Acousto-plasmonic hot spots in metallic nano-objects

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In this work we investigate the acousto-plasmonic dynamics of metallic nano-objects by means of resonant Raman scattering. Acousto-plasmonic interactions still presents several theoretical challenges to correctly interpret the Raman-Brillouin scattering. A variety of theoretical and experimental works have been devoted to the study of shape, size, matrix and ordering effects on the surface plasmons whereas there are only few studies of the dynamical properties of the surface plasmons such as coupling mechanisms to the acoustic vibrations [1], renormalization and damping effects or inelastic light scattering properties.

The issue

We experimentally observe an unexpectedly strong acoustic vibration band in the Raman scattering (Fig. 1b) of silver nanocolumns (NCls) (Fig. 1a), usually not found in isolated nano-objects. The frequency and the polarization of this unexpected Raman band allow us to assign it to breathing-like acoustic vibration modes. To understand this “anomalous” Raman scattering (“?” on Fig. 1b), we address a theoretical and experimental study of the interactions between acoustic vibrations (Fig. 2a) and surface plasmons (Fig. 2b) [2-4]. The modulation of the surface plasmon nearfield (Fig. 2c) allows for the interpretation of experimental Raman-Brillouin spectra in these objects.

Methods

Based on full electromagnetic near-field calculations coupled to the elasticity theory, we introduce a new concept of “acousto-plasmonic hot spots” which arise here because of the indented shape of the nanocolumns. These hot spots combine both highly localized surface plasmons and strong shape deformation by the acoustic vibrations at specific sites of the nano-objects. In order to investigate this new concept, we integrate the Boundary Element Method [5,6] for the electromagnetic calculations (Fig. 2b) and the elasticity theory by the means of the RUS method [7] for the vibrational calculations (Fig. 2a), which allows calculating the modulation of the surface plasmon polarization for these acoustic vibrations (Fig.2c)

Results & Conclusions

We show that the interaction between breathing-like acoustic vibrations and surface plasmons at the “acousto-plasmonic hot spots” is strongly enhanced, turning almost silent vibration modes into efficient Raman scatterers. The indentations of the silver NCls are responsible for the strong localization of the surface plasmon nearfield and its modulation by breathing-like acoustic vibrations. The concepts, the numerical and experimental approaches developed in this work are not specific to indented NCls. They can be extended to other isolated nano-objects exhibiting strong field localization, to dimers of nano-objects and to more complex metallic nanostructures combining size, shape and interaction effects.

References
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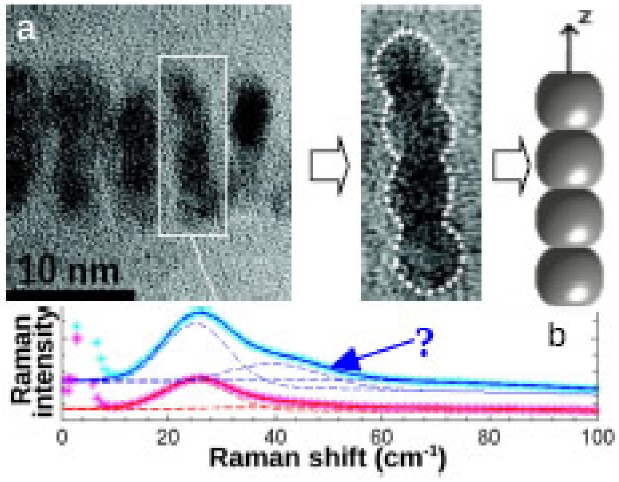

Figure 1 – (a) TEM images of silver NCls and the modelled indented NCl. (b) Raman spectra of the NCls.

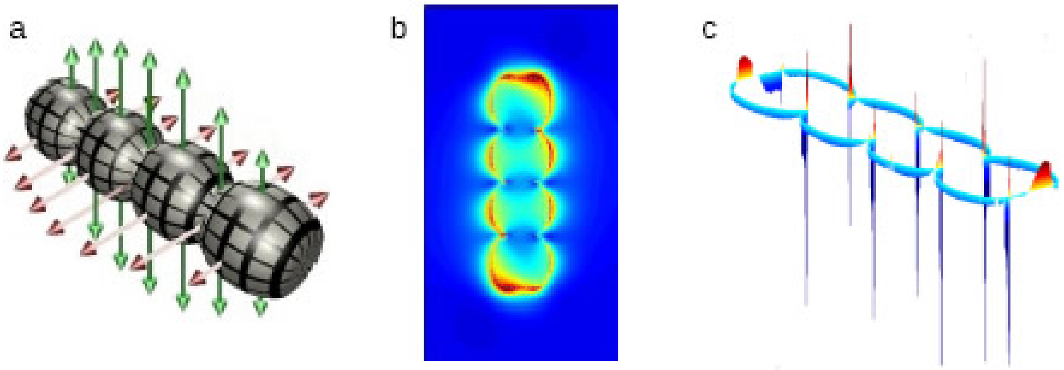

Figure 2 – (a) Displacement field of the breathing-like acoustic vibrations. (b) Surface plasmon nearfield for silver indented NCls. (c) Relative modulation of the surface plasmon polarization by the breathing-like acoustic vibrations δvibP(r)/P0(r).