Elucidating effects of nanoscale structural variations on local plasmonic modes via photon localization microscopy

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Plasmonic devices are primarily based on metallic (nano)structures with nanoscale feature sizes, and many are designed to enhance and concentrate fields at ~ 10 nm length scales. This leads to an enhanced sensitivity of local plasmonic modes to locally-varying material properties, device substructure, and environment, which must be taken into account for each specific application. Advancement of plasmonics technologies depends on reliable fabrication of these structures, often repeatably over large areas. However, the resolution and reproducibility limits of current nanofabrication techniques, such as electron-beam (e-beam) lithography, focused ion beam (FIB) milling, and e-beam- and FIB-induced deposition, tend also to be ~ 10 nm, often struggling to break the so-called “single-digit-nano” barrier. In some cases, bottom-up chemically synthesized nanoplasmonic structures can have less structural variation, but these require specific arrangements and orientations for most devices, and specific binding events and placement accuracies are again limited to ~10 nm or greater. Therefore, it is expected that the properties of plasmon modes with localized fields will be influenced by these unavoidable structural variations.

In this work, we use the recently-demonstrated non-perturbative all-optical probing technique of two-photon photoluminescence-based photon localization microscopy[1] as well as electromagnetic simulations to reveal how small structural variations often significantly impact plasmonic properties, particularly for devices with ~ zeptoliter mode volumes. The photon-limited localization accuracy of nanoscale mode positions is determined for many of the measured devices to be within a 95% confidence interval of +/- 2.5 nm. Specifically, we find that for our test devices, local plasmonic behavior is primarily influenced by two classes of fabrication-related variations: 1) random, “incoherent” localized defects (Fig. 2); and 2) small changes in structure size (Fig. 3), which are directly related to the cavity length of the plasmon resonator. For this second case, we show that relatively modest changes in device length of about 10% can nearly double the spatial separation between modes.

To investigate the effects of fabrication-related imperfections, we chose as our test samples asymmetric bowtie nano-colorsorter (ABnC) optical antennae (Fig. 1 a-b). Here we correlated relative spatial movements of the two modes with the structural variations we observed via SEM characterization.

[1] A. McLeod, *et al.*, Phys. Rev. Lett. in press (2011)

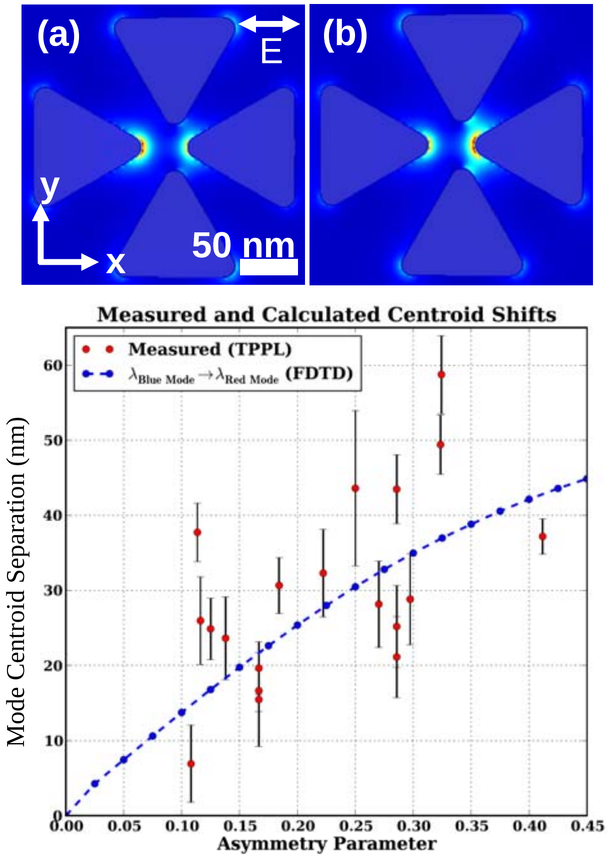

Fig. 1. Calculated $|E|^2$ near-field distributions for the "blue" mode (a) and "red" mode (b) of a two-color ABnC. (c) TPPL image mode centroid separations plotted as a function of normalized offset of the vertical bowtie component in the +x direction. Experimentally measured values are marked by red circles and FDTD-calculated values are shown by the blue line and circles. Error bars correspond to the one-sigma accuracy of the experimentally-determined shifts along the x-axis.

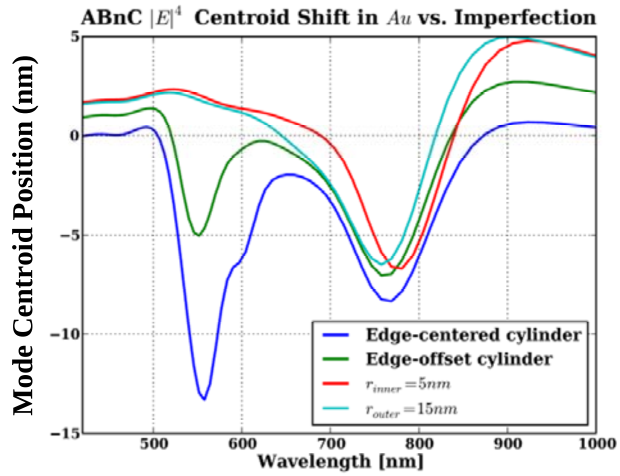

Fig. 2. Calculated mode centroid positions are plotted as a function of wavelength, where it is seen that maximum position separations are much smaller than for a device without the defect (compare fig. 3).

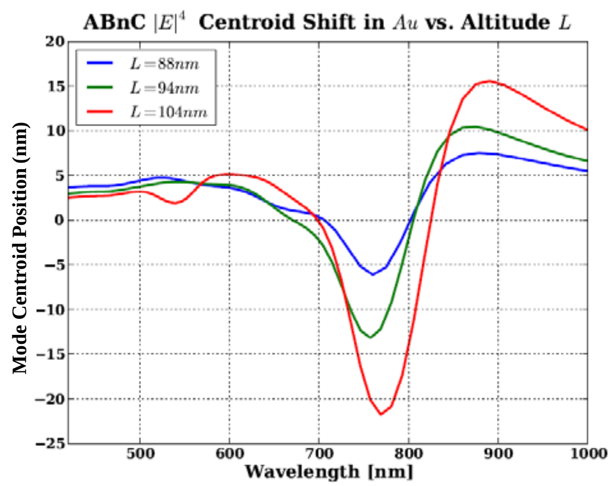

Fig. 3. Small variations in triangle size greatly effect E^4 centroid separations, leading to deviations from the theory line in fig. 1c. Centroid separations are determined by subtracting the minimum centroid position (near 780 nm) from the maximum position (near 850 nm). Here, triangle altitudes, L, for the horizontal-oriented bowtie are varied (triangles remain equilateral) while holding the gap size constant.