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Sub-nanosecond coherent optical manipulation of a single aromatic molecule at cryogenic temperature

¹Q. Deplano, ¹Ph. Tamarat, ¹B. Lounis, J.-B. Trebbia^{1*}

¹jean-baptiste.trebbia@u-bordeaux.fr, Université de Bordeaux, CNRS, Institut d'Optique, LP2N, F- 33405 Talence, France

The second quantum revolution presents challenges in manipulating and controlling quantum systems coherently. In solid-state physics, single emitters like molecules, quantum dots, and defects in diamond (NV centers) are promising for optical qubits or quantum networks due to their controllability with light and compatibility with nano-devices.

Single molecules trapped in the solid state at liquid helium temperatures are promising quantum emitters for the development of quantum technologies owing to their remarkable photostability and their lifetime-limited optical coherence time of the order of 10 ns. The coherent preparation of their electronic state requires resonant excitation with a Rabi period much shorter than their optical coherence time. Sculpting the optical excitation with sharp edges and a high on-off intensity ratio ($\sim 3 \times 10^5$) from a single-frequency laser beam, we demonstrate sub-nanosecond drive of a single dibenzanthrene molecule embedded in a naphthalene matrix at 3.2 K, over more than 17 Rabi periods, which to our knowledge constitutes the largest number of optical Rabi oscillations observed to date with a single solid-state emitter.

Coherent manipulation of the molecular electronic state, such as population inversion or superposition of the ground and excited states with maximal coherence, requires controlled pulse areas. With pulses tailored for a half-Rabi period, the electronic excited state is prepared with fidelity as high as 0.97. In order to characterize the robustness of the molecular transition dipole against decoherence induced by electron-phonon interaction, we have investigated the temperature dependence of the dephasing rate using single-emitter Ramsey spectroscopy. Using single-molecule Ramsey spectroscopy, we prove up to 5 K that the optical coherence lifetime remains at its fundamental upper limit set by twice the excited-state lifetime, making single molecules suitable for quantum bit manipulations under standard cryogen-free cooling technologies.

Reference :

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