC sensor device. The experimental results show a highly nonlinear relationship between the amplitude of the sensor signal and the BSA concentration, where the binding rate increases with the increase in the concentration of the BSA/phosphate solution. Furthermore, there exists an upper limit of the concentration of the BSA/phosphate solution as low as 1 mg/ml for sensor applications, which is determined such that no refractive index change is introduced due to the increase in the solution concentration.

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1. Introduction

Various biosensors based on localized surface plasmon resonance (LSPR) [1–5] or particle plasmon resonance (PPR) [6,7] using nanostructured noble-metal photonic crystals or nanoparticles have been demonstrated for the detection of molecular concentration [8] or bioreaction processes through biomolecular-binding events [9–11]. All of these devices make use of the strong and environmentally sensitive response of plasmonic nanostructures due to the change of the dielectric constant of medium surrounding or binding to the surface of the nanostructured metals. Sensors based on the coupling between the waveguide resonance mode and particle plasmon resonance (PPR) introduce a unique enhancement mechanism that amplifies the sensor signal significantly [12,13]. This kind of Fano-like resonance leads to narrow-band response with sharp leading or falling edges of the signal spectrum. Even a small spectral shift of plasmon resonance may result in a large difference in the derivative spectrum of the coupled mode and this is the basic principle of such sensors. These kind of metallic photonic crystal (MPCs) sensors have been applied in the detection of both bioreactions [12] and molecular concentrations in liquids [8].

Sensors based on MPCs are advantageous in real-time monitoring of bioreactions and recording status of the system at different stages, which is important in selecting materials and optimizing the experimental route for bioreactions. Bovine serum albumin

(BSA) is an important protein that has been extensively used in sensor experiments, which can be employed either as a blocking medium or as a kind of convenient biomolecules for testing the sensitivity of biosensors. In particular, it is a kind of stable biomolecule with constant properties over a long time. In this paper, we present an optical sensor based on MPCs to demonstrate its success by efficient and sensitive detection of the BSA molecular concentration in phosphate and to show the spectroscopic dynamics as the BSA molecules are bound to the surface of MPCs.

2. Fabrication and spectroscopic characterization of the MPCs

MPC structures were fabricated using solution-processable techniques, as have been reported in our previous publications [12]. Interference lithography was used to produce template photoresist (PR) gratings before a spin-coating of colloidal gold nanoparticles and subsequent annealing processes. In the practical fabrication, the PR grating was fabricated on top of an indium-tin-oxide (ITO) glass substrate, colloidal solution of gold nanoparticles with a concentration of 100 mg/ml was spin-coated onto the PR grating, and the sample was then annealed at 450 °C to remove the PR template. As a result, grating structures consisting of gold nanowire segments sitting on the ITO waveguide were produced as MPCs for biosensor applications. As has been discussed in our previous publications, the breaking of gold nanowires into segments due to the high-temperature annealing process does not influence the performance of the sensors [8].

Fig. 1(a) shows the atomic force microscopic (AFM) image of the grating structures, where the grating has a period of about

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330 nm and a modulation depth of more than 76 nm. Each grating line consists of gold nanowire segments with a length ranging from below 200 nm to longer than 5 μm . However, width of the gold nanowire segments is basically homogeneous and is smaller than 200 nm in average.

Fig. 1(b) shows a schematic drawing of the optical setup for spectroscopic measurements.

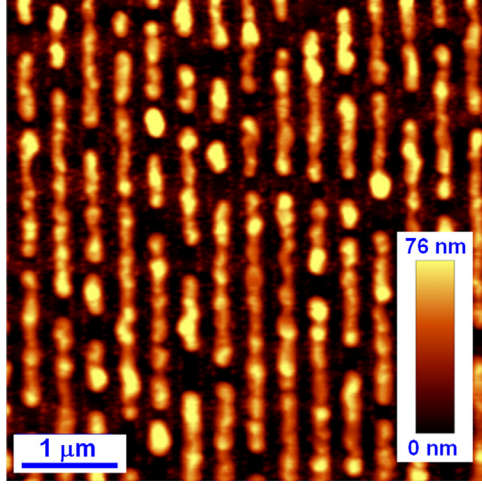


Fig. 1. AFM image of the MPC structures on top of the ITO glass substrate.

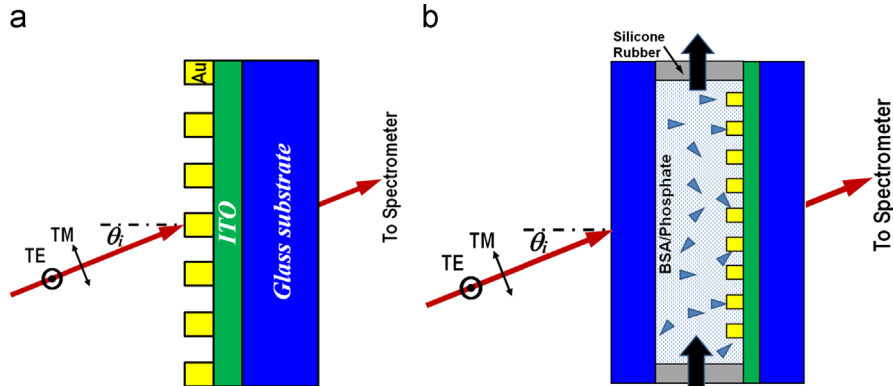


Fig. 2. Schematic illustration of the optical setup for the optical extinction measurements on: (a) angle-resolved tuning properties of MPCs in air, corresponding to measurement results in Fig. 3; (b) sensor performance in the detection of BSA/Phosphate solution, corresponding to the measurement results in Fig. 4–6. Polarization directions of the incident light are marked out by TE and TM.

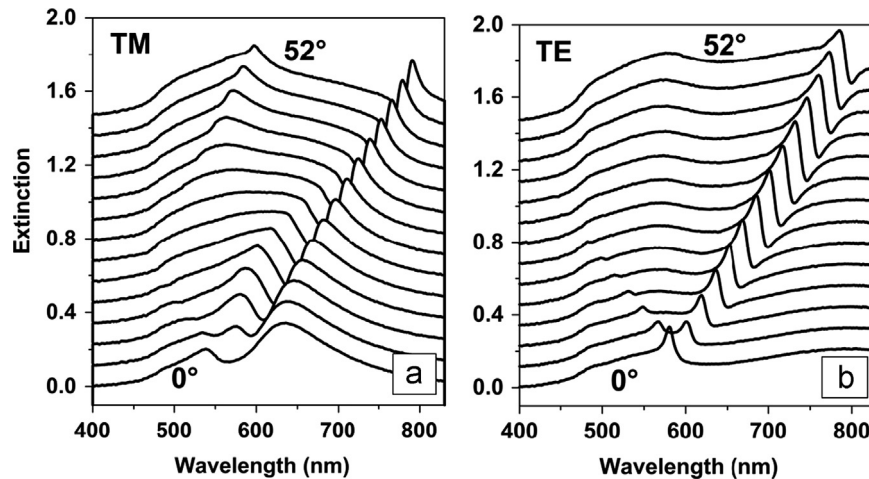


Fig. 3. Angle-resolved tuning properties of the optical extinction spectrum of the MPC device in air for (a) TM and (b) TE polarizations.

Fig. 2 shows the scheme for the optical spectroscopic measurements. Fig. 2(a) shows the geometry for the measurements on the angle-resolved optical extinction spectra, where the white-light beam is incident at an angle of θ_i . Assuming that transmission spectrum through the ITO glass is $I_0(\lambda, \theta_i)$ and that through the MPCs is $I_S(\lambda, \theta_i)$, where λ is the wavelength of light, the optical extinction spectrum is calculated by $-\log_{10}[I_S(\lambda, \theta_i)/I_0(\lambda, \theta_i)]$. Fig. 2(b) shows the geometry of the setup for the detection of BSA/phosphate solution, where the sensor consists of a chamber closed by the MPCs on a piece of ITO glass, a piece of cover glass, and a silicone rubber for sealing and separation between the two pieces of glass. The light beam propagates at θ_i through the window of the sensor chamber. The measurement procedures and definition of the sensor signal are depicted in Section 3.

Fig. 3(a) and (b) shows the optical extinction spectra of the MPCs for TM and TE polarizations, respectively, which have been measured in air with the incident angle increased from 0° to 52° in steps of 4° . Fano-like coupling between plasmon and waveguide resonance modes is observed only for TM polarization, as can be identified by the extinction dips in Fig. 3(a). Mechanisms for the angle-resolved tuning properties of waveguided MPCs in air have been described in detail in Ref. [13] and will not be addressed here.

3. Detection of bovine serum albumin (BSA) in phosphate with different concentrations

BSA is an important protein for biosensor experiments, which is generally used as the blocking agent for specific bioreactions.

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