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Enhanced UV/blue fluorescent sensing using metal-dielectric-metal aperture nanoantenna arrays

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ABSTRACT

Subwavelength aperture antenna arrays are designed and fabricated for potential applications in fluorescence sensing in the near UV/blue range. They are designed using finite-difference time-domain (FDTD) simulation, fabricated using focused ion beam etching and characterised using angular Fourier spectroscopy. The aperture arrays are formed in the top layer of an aluminum-silica-aluminum trilayer and produce a maximum simulated field intensity enhancement of 5.8 times at 406 nm and highly directive emission with a beamwidth of 8.3 deg. The normal incidence reflection response has been measured and shows reasonable agreement with modelled results. In addition, to investigate higher field intensity enhancements, bowtie aperture arrays are simulated and the influence of parameters such as dielectric gap, position of dipole source, and aperture shape and size are discussed and show enhancements up to 67 times are possible.

1. Introduction

The concept of surface plasmons or plasmonics has seen widespread use in implementing optical devices and photonic circuits [1–3] as well as in chemical sensing technologies [4–8]. Recent advances in metallic nanoparticle synthesis and nanofabrication methods have led to a variety of new nanostructures composed of nanoparticles, nanoholes, and other structures with precisely controlled shapes, sizes, and/or spacers, with novel physical and chemical properties [9–12]. Such exquisite fabrication and synthesis control in combination with advances in theory and the emergence of quantitative electromagnetic simulation tools has provided a better understanding of the optical properties of single and coupled metallic nanostructures of various sizes and shapes [10,11]. Various approaches for modifying metallic surfaces and structures make it possible to effect the selective binding and detection of specific targets for chemical and biological sensing. Recently metallic nanoparticles and nanoantenna arrays have been used because of their ability to greatly enhance local electromagnetic fields and confinement [13–17]. Specifically it was demonstrated that metal-dielectric-metal (MDM) structures of nanoantennas are a very effective alternative to increase the performance of nanoantennas for chemical and biological [15,16] and humidity [17] sensing applications. Nanoantennas are also very suitable for biological applications because they are able to track emission from fluorescent makers in cells with sub-diffraction limited resolution, as well as the destruction of cancer cells using the heating of resonant nanoparticles [18,19].

In 1998, Ebbesen et al. demonstrated the extraordinary optical transmission through the subwavelength hole arrays perforated in a gold or silver film which is generally attributed to the resonant excitation of surface plasmons [20]. Subsequent work [21] confirmed that surface plasmons formed on both sides of the metallic surface resonantly couple through the subwavelength hole arrays, which enhances the light transmission for specific wavelengths depending on the lattice constant of the hole arrays and the dielectric constants of the metallic and the surrounding media. This structure has shown potential for surface plasmon resonance sensing, since they can couple incident light directly into surface plasmons. The feasibility of using subwavelength hole arrays for biosensing where a spectral shift is observed after the immobilization of molecules on metallic film using a broadband source and a spectrometer has also been shown previously [22–24]. In the context of fluorescent biosensing applications, plasmonic nanoantennas can create highly enhanced local fields when pumped resonantly, and through Purcell enhancement can significantly enhance fluorescent emission, both of which can improve fluorescence based sensors [25–27]. Recently we have shown fluorescent emission enhancement using aluminium nanoantenna arrays in the near-UV [10]. Scanning confocal photoluminescence measurements of the arrays showed up to 1.9 times more enhancement of the fluorescent signal

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compared to a reference on glass. The simulations and measurements showed strong dependence of the emission on angle, with maximum emission at high angles with respect to the normal, so that a high numerical aperture (NA) is necessary to collect the emission. In this work, aperture arrays in a MDM structure are considered as another choice which give high field enhancement and strong normal emission enabling straightforward collection of fluorescence. This is an extension of a single metal layer aperture array, with the addition of a lower metal layer which prevents downward emission of fluorescence, further enhancing upward emission.

The structure consists of a subwavelength aperture array which is Focused Ion Beam (FIB) etched into the upper aluminium layer of the MDM structure. We fabricated a 14 by 14 element array and performed angle resolved reflectance measurements. We simulated the enhancement with the FDTD method using electric dipoles placed inside the subwavelength aperture arrays. The simulated enhancement definition here is a very simple one which is the ratio of total emitted power from a dipole with and without the antenna which does not account for the very strong focusing effects that occur due the periodic nature of the array [10]. In addition, we study the influence of antenna parameters such as silica gap thickness, aperture size and shape, and also position of the electric dipole sources on the fluorescent enhancement. The paper is composed of three sections. The first section introduces the design and fabrication of subwavelength aperture antennas. The second section presents the results and discussion. In the final section, we present the conclusions.

2. Design and fabrication

2.1. Simulated reflection spectra

Our study uses a silicon substrate covered by a 100 nm thick aluminium (Al) layer, which is thick enough to prevent optical transmission into the silicon, further capped by a continuous silica (SiO_2) film of thickness *d*. The nanoantenna structure is formed by a square lattice (lattice constant $L = 350$ nm) of subwavelength square apertures (width, $w = 175$ nm) perforated through an Al layer (thickness *t*) on top of the SiO_2 film, as illustrated in Fig. 1(a). The simulations were performed using a commercial 3D full-wave electromagnetic wave solver Lumerical FDTD [28]. To calculate the reflectance spectra of the nanoantenna structure, perfectly matched absorbing boundary conditions are used at the top and the bottom boundaries [29]. In the lateral direction, periodic boundary conditions are applied. Incident light to the unit cell was a plane wave. The reflectance was collected with a power monitor placed behind the radiation source. Fig. 1(b) shows the reflectance spectra of the normally incident wave ($\theta_i = 0°$) for several SiO_2 inter-layer thicknesses (*d*) and upper layer thicknesses (*t*). As shown in Fig. 1(b), the reflectance spectrum shows two dips for each structure. The dip at longer wavelengths, above 480 nm with quality factor (*Q*-factor) of ~8.5 corresponds to the surface plasmon resonance supported by the periodic array, which can be understood as a plasmonic standing wave on the surface of the antenna array. The more pronounced dip at shorter wavelengths around 406 nm with *Q*-factor of ~10.5 corresponds to a plasmonic standing wave localized inside the apertures, which is not strongly affected by changes in the upper or SiO_2 layers [13].

These resonant characteristics can be seen in Fig. 1(c) and (d) which show the electric field distributions in side and top view at the two resonances. Fig. 1(c) shows the distributed nature of the longer wavelength resonance and Fig. 1(d) shows the strong localisation for the shorter wavelength resonance. Both of these may be useful for different types of sensing applications.

To understand the angular dependence on the optical properties, we have next calculated the reflectance of the structure versus the incident angle. This also relates to the type of optical characterization that will be carried out later in the paper. The oblique incident angle θ_i is defined by the angle between wavevector *k* and the *z* axis. An inter-layer SiO_2 of 40 nm and upper Al thicknesses of 140 nm are chosen for the simulation. The structure is not sensitive to polarization due to its symmetry in *x* and *y* directions, so that only dependence of the reflectance spectrum on the incident angle θ_i for electric polarization along *x* direction is shown in Fig. 2. The blue shaded region around 400 nm in these figures represent a decrease in reflectance and hence indicates the presence of a strong optical mode. This resonance remains for a wide range of angles, which could be useful in a number of sensing applications in fluorescence based biosensing.

2.2. Sample preparation and characterization

The Al-SiO_2-Al structure was fabricated on a 24 mm × 24 mm x 1 mm silicon (100) substrate. The lower Al layer was deposited by sputter coating (Leybold L560) at a fixed deposition rate of 2 Å/s. An ultra-thin chromium (Cr) layer (~5 nm) was used as an intermediate layer to improve Al adhesion [15]. Then, the SiO_2 buffer-layer was deposited using Plasma Enhanced Chemical Vapour Deposition (PECVD) and finally, the upper Al layer was deposited by sputter coating (Leybold L560) at a fixed deposition rate of 2 Å/s. The second step was patterning of antenna arrays using the focused-ion-beam (FIB). The FIB current and dwell time are the main parameters that effect the etching width and depth for a fabricated antenna arrays. Fig. 3 (a) shows a cross-section of this tri-layer structure viewed after FIB cross-sectioning. A thin layer of platinum (Pt) has been deposited over the upper Al layer to aid the imaging and cross-sectioning process. A dark SiO_2 layer can be seen between the two Al layers. The thicknesses of the layers were measured as: upper Al ~140 nm, SiO_2 ~ 40 nm, and lower Al ~100 nm. The thickness of upper Al layer is larger than expected thickness of 100 nm, but this does not greatly influence the optical properties of the antenna arrays. Fig. 3(b) shows the surface of the ~5 × 5 μm^2 antenna arrays, the average period is 350 nm and square size is 175 nm × 175 nm.

The reflectance (*R*) measurements of the nanoantenna arrays were carried out by Fourier spectroscopy. The wide angular reflected light was collected from a 5 × 5 μm^2 focusing area using an objective with a high numerical aperture (Zeiss EPIPLAN 100× NA = 0.8). In order to measure reflectance from a device we first measure a background reference reflectance from an Al mirror. The Fourier microscope uses a spectrometer (Ocean Optics USB 2000+) for the far UV and visible parts of the spectrum and the results shown here have some noise in the UV range due to the detector and the limited transmission of the optics in this range. Fig. 4(a) shows the reflectance colour map of the fabricated sample depicted in Fig. 3 for *y* electric field polarization. As can be seen in Fig. 4(a), the resonance at around 400 nm can be observed along with a broader -shallower resonance around 530 nm. The comparison between the measured and simulated results at normal incidence is shown in Fig. 4(b) and reasonable level of agreement is obtained, considering the fabrication imperfections which will be present in the measured structure. Also the finite size of the measured sample will cause some discrepancies.

3. Results and discussion

In order to model the fluorescence emission enhancement produced by the MDM antenna arrays shown in Fig. 1(a), we simulated the interaction of an electric dipole source with the MDM antenna arrays. In order to simplify the analysis an E_y electric dipole is positioned at the middle of the upper Al layer. We plotted the power enhancement with the electric dipole source 15 nm away from the surface aperture as depicted in inset of Fig. 5(a) showing a single unit cell. The enhancement is the total power through a plane 50 nm above the upper surface normalized to the power that would have been emitted through the plane by the source in free space. The infinite antenna arrays are simulated by using periodic boundary conditions in the lateral directions.