**Javier Aizpurua**
Center for Materials Physics (CSIC-UPV/EHU) and DIPC, Paseo Manuel Lardizabal 5, Donostia, Spain
aizpurua@ehu.eus

# Optomechanical approach to Surface-Enhanced Raman Spectroscopy

Molecular vibrations constitute one of the smallest mechanical oscillators available. The energy and strength of molecular oscillations depend on the attached specific functional groups as well as on the chemical and physical environments. Raman scattering can access the information contained in molecular vibrations, however, the low efficiency of the Raman process typically allows only for characterizing large numbers of molecules. To circumvent this limitation, plasmonic resonances supported by metallic nanostructures and nanocavities can be used because they localize and enhance light at optical frequencies, enabling surface-enhanced Raman scattering (SERS), where the Raman signal is increased by many orders of magnitude. This enhancement enables few- or even single-molecule characterization. The coupling between a single molecular vibration and a plasmonic mode constitutes an example of an optomechanical interaction [1], analogous to that existing between cavity photons and mechanical vibrations (see Fig. 1). Optomechanical systems have been intensely studied because of their fundamental interest as well as their application in quantum technology and sensing. In this context, SERS brings cavity optomechanics down to the molecular scale and gives access to larger vibrational frequencies associated with molecular motion.
Here, we describe this novel approach to SERS [2], and note that the molecular optomechanics framework naturally accounts for a rich variety of nonlinear effects in the SERS signal with increasing laser intensity. In this context, we review collective phenomena involving many molecules [3], and the modification of the effective losses and energy of the molecular vibrations due to the plasmon−vibration interaction. The quantum description of SERS also allows us to address the statistics of the Raman photons emitted, enabling the interpretation of two-color correlations of the emerging photons [4], with potential use in the generation of nonclassical states of light.

## References

[1] M. K. Schmidt, R. Esteban, A. González-Tudela, G. Giedke, J. Aizpurua, ACS Nano, 10 (2016) 6291-6298.
[2] R. Esteban, J. J. Baumberg, and J. Aizpurua, Accs. Chem. Res., 55 (2022) 1889-1899.
[3] Y. Zhang, J. Aizpurua, R. Esteban, ACS Photonics 7, (2020) 1676-1688.
[4] M.K. Schmidt, R. Esteban, G.Giedke, J. Aizpurua, A.González-Tudela, Quant. Sci. and Tech. 6 (2021) 034005.

## Figures

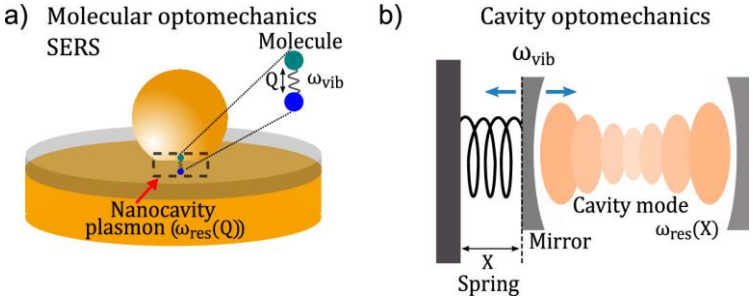


**Figure 1:** a) Schematics of a plasmonic nanocavity with resonance frequency $\omega_{res}$, and a molecule with vibrational frequency $\omega_{vib}$ located in it. It is possible to explore molecular optomechanics effects in this SERS configuration. b) Schematics of a canonical optomechanical system composed of a Fabry-Perot cavity with resonance frequency $\omega_{res}$, and a vibrating mirror with frequency $\omega_{vib}$.