Critical influence of gap morphology in the optical response of ultranarrow gap-plasmonic nanoantennas

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Plasmonic nanoantennas are resonant structures that support localized plasmonic modes, typically in the visible and near-infrared range of the electromagnetic spectrum, due to free-electron oscillations[1]. Plasmon resonances can be easily tuned and can concentrate incoming radiation to very small volumes. Notably, when two plasmonic particles are separated by a small gap, Coulomb interaction induces hybridized resonances that localize and enhance the field into the gap region. In this work, we demonstrate that, for very narrow separation distances, the gap morphology strongly affects the optical response. In particular, we consider linear gap antennas comprising two metallic rods, and study the difference in the optical response between spherical and flat gap ends (see sketches in Fig.1). Furthermore, we decrease the gap down to sub-nanometric distances where quantum tunneling plays an important role in the optical response of the system. To study this quantum regime, we use the Quantum Corrected Model [2,3], which enables to include electron tunneling within a classical framework [4] and therefore enables to adress large plasmonic systems.

We first consider a classical treatment of narrow gaps at large enough separation distances where electron tunneling is not present. The spherical-ended antenna (Fig.1a) behaves similarly to a metallic sphere dimer [5]. As the gap narrows, the plasmonic resonances strongly redshift showing a large extinction cross-section and very strong near-field enhancement at the gap. When the gap separation distance is equal to zero, an abrupt change in the modal distribution is clearly observed in the extinction spectrum. Once the two arms are in contact, we observe charge transfer plasmons that blueshift as the overlap increases.

The flat-gap antenna (Fig.1b), the optical response of flat-gap antennas is very different, particularly for very narrow gaps. Several longitudinal antenna modes can be observed in the extinction cross section. The dominant lowest energy mode, which is spectrally broad, also redshifts as the gap distance is reduced but this redshift saturates for very narrow gaps. The spectral position of higher order modes is hardly affected by the gap separation distance. A simple RC model is able to explain the spectral behavior and relates the modes to those of the constituent individual nanorods [5]. After contact, the resulting modes correspond to the resonances of a rod of twice the initial length. In contrast, in the near-field spectrum at the gap two different sets of modes are clearly appreciable: (i) the longitudinal antenna modes and (ii) a set of spectrally narrow modes that strongly redshift with decreasing gap, identified as cavity modes that radiate very weakly [6,7]. We use a simple analytical expression based on the modes of a Fabry-Pérot resonator to interpret the spectral evolution of these plasmon cavity modes as the gap narrows. High order cavity modes exhibit very fast spatial variation of the fields at the gap. For narrow gaps, the longitudinal antenna modes and the cavity modes can be tuned independently by changing the respective geometrical parameter. The field enhancement is maximum when the resonant energy of a cavity mode coincides with that of an antenna longitudinal resonance. To access this ultrananarrow gap regime at visible frequencies is very challenging experimentally since distances below 1 nm along extended areas are necessary. A situation experimentally at hand that would reproduce this behavior for larger separation distances could be obtained with use of phononic resonances in the infrared. We therefore apply the same concepts to phononic antennas made of SiC.

For distances smaller than approximately half a nanometer, electron tunneling must be considered. The main effect observed in this tunneling regime is a very strong quenching of the near-field enhancement at the gap [2,3] for both spherical-gap and flat-gap geometries. The cavity modes appear to be quenched for larger separation distances in flat gaps compared to the case of the spherical-gap antennas. Moreover, the effect of the tunneling on the extinction depends on the morphology of the gap. In the spherical case, the tunneling induces a smooth spectral transition between the modes of the non-touching situation and the charge transfer plasmons found in the particle-overlapping situation. The charge transfer plasmons already appear slightly before contact. This continuous spectral transition is in contrast with the unphysical discontinuity found within a classical treatment of the response. For the flat-gap antenna however the tunneling does not appreciably change the extinction cross-section found classically.

Our work shows that the gap morphology influences dramatically the optical response of plasmonic gap nanoantennas separated by ultrashort distances not only quantitative but also qualitatively, an influence that should be considered in design strategies for sensing or spectroscopy applications. Importantly, when sub-nanometric gaps are studied, electron tunneling needs to be included in the description.

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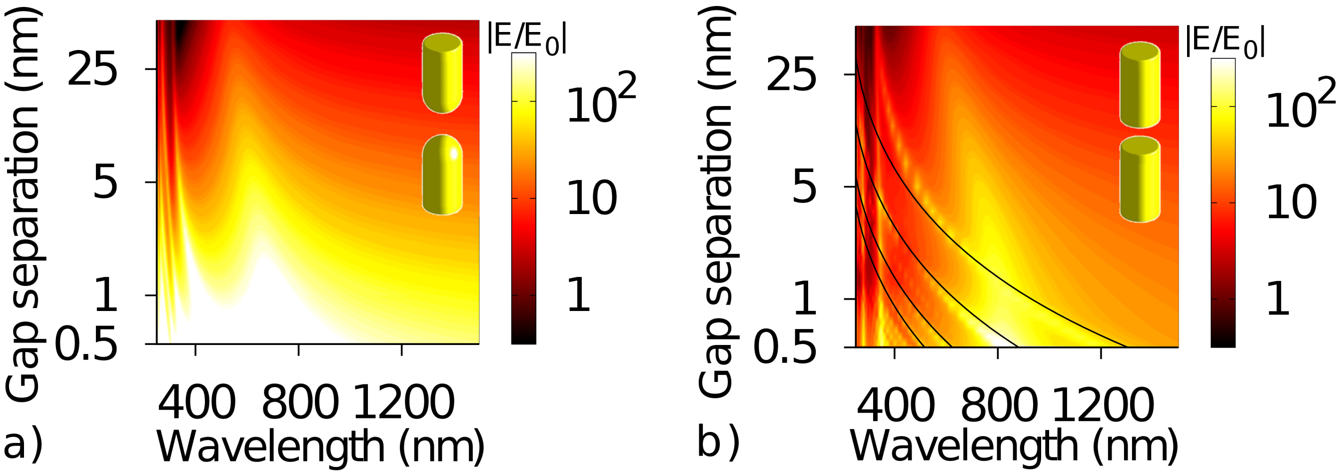

Fig. 1: Near-field enhancement at the spherical-gap (a) and flat-gap (b) antennas as a function of distance and wavelength. For the distances considered in these calculations tunneling is not present. In (b), the resonant wavelength of the cavity modes, as predicted by a simple model, are labeled with black lines.