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Quantum optics with organic molecules

Recent years have seen great progress in solid-state realizations of quantum optics experiments, which was enabled by sophisticated experimental techniques and advances in nanofabrication. In many cases these experiments rely upon and are driven by the specific quantum emitters, e.g., diamond color centers, semiconductor quantum dots, or rare-earth ions. In this talk I present one such emitter class pursued in our group: polyaromatic hydrocarbon (PAH) dye molecules embedded into solid-state matrices at cryogenic temperatures. Specifically, we use dibenzoterrylene (DBT) dye molecules embedded into anthracene or para-dichlorobenzene (pDCB) matrices. These systems show very attractive properties at low (2 K) temperatures: unlimited photostability, close to unity quantum yield, very little dephasing or spectral diffusion, strong (up to 50%) zero-phonon line (ZPL), and high brightness (typical lifetime of several nanoseconds). In my talk I will touch on two recent experiments with these emitters carried out in our group.

In the first [1], we placed a DBT-doped anthracene crystal into a microscopic free-space Fabry-Perot cavity, thus realizing a canonical Jaynes-Cummings model at the onset of the strong coupling regime. The Purcell enhancement induced by the cavity turned the molecule into a near-perfect two-level system, increasing its ZPL contribution from 30% to 95%. It also resulted in such effects as saturation of the emitter below single-photon level, strong light extinction and phase shift induced by a single molecule, and generation of non-classical light.

In the second experiment [2], we took first steps to make the system more scalable by moving from a free-space cavity to nanofabricated structures. The advantage of this approach is in having multiple molecules coupled to the same light mode, which facilitates long-range emitter-emitter interaction mediated by propagating light. I will present our results on coupling molecules to 1D nano-waveguides and waveguide-based ring resonators. In the waveguide case we could observe up to 7% extinction of the guided light induced by a single molecule. Replacing the waveguide with a resonator increased this extinction up to almost 25% due to the Purcell enhancement, showing good prospects for this approach.

[1] D. Wang, et al., *Nature Physics* 15, 483–489 (2019)

[2] D. Rattenbacher, et al., *arXiv: 1902.05257* (2019), accepted into *New Journal of Physics*

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