Mechanical coupling in gold nanoparticle dimers revealed by plasmon-enhanced ultra low frequency Raman spectroscopy

A. Girard[1],H. Gehan[1], A. Crut[1],L. Saviot[2], E.Cottancin[1], C.Bonnet[1], A.Mosset[1], A.Mermet[1] & J.Margueritat[1]

[1] *Institut Lumière Matière, Université Claude Bernard Lyon1, CNRS UMR 5306, Villeurbanne, France*

[2] *Laboratoire Interdisciplinaire Carnot de Bourgogne, Université de Bourgogne Franche Comté, CNRS UMR 6303, Dijon, France*

Acoustic vibrations of isolated gold nanoparticle dimers were investigated using ultra low frequency micro-Raman scattering and finite element simulations. When exciting the dimer resonantly with the surface plasmon resonance of electromagnetically coupled nanoparticles, the Raman spectra show two ultra-low frequency bands whose frequencies lie below the lowest Raman active Lamb mode of single nanoparticles. These features were ascribed to the vibration modes of gold nanoparticles "supermolecules", i.e. nanoparticles mechanically coupled by a surrounding polymer. Their measured frequencies are inversely proportional to the nanoparticle diameter and sensitive to the elastic properties of the interstitial polymer. The latter dependence as well as finite element simulations suggest that these modes are activated by the mechanical coupling between the nanoparticles, and they corresponds to the out-of-phase semirigid translation ($\ell = 1$ Lamb mode) of each nanoparticle of the dimer inside the matrix, along the direction of the short axis (lower frequency mode) or along the long axis of the dimer. In order to investigate in details the plasmon/vibration coupling, the optical properties of each studied dimer was acquired using SMS spectroscopy, and correlated to the Raman measurements and finally compared to the exact morphology of each dimer deduced by TEM. These measurements demonstrate that our observations were permitted only thanks to the resonant excitation with the coupling plasmon excitation, leading to an enhancement up to 10^4 of the scattering by these vibrations. Moreover it shows that the frequencies of the vibrations modes are extremely sensitive to the interstitial distance between nanoparticles.

These enhanced ultra low frequency Raman scattering opens a new route towards mechanical resonators such scattering properties are driven by the plasmon resonance ; i.e. nano-opto-mechanical resonators.