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Defect-assisted plasmonic crystal sensor

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Abstract

We demonstrate enhanced sensitivity of a nanostructured plasmonic sensor that utilizes resonance in intentional structural defects within a plasmonic crystal. The measured sensitivity of the fabricated nanosensor is ~500 nm/RIU showing improvement over traditional nanohole array sensors. Furthermore, the defects provide an additional design parameter to increase sensitivity by engineering plasmon lifetime.

Optical sensors provide many benefits over traditional sensing schemes as they are highly sensitive, amenable to multiplexing, and immune to electromagnetic interference. Recent research has focused on a class of ultrasensitive optical sensors that utilizes the extreme excitation sensitivity of surface plasmons (SPs). Plasmonic sensing encompasses many types of sensors, each exploiting different characteristics of the SP waves. For example, surface plasmon resonance (SPR) [1, 2], extraordinary optical transmission (EOT) [3–6], and localized surface plasmon resonance (LSPR) [7, 8] of metal nanoparticles have been used for optical sensing applications.

Planar SPR sensors, such as Biacore™, use Kretschmann configuration to launch SPs along a metal-dielectric interface using a coupling prism. This setup employs angular interrogation to monitor the phase matching condition between incident light and the excited SP waves. Nanoparticle-based sensors monitor changes in the extinction spectrum of metal nanoparticles in sensing media. These sensors have demonstrated viability in practical sensing applications including: protein-protein interactions [9], tumor markers [10], bacteria detection [11], and antibody-antigen interactions [7, 12]. Despite the many advantages of these plasmonic sensors, each has inherent limitations. Planar SPR sensors use complicated coupling setups which make the overall system bulky and susceptible to ambient vibrational noise. Nanoparticle-based sensors are vulnerable to inconsistent size, shape, and nanoparticle composition.

EOT-based nanostructures, such as nanohole arrays (NHAs), have been widely used in this field as well. Unlike planar or particle-based sensors, the spectral response of NHA sensors can be precisely controlled and is highly sensitive to external perturbation. EOT-based

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sensors have been used for biosensing [3, 4, 6, 13], gas sensing [14], and refractive index sensing [15] by monitoring changes in resonant peaks within the transmission spectrum. The sensitivity of NHA sensors is governed by many parameters, including field overlap and

their dispersion relation, $\omega = \beta c \sqrt{(\epsilon_m + \epsilon_d) / \epsilon_m \epsilon_d}$ where β is the propagation constant of the SP wave, c is the speed of light, and ϵ_m and ϵ_d are the permittivity values of the metal and dielectric medium, respectively [16]. Changes in the refractive index of the dielectric medium alter β , resulting in a shift in the plasmonic resonance wavelength.

In this paper, we demonstrate for the first time, a new class of plasmonic sensor that uses intentional defects within a plasmonic crystal (PC). The sensor reported here utilizes in-plane plasmonic Fabry-Perot (FP) modes within the defects for refractive index sensing. The plasmonic FP modes can significantly enhance light-matter interactions by creating high quality factor resonance, thus increasing the lifetime of the plasmon wave. The increased interaction directly affects the overall sensitivity of the sensor. Plasmon lifetime engineering can be achieved through optimizing the defect's geometry and physical dimensions. This also provides an additional degree of freedom for sensor design.

The defect modes of a photonic crystal (PhC) have been developed and extensively studied for a number of applications including sub-diffraction lasers [17], control of spontaneous emission [18], optical filters [19], and sensors [20, 21]. The fabricated sensor is a plasmonic analogue of the optical cavity using a defect-based PhC. As shown in Fig. 1, this structure is composed of a thin metal slab for guiding an SP mode bound to a metal-dielectric interface in the vertical direction and the two-dimensional (2D) PC for excitation of the SP mode in the horizontal direction. The defect creates a cavity, thereby forming in-plane plasmonic FP modes. Unlike the optical mode in a PhC, the SP mode in the PC has a field maximum at the metal-dielectric interface, providing a large overlap between the surface-bound field and analyte. This unique property provides high refractive index sensitivity in both bulk and thin film sensing environments.

Excitation of the surface wave is achieved by providing additional momentum, K_G , to incident light according to $K_G = m \cdot (2\pi/\Lambda_1) \hat{x} + n \cdot (2\pi/\Lambda_2) \hat{y}$ where Λ_1 and Λ_2 are the lattice constants and m and n are integers [22]. Fig. 1(c) depicts the formation of the plasmonic FP modes within defects of the crystal. When the period Λ is matched to the effective wavelength of the SP wave, partially reflected waves arriving at the defect are in-phase, creating in-plane FP resonance. The field buildup and the lifetime of the resonant modes are proportional to the reflectance from the nanoholes. By controlling the physical dimensions and number of nanoholes, the plasmon lifetime of the defect mode can be engineered.

For fabrication of the crystal, nanoscale holes were patterned on a 100nm-thick Au film on a glass substrate using focused-ion-beam milling. The Au film was deposited on glass using a 5nm-thick Ti adhesion layer through electron beam evaporation. Patterning was performed using FEI's Nova 600 at 30kV resulting in a square NHA with a period of 500nm ($\Lambda_1 = \Lambda_2$), defect size of approximately 1.3 μm by 1.3 μm , and overall pattern size of 30 μm by 30 μm . An identical NHA, but without defects, was fabricated on the same substrate as a control

sample. A close-up SEM image of the defect within the PC is displayed in Fig. 1(b). Each pattern was slightly over-milled to ensure all metal within the nanohole was removed down to the substrate. The over-milling process produces a slight undercut in the sidewall profile of the patterned structure and has been confirmed by performing a cross-sectional cut on a nanohole array sample. A polyethylene ring was then mounted on the Au film to serve as a fluidic chamber and confine the analyte to the sensor's surface.

To understand how the presence of structural defects affects the overall spectral response, we performed 2D electromagnetic simulations of two types of PCs: 1) NHAs and 2) NHAs with defects on glass. Fig. 2 presents calculated transmission spectra of the nanostructures using a commercial finite-element-method solver COMSOL MULTIPHYSICS™. For simulation, the period of the NHAs and the size of the defects were matched to the values of the fabricated sensor and a modified Lorentz-Drude model [23] was used to account for dispersion of the metal film. Periodic boundary conditions were used to accurately model repeated nanostructures representing the array. Perfectly matched layers were included to avoid undesired numerical reflections from the boundary of the computational window. The values of the cover layer, ϵ_d , and substrate, ϵ_s , were 1.769 and 2.250, respectively. A slight undercut was included in structure ii) to represent the fabricated structure. The transmission spectrum of NHAs shows a resonance peak from EOT. Defects in the NHAs introduce additional resonance peaks in the output spectra. This is due to the excitation of the in-plane FP modes in the defects. The inset of Fig. 2 illustrates the calculated magnetic field distribution in the defect, showing the formation of an FP mode.

The fabricated plasmonic sensor was illuminated through a high numerical aperture (NA) microscope objective (Mitutoyo 20 $\times$, NA .42) by a quartz-tungsten-halogen lamp. The transmitted signal was collected by a fiber-coupled spectrometer. An optical fiber was aligned under the microscope objective at the optical axis of the measurement system. The sensor was then mounted on a translation stage and aligned under the microscope.

The following procedures were carried out to measure the refractive index sensitivity of the sensor. Deionized (DI) water was introduced into the fluidic chamber and the resulting spectral signature was recorded as the baseline measurement and notated as $n = 0$. Subsequent measurements were performed by introducing an analyte of higher glucose concentration to the sensor. Sensitivity characterization of the fabricated sensor can be performed by introducing an analyte of known refractive index and monitoring changes in the transmission spectrum. To achieve this, we varied glucose concentrations in DI water according to referenced literature [24, 25].

Fig. 3 shows the measured changes in the resonant peak at $\lambda_r = 744\text{nm}$, plotted versus time, as each analyte was introduced. Each introduction of analyte provided changes in refractive index of 0.005 refractive index units (RIU). After the final glucose measurement, the sensor was removed and thoroughly rinsed with DI water to reset the surface. The sensor was then remounted and the spectral response was measured. This result is represented by the 'Reset' values seen in Fig. 3. As the refractive index is changed through the introduction of higher glucose concentrations a noticeable shift in resonant peak wavelength is observed. The inset of Fig. 3 is the measured transmission spectrum of the fabricated sample with DI water. This

same procedure was repeated on the control sample. To calibrate the measured spectral responses of the sensors, the overall transfer function of the measurement setup was measured and de-embedded from each measured transmission spectrum. Simple linear regression estimates the sensitivity to be ~500 nm/RIU and ~363 nm/RIU, in the visible spectrum, for the defect-based sensor and the control, respectively. Current literature indicates traditional NHA sensors operate with a sensitivity of 300–400 nm/RIU in the visible spectrum [26, 27]. Enhanced light-analyte interactions from the FP modes within the defect show improved sensitivity over typical EOT sensors.

In conclusion, we experimentally demonstrated enhanced sensitivity of a new class of plasmonic nanosensor characterized by intentional defects within a PC. Sensitivity of the nanosensor was estimated through simple linear regression to be ~500 nm/RIU. The improved sensitivity is due to enhanced light-analyte interactions from the defect mode.

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Fig. 1.

- (a) Schematic of the fabricated sensor representing resonance within the structural defect.
 (b) Close-up scanning electron microscope (SEM) image of the defect sensor after patterning.
 (c) 2D cross-sectional view of the formation of plasmonic FP modes in the defect.

Fig. 2. Comparison of the calculated transmission spectra of the nanostructures: i) NHA in a homogenous medium ϵ_d (dashed line), and ii) fabricated NHA with defect of length L_d , cover layer ϵ_d , and substrate ϵ_s (solid line). A slight undercut, typical of lithographic processes, was included in ii) to represent the sidewall profile of the fabricated structure. Inset- Calculated magnetic field distribution of the undercut structure at the second resonant peak.

Fig. 3.
Changes (λ_r) in resonance peak due to refractive index perturbation plotted versus time.
Inset- Transmission spectrum of the sample at $\Delta n = 0$.