Spectroscopy and cavity-enhanced emission of Europium-based molecular systems

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Rare-earth ions in solid-state hosts are a promising candidate for optically addressable spin qubits, owing to their excellent optical and spin coherence times. Recently, Eu^{3+}-based molecular materials have also been shown to possess excellent optical coherence properties [1]. However, Eu^{3+}-doped nanocrystals have a long optical lifetime of the $^5D_0$-$^7F_0$ transition ($T_{1,opt}$ ~ ms) and a low branching ratio (<1%) [2], limiting single-ion experiments. Both issues can be solved by enhancing the emission of Eu^{3+} with high-finesse fiber-based microcavities.

We study Eu^{3+}-doped molecular crystalline materials and powders, exhibiting long spin lifetimes and narrow homogeneous linewidths at 4.2K [1,3]. On a single, macroscopic molecular crystal of [Eu(Ba)4(pip)] [see Figure 2], we measure narrow inhomogeneous linewidths, hour-long spin T_1, and photon echoes at <1K. Steps to integrate molecular crystals into a fiber cavity in the form of a crystalline thin film are reported. Open-access Fabry-Pérot fiber cavities have been demonstrated to achieve high quality factors and low mode volumes, while simultaneously offering large tunability and efficient collection of the cavity mode [4]. The home-built cavity setup was successfully integrated into a cryostat and demonstrated high mechanical stability during operation, which is required for cavity-enhanced ensemble spectroscopy.

The presented results are important steps towards single-ion readout and control being necessary for scalable quantum registers.

References

[1] Serrano et al., Nature, 603, 241–246 (2022)
[2] Bartholomew et al., Nano Lett., 17, 2, 778–787 (2017)
[3] Kuppusamy et al., J. Phys. Chem. C 127, 22 (2023)
[4] Hunger et al., New J. Phys 12, 065038 (2010)

Figures

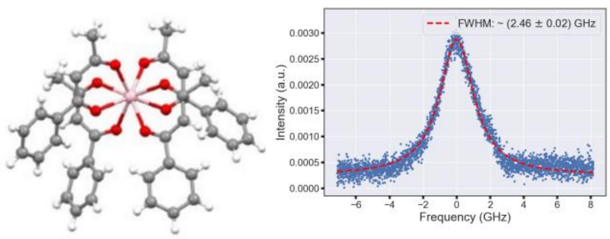

Figure 1: Left: Mononuclear Eu^{3+}-based molecular complex [1]. Right: Narrow inhomogeneous line of the $^5D_0$-$^7F_0$ transition measured in a powder sample.

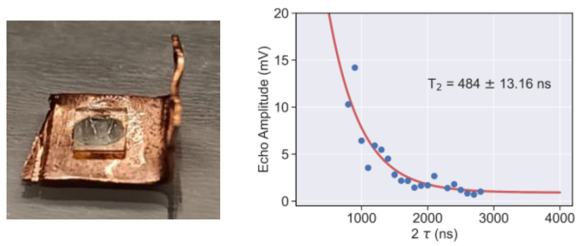

Figure 2: Left: Macroscopic molecular crystal on a copper holder was installed in a dilution refrigerator. Right: Measured two-pulse photon echo decay for the $^5D_0$-$^7F_0$ transition.

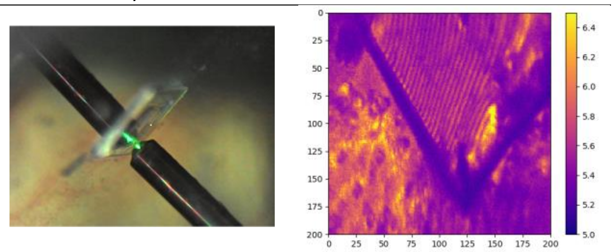

Figure 3: Left: Cavity integration of molecular crystals. Right: Single 2D – transmission raster scan of a crystalline region, carried out at room temperature. X- and Y-axes are pixel numbers.