Cathodoluminescence Imaging of Plasmonic Modes of Au Nanostructures

Anil Kumar[1], James Mabon[2], Edmond Chow[3], and Nicholas Fang[4]*
[1]Department of Electrical and Computer Engineering; [2]Frederick Seitz Materials Research Laboratory; [3]Micro and Nanotechnology Laboratory; [4]Department of Mechanical Science and Engineering; University of Illinois at Urbana-Champaign, Urbana, IL 61801 USA

I. INTRODUCTION

Recent advances in resonant sub-wavelength optical antennas [1-4], as an optical counterpart of microwave antennas, has offered researchers a continuum of electromagnetic spectrum to design, analyze and predict new phenomena that were previously unknown [1, 4-5]. Their applications in areas with pressing needs, e.g., in sensing [4], imaging [6], energy harvesting, and disease cure and prevention have brought revolutionary improvements. However, understanding of the physics behind the optical interaction with metal nanostructures is still in the early stage. By patterning metal films in unique shapes and sizes, highly localized optical modes can be engineered. Exciting and observing these modes require state-of-the-art techniques, e.g., near-field scanning optical microscopy (NSOM), electron energy loss spectroscopy (EELS), and cathodoluminescence (CL) spectroscopy. Among these, CL is advantageous because of its high resolution, ease-of-excitation, and less stringent demands for sample preparation. CL has been previously used to study the optical modes of optical antennas [6-9], however, the luminescence peaks from substrates can overshadow any possible antenna modes. Si has been used to overcome this issue; however, its higher refractive index shifts the resonant wavelength [9] beyond the range of detectors used for current CL systems. Here we take a new approach to design, excite and observe optical modes of triangular Au nanoantennas using CL spectroscopy.

II. EXPERIMENT

To obtain a low index substrate with minimal substrate luminescence, we fabricated gold nanostructures on 100 nm thick PECVD-deposited SiO_2 film using electron-beam lithography. The SiO_2 outside the nanostructures was removed using Freon-based RIE. A 50:1 BOE solution was used for further undercutting of the oxide underneath the nanostructures to provide an effective index close to vacuum. Figure 1 shows an SEM image of a 50 nm thick triangle dimer showing the undercut region. The left triangle shows a reduced SiO_2 substrate region that has shrunk due to sidewall etching. The Au-SiO_2 interface is observed to be further reduced, as seen from the left exposed oxide surface. This allows us to carefully control the SiO_2 region with the nanostructure experiencing a low effective index close to vacuum.

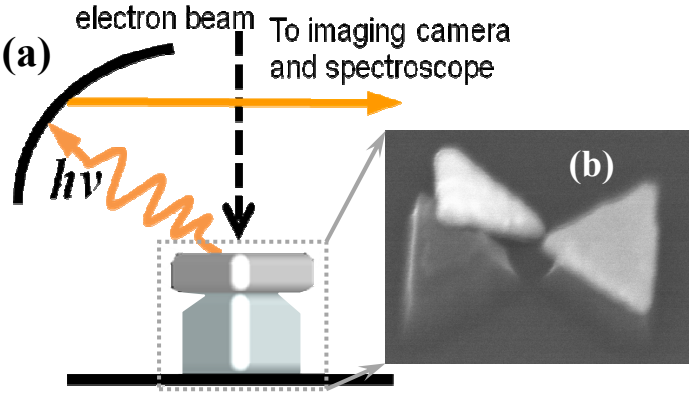

Fig. 1. (a) Schematic of the cathodoluminescence setup. A parabolic mirror collects the light from nanostructures and directs it to a photomultiplier tube (PMT). (b) SEM image of an Au triangle-dimer on 100 nm thick SiO_2 deposited on Si. After e-beam fabrication of 50 nm thick Au structures, SiO_2 outside the nanostructures was removed using Freon RIE and further undercutting was achieved using a 50:1 BOE solution.

Measurements were carried out on a CL setup (MonoCL from Gatan) attached to an SEM (JEOL JSM-7000F) and the spectra were taken using a photomultiplier tube (PMT) with a range of 250 nm – 900 nm. A schematic of the setup is shown in Figure 1.

III. RESULTS AND DISCUSSION

To predict the effect of substrate on resonant wavelength, we carried out FDTD simulations for various substrates. Resonant peaks as a function of substrate index for triangles with tip-to-base height of 120 nm and 150 nm are shown in Figure 2. A narrow window of substrate index in the visible range is observed. The square in Figure 2 predicts a peak at 680 nm for a 150 nm triangle sitting on a 100 nm thick triangle of same size. This peak lies at the average of vacuum-SiO_2 indices; however, the actual peak should be red-shifted due to smaller SiO_2 triangle.

Observing the plasmonic modes during CL spectroscopy has a practical problem of separating the nanostructure modes from substrate luminescence peaks. Most commonly used materials in device processing show luminescence under electron

excitation making the nature and purity of the substrate critical. As an example, float glass shows nine peaks in the visible region [10]. To avoid the problem of substrate peaks, high purity materials like Si, InP, or other single crystals that do not show any peaks in visible region can be used; however, their high index shifts the dipole peaks beyond the detection range of our setup. SiO_2 also shows two peaks in this region [11]; however, the high energy peak lies outside the dipole peaks of our designs, while the low energy peak is weak and its effect can be reduced by normalizing the spectra with substrate.

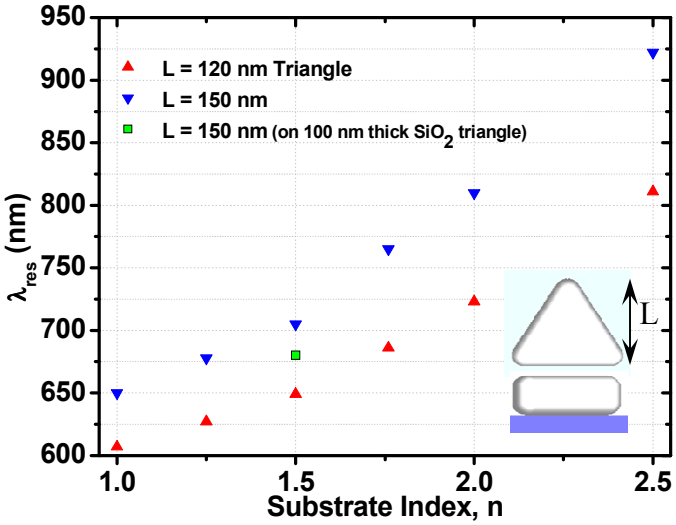

Fig. 2. Simulated resonant peak positions for the in-plane-dipole mode as a function of substrate index for 50 nm thick Au triangles (L=120 nm and 150 nm). A 15 nm curvature was used for the triangle edges and tips. The square shows a resonance peak for triangle (L=150 nm) sitting on another SiO_2 triangle of same size but 100 nm thickness and Si substrate.

Figure 3 shows a spectrum of an Au triangle (red) with L = 150 nm. A broad peak at 575 nm is observed after grating correction and normalization with a SiO_2 triangle on Si. For comparison, spectrum taken on Au film (blue) shows a surface plasmon peak near 525 nm as predicted earlier [12].

Panchromatic CL imaging allows us to observe the possible spatial modes of the triangles. Figure 4 shows a preferential excitation of tips observed during raster scanning of the electron beam. This spatial mode has earlier been associated with an out-of-plane dipole mode for Ag triangles [9]. From panchromatic images taken for various triangle dimers, CL resolution was observed to be approx. 22 nm (FWHM), which is comparable to previously reported value [13].

III. CONCLUSION AND OUTLOOK

Au triangular nanostructures were fabricated on a low index substrate by using undercutting of SiO_2 deposited on Si. CL imaging showed high contrast tip modes excited in panchromatic imaging and a peak position at 575 nm was observed. FDTD simulations were performed to predict the in-plane dipole peaks. Further undercutting of oxide gives a tilt to the nanostructures, allowing electron-beam to be parallel to the surface; this approach will be used to further explore the in-plane modes of the nanostructures.

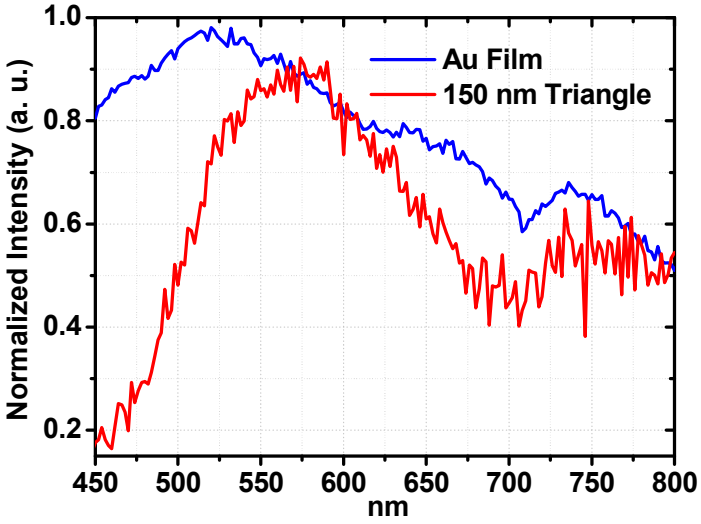

Fig. 3. CL spectrum for Au triangle after grating correction and normalization with SiO_2/Si background. Spectrum for Au film with surface plasmon peak at 525 nm [12] is shown for comparison.

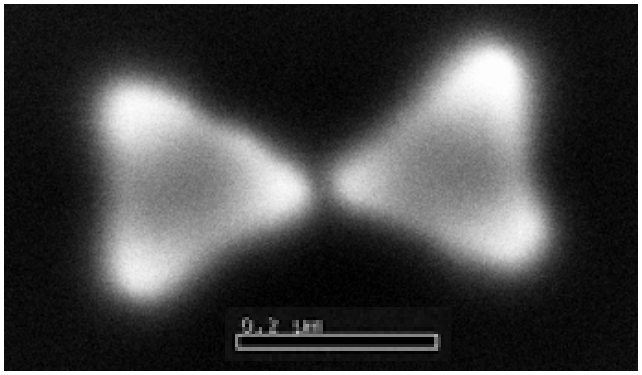

Fig. 4. Panchromatic CL image of Au triangle dimer (30 kV, 12 nA). The scale bar is 200 nm.

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