Plasmonics At The Single-Nanometer Scale

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Nanometer-sized gaps or contacts between conductors are of great interest, as they respectively exhibit the phenomena of quantum tunneling and quantized conductance at low frequencies. At optical frequencies, however, the transport properties of these nanosized features are challenging to probe. As a result, theoretical simulations often use parameters at DC to predict the behavior of systems operating at significantly higher frequencies, resulting in inaccurate predictions. In this work, we demonstrate progress towards probing the transport properties in several-atom-wide gaps and contacts placed in the center of collectively-oscillating electrons, i.e. plasmon resonances, at optical frequencies. At these frequencies, applications of plasmonics can be found for optical interconnects and surface-enhanced Raman spectroscopy.[1]

Using a recently developed process[2] and alternatively by electron-beam sculpting in a transmission electron microscope (TEM), we fabricated a series of Au bowtie antennas with gaps varying by an order of magnitude (0.5 nm to 14 nm) and nanoprisms connected by conductive contacts with widths of ~1 to 13 nm. These nanostructures were fabricated on a single free-standing silicon nitride membrane (30 nm thick), making it suitable for measurements in a TEM equipped with monochromated electron energy-loss spectroscopy (EELS).

This process enabled the in-situ probing of plasmon resonances of the structures by electrical excitation while obtaining their high-resolution TEM images. The lithographically-defined structures were nominally identical except for the systematically-varied gap/contact dimensions between the nanoprisms. In EELS, energetic electrons drive the collective oscillation of electrons in the metal nanostructures, allowing for a high-resolution imaging of the resulting electric fields, which we call plasmon maps. Figure 1 shows examples of TEM images of coupled nanoprisms with a gap of 3 nm and a contact of 4 nm with their corresponding “ground-state” plasmon maps.

We will discuss the systematic trends of the evolution of plasmon-resonance modes as the nanoprisms come into contact. Our results further demonstrate, rather surprisingly, that the plasmonic behavior even at the last nanometer can be described adequately by classical electrodynamics.

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[1] Huigao Duan, Hailong Hu, Karthik Kumar, Zexiang Shen, Joel K.W. Yang, ACS Nano, 5 (9), pp 7593–7600 (2011)

[2] Ai Leen Koh, Antonio I. Fernndez-Domnguez, David W. McComb, Stefan A. Maier, Joel K. W. Yang, Nano Letters, 11 (3), pp 1323–1330 (2011)

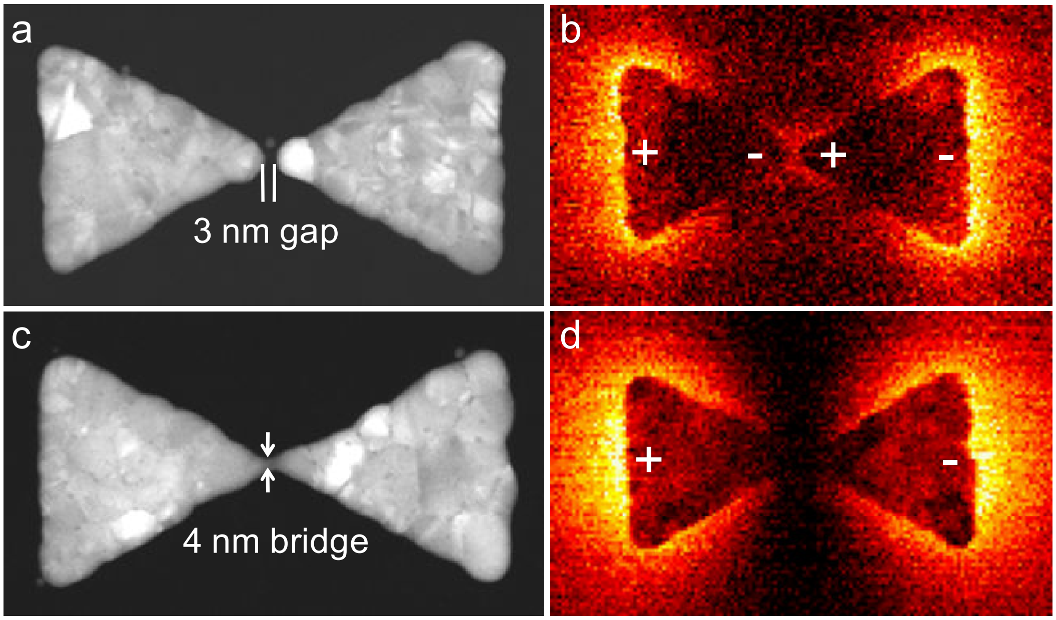

Figure 1: (a) Dark field STEM image and (b) EELS map at 1.37 eV of a bowtie nanoantenna with a gap of 3 nm; (c) STEM image and (d) EELS map at 0.53 eV of a bowtie nanoantenna with a contact of 4 nm width. The EELS maps show that the "ground-state" plasmon resonances supported by these two systems that differ only at the nanometer scale have distinct energies. The plasmon map in (b) shows the presence of electric fields bridging the gap of the accompanying charge distribution of two coupled dipoles, while the fields in (d) correspond to that of a single dipole, due to electron transport across the 4-nm contact.