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Inelastic light scattering by the vibrational modes of single gold nanocrystals

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The measurement of acoustic vibrational modes is fundamental to the full comprehension of the mechanical properties of nano-objects, since they are related to the intrinsic characteristics of the material, such as crystallinity, size, shape, and elasticity.^[1] Furthermore, it has been shown that modifications in local environment can induce the appearance of new features in the inelastic light scattering spectra of nanoparticles that do not follow the usual selection rules.^[2] In this sense, optical spectroscopy techniques represent a powerful tool to study the vibrational modes of nano-objects and address both their intrinsic properties and their interaction with light, which plays a key role for applications in nanophotonics and optomechanics. In this contribution, we report low-frequency inelastic light scattering measurements on single gold nanocrystals, combined with detailed characterizations of their morphology and environment by transmission electron microscopy and tomography, and numerical simulations of their optical and vibrational responses. We demonstrate that low-frequency Raman spectroscopy of single nanoparticles, a simple to implement and non-destructive technique, can be used to study the coupling between light and vibration at the nanoscale, by monitoring the changes in the internal electric field of the nanoparticle. We show that shape anisotropy and electric field inhomogeneities within a particle, induced by a local change in the particle environment, lead to the appearance of inelastic light scattering by acoustic modes that are not predicted by usual Raman selection rules, which are based on the (here invalid) assumption of a symmetric internal electric field.^[3]

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