Beyond the Two-Level Description of Molecular Single-Photon Emitters in the High-Saturation Regime

Hugo Levy-Falk[1], Daniele De Bernardis[1], Elena Fanella[1,3], Louise Morlae[1,2], Marco Arzilli[1], Maja Colautti[1,2], Costanza Toninelli[1,2]

[1]*National Institute of Optics (CNR-INO), c/o LENS, Italy*
[2]*European Laboratory for Non-Linear Spectroscopy (LENS), Italy*
[3]*Physics Department, University of Naples, Italy*

Our group has strongly demonstrated the excellent properties of organic molecules trapped in solid matrices as single-photon sources in terms of indistinguishability, coherence, and photostability. [1, 2] When brought to cryogenic temperatures, such emitters can be described by a simple two-level system model. [3] Recently we proposed extending this toy-model description by explicitly including the internal vibrational degrees of freedom, with the final goal of turning them into compact optomechanical devices. [4] This however requires to excite them in the high-saturation regime to reach strong optomechanical coupling. It has been reported that in this regime molecular emitters show an anomalous behavior above a certain excitation intensity. [5]
The goal of this work is to clarify the physical origin of this effect. Our work shows that the latter is not compatible with a simple laser-induced heating of the matrix. Instead, the anomalous saturation is well described by absorption from the excited state towards a fast decaying dark state. We propose that this dark state is related to the matrix, linking anomalous saturation to the optical Stark shift.

References

[1] C. Toninelli *et al.,* "Single organic molecules for photonic quantum technologies" Nat. Mater., Vol. 20, No. 12, 1615–1628, 2021.

[2] S. Pazzagli *et al.,* "Self-Assembled Nanocrystals of Polycyclic Aromatic Hydrocarbons Show Photostable Single-Photon Emission" ACS Nano, Vol. 12, 4295, 2018.

[3] Q. Deplano *et al.,* "Sub-nanosecond coherent optical manipulation of a single aromatic molecule at cryogenic temperature", AVS Quantum Science, 5 (4), 2023.

[4] D. De. Bernardis *et al.,* "Hybrid interfaces at the single quantum level in fluorescent molecules" Quantum Sci. Techno. , Vol. 10, 045051, 2025.

[5] T. Plakhotnik, *et al.,* "Single-molecule spectroscopy" Annual review of physical chemistry, 48(1), 181-212, 1997.