Plasmonic Breathing and Edge Modes in Aluminum Nanotriangles

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The optical properties of metallic nanoparticles are dominated by the surface plasmons (SPs), which are collective electron oscillations. In a metallic nanoparticle these oscillations are confined by the boundaries of the particle and can be tuned by shaping the geometry, resizing the nanoparticle or changing the dielectric environment. Recently, a detailed analysis of surface plasmons in flat structures [1] proposed to classify surface plasmon modes into two families. The first one corresponds to the so-called edge modes, which are localized at the periphery of two-dimensional nanoobjects. These edge modes are well-known in the literature and have been reported for several geometries. On the other hand, the second group of modes corresponds to the so-called radial breathing modes (RBMs), which are localized inside the two-dimensional nanoobjects. Such modes have been reported experimentally for silver nanodisks [1, 2], silver nanosquares, and silver nanotriangles. Moreover, although a large number of studies have been performed on 2D plasmonic objects, only the plasmonics disks received a comprehensive description [1, 2]. In contrast, the widely studied case of triangular prisms could not be comprehensively described, each study leading only to a partial understanding of the mode structure. In this work we extend the comprehension of edge modes and RBMs in triangular nanocavities.

In order to carry out this study, equilateral aluminum nanotriangles have been characterized by electron energy loss spectroscopy (EELS) coupled with a transmission scanning electron microscope (STEM) which allowed us to obtain spatial and spectral information at the same time. Although the spectral resolution of our EELS spectrometer is 0.4 eV, the systematic use of Richardson−Lucy Deconvolution (with typically 50 iterations) yields a ~ 0.15 eV spectral resolution. An automatic fitting procedure with several Gaussian functions was used to generate intensity maps. The aluminum nanotriangles were fabricated by electron beam lithography and lift-off process on a STEM-EELS compatible substrate (30 nm thick Si_3N_4 membrane). The triangle side length varies from 125 to 700 nm, while the thickness of all structures is 40 nm.

STEM-EELS intensity maps of aluminum nanotriangles (figure 1) present two kinds of modes, the edge modes and the pseudo-RBMs. The symmetry of these modes and their energy dispersion have been theoretically verified by rigorous EELS-GDM simulations. One dimensional and two-dimensional Fabry−Perot analytical models allowed us to label the edge modes (by one quantum number) and RBMs (by a set of two quantum numbers) and to study the phase shift upon reflection. The wavenumbers extracted from the analytical models were used to plot the dispersion relation (E vs k) of edge modes and RBMs (figure 2). The different behavior in the dispersion relation of both modes reveals their distinct nature. It has been proven experimentally (for the first time) that edge modes in Al nanotriangles can be related to Al nanoantenna modes. On the other hand, the RBMs have been related to the dispersion relation of quasistatic stationary short-range surface plasmons (SRSPs) in Al thin film, as in the case of disk RBMs [1, 2].

References:

[1] Schmidt, F.-P. et al. Universal dispersion of surface plasmons in flat nanostructures. *Nat. Commun*. 5, 3604 (2014)
[2] Schmidt, F.-P. et al. Dark plasmonic breathing modes in silver nanodisks. *Nano Lett*. 12, 5780 - 5783 (2012)

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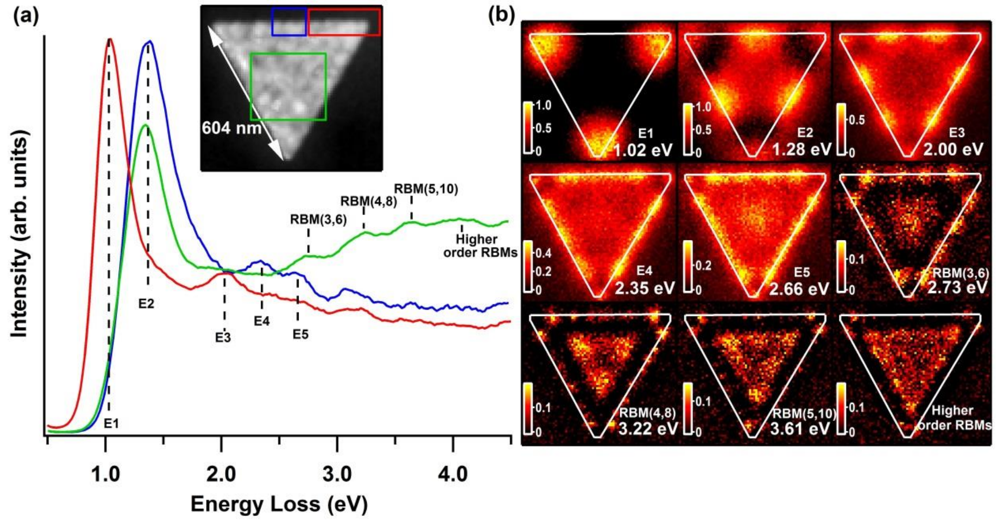

Figure 1. (a) Deconvoluted EEL spectra acquired at different locations (color boxes in the inset) on a single Al nanotriangle with 604 nm side length. The labels E and RBM stand for the edge and pseudoradial breathing modes observed in the nanotriangle. Inset: corresponding HAADF image. (b) EELS fitted intensity maps of the plasmon resonances sustained by the Al nanotriangle at the energy of the resonances evidenced in a.

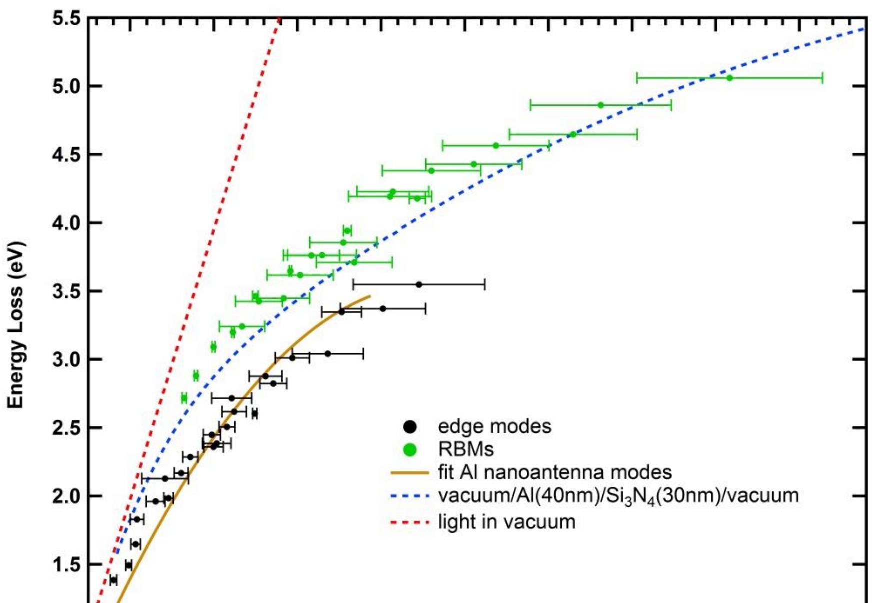

Figure 2. Dispersion relation of the edge modes and pseudo-RBMs of Al nanotriangles. The solid brown curve represents the experimental dispersion relation of Al nanoantenna modes. The dashed blue curve is the analytical dispersion relation of an Al thin film (40 nm thickness) on a Si_3N_4 substrate (30 nm thickness) immersed in vacuum. We observe that Al edge nanotriangles modes and Al nanoantenna modes match very well. An analogous match is observed between RBMs and the analytical dispersion relation of an Al thin film.