Acousto-Plasmonic Coupling: The Raman Energy Density (RED)

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Interactions between elementary excitations, such as plasmon-exciton and plasmon-phonon, are of great interest from a fundamental point of view and for applications. While plasmon-exciton interactions have been extensively studied both experimentally and theoretically [1], the interaction mechanisms between acoustic vibrations (phonons) and localized surface plasmons (LSPs) remain largely unexplored [2]. Here we present a theoretical investigation of the interactions between confined acoustic vibrations and LSPs (Fig. 1, left) in the framework of resonant acoustic Raman scattering. We express the Raman scattering process in the framework of Fermi golden rule and introduce for the first time the concept of Raman energy density (RED; Fig. 1, center). Similarly to the Raman-Brillouin electronic density (RBED) introduced for semiconductors [3–4], this new physical quantity is used as a theoretical tool for the interpretation of resonant Raman scattering mediated by LSPs in metallic nanoparticles. The RED represents the electromagnetic energy density excited by the Raman probe and modulated by the acoustic vibrations of the nanoparticle. We show that, similarly to the local density of optical states (LDOS) and the RBED, the RED is a local physical quantity that can be mapped in the near-field region. It provides a clear picture of the interaction between LSPs and acoustic vibrations which give rise to inelastic Raman scattering measurable in the far-field. Here, we use the newly introduced RED concept to investigate elastic (an)isotropy effects and calculate the Raman selection rules of spherical nanoparticles in a dielectric environment (Fig. 1, right).

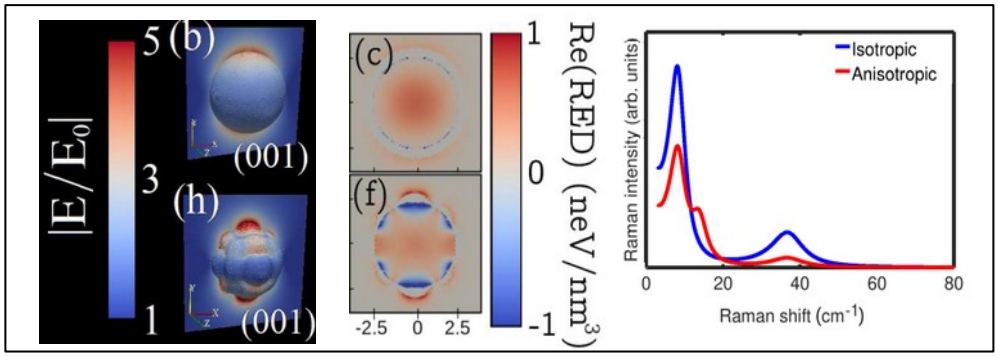

Fig. 1 Left: Isotropic (l=0) and Anisotropic (A_{1g}) breathing acoustic mode of a AuNP modulating the near electric field induced par the dipolar localized surface plasmon (LSP). Center: Raman energy density (RED) resulting from the interaction of the l=0 and A_{1g} vibration mode with the dipole LSP. Right: Calculated acoustic Raman spectra for the isotropic and anisotropic nanoparticles.

References

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