Individual Addressing of Molecular Carbene Spin-Qubits for Spin-Selective Single-Photon Emission

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The realization of large-scale quantum networks and distributed quantum computing architectures requires robust, scalable spin-photon interfaces capable of deterministic entanglement. While solid-state color centers provide excellent spin coherence, their integration into scalable nanophotonic architectures is fundamentally hindered by stochastic defect creation, challenging heterogeneous integration and susceptibility to spectral diffusion near surfaces.

Conversely, molecular spins offer atomic-level reproducibility, high tunability and relatively simple integration into photonic integrated circuits (PIC). Recent progress has shown that molecular emitters integrated on PICs can be used as single photon sources, producing remarkably indistinguishable photons [1]. However, as mentioned in the most up to date review of organic molecules as single photons sources [2], all current molecular emitters with an internal spin degree of freedom have been detected only in bulk, and could not be addressed individually.

Here, we present for the first time individual addressing of molecular emitters with an internal spin degree of freedom, leading to *spin-selective single photons*. Our novel purely organic molecular quantum platform is based on photo-active ground-state triplet carbenes embedded in a rigid crystalline host matrix [1]. Following iterative optimization of the carbene molecules and crystal host, we have achieved individual addressing of the electron molecules, which act as single-photon sources with exceptional spin-photon interface properties.

We demonstrate the isolation and coherent control of single molecular electron spins, reporting highly stable, narrow optical emission and extremely long spectral stability without spectral wandering. By exploiting highly spin-selective intersystem crossing pathways, we achieve single-molecule optically detected magnetic resonance (ODMR) with record-high fluorescence contrast, and we perform fast coherent spin manipulation via microwave driving, observing high-visibility Rabi oscillations. Furthermore, using XY-8 dynamical decoupling sequences, we achieve $T_{2dd} > 1$ms. For integration with PICs, we show that these molecular crystals can be grown as thin films that retain their exceptional optical and spin properties. Our results establish carbene-based molecular architectures as a highly coherent and photonics-ready platform for the generation of entangled photons and distributed quantum processing.

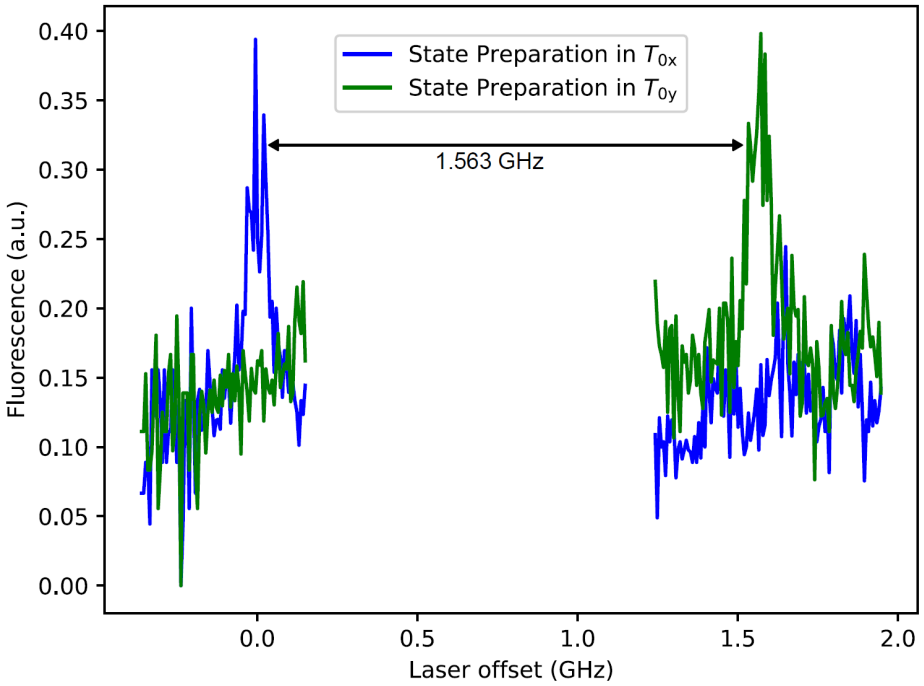

Fig. 1: Selective excitation of single photons depending on the internal spin state of the molecular emitter. Each spin state has a different excitation frequency (and produces a different photon frequency), separated by 1.563 GHz, thus enabling generation of photons entangled to the spin state.

References

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