Raman Pressure in Metallic Nano-Objects: a Picture of the Acousto-Plasmonic Interactions

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Metal nano-objects and nano-structures have recently attracted much interest due to their possible use in new nanoscale optical devices (nano-antennas, negative refraction lenses, surface wave guides) [1], for biomedical application and for the control of the light emission, absorption and scattering (Raman-Brillouin and Rayleigh scattering) in surface-enhanced spectroscopies (SERS, SEIRA) [2]. We focus here on the interaction between localized surface plasmons (LSP) and acoustic vibrations of metallic nano-objects. The coupling between acoustic vibrations and LSP still presents several theoretical challenges to correctly interpret Raman-Brillouin and pump-probe experiments. Indeed, a lot of theoretical and experimental works have been devoted to the understanding of shape, size, matrix and ordering effects on the LSP whereas only few deal with their dynamical properties (coupling mechanisms to the acoustic vibrations and inelastic light scattering properties).

Methods

We present a theoretical study of the interactions between acoustic vibrations (Fig. 1a) and localized surface plasmons (Fig. 1b) [3,4]. In this work, we introduced the concept of Raman pressure for the first time. It is used as a theoretical tool for the interpretation of resonant Raman scattering mediated by surface plasmons in metallic nano-objects. This physical quantity represents the electromagnetic energy density excited by the Raman probe and modulated by acoustic vibrations of the nano-object (Fig. 1c). The Raman pressure is a local quantity and can be mapped in the near-field region, thus providing a clear picture of the interaction between LSP and acoustic vibrations which give rise to inelastic scattering measurable in the far-field. The surface plasmons/acoustic vibrations coupling is calculated using the Boundary Element Method [5,6].



Figure: (a) Deformation of a silver nanocolumn induced by an acoustic vibration mode. (b) Near-field distribution associated with the LSP of the nanocolumn. (c) Modulation of the near-field by the acoustic vibration mode close to the nanocolumn surface.

Results

We show results on spherical nanoparticles, dimers, and nanocolumns. These results clearly show that for every particular acoustic vibration mode, the modulation of the LSP strongly depends on the size, the shape, the defects of the nano-object, and on the interaction between the nano-objects. The calculations/simulations allow us to understand and correctly interpret the activation of acoustic vibration modes and their relative Raman-Brillouin intensities.

References

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