Resonant Raman quantum engineering with fluorescent molecules

Daniele De Bernardis[1], Hugo Levy-Falk[1], Rocco Duquennoy[1], Elena Fanelli[1], Maja Colautti[1], and Costanza Toninelli[1]

1. *National Institute of Optics [Consiglio Nazionale delle Ricerche CNR–INO],*
care of European Laboratory for Non-Linear Spectroscopy (LENS),
Via Nello Carrara 1, Sesto Fiorentino, 50019, Italy

Abstract: Organic molecules trapped in solid-state matrices can be manipulated with Raman fields to produce new interesting vibrational states or to act as a THz-to-optical transducer.

We theoretically investigate a single fluorescent molecule as a hybrid quantum optical device controlled by multiple external laser sources. The theory is developed in the polaron framework, accounting for frequency filtered photon detection and multi-frequency Raman processes due to the coherent interactions between the electronic and vibrational degrees of freedom of the molecule. In this way the system is presented as a quantum toolbox that can simulate various cavity QED toy models exploiting a specific vibrational mode as the Bosonic single cavity mode. Each specific configuration tailors the steady-state of the open system, driving it into possibly exotic states, like vibrational cat states. Assuming to work with molecules with prominent spatial asymmetry, the resonant Raman engineering technology can be also combined with an external THz(IR) driving turning the system into a single photon transducer. Two realistic implementations based on the coupling to a subwavelength THz patch-antenna or metamaterial are proposed

Figure 1: Cartoon representation of the described system. A combination of various laser sources drives the molecule, mixing the electronic and vibrational dynamics. The signal is detected selecting specific side-bands.

Figure 2: (a) Fluorescence (Stokes, ZPL, anti-Stokes, from left to right) under the influence of various Raman schemes. (b) Wigner function of the molecular vibrational steady-state under the different schemes.