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Multi-Quantum Spectroscopy of Anisotropic Muonium

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Multi-quantum (MQ) spectroscopy allows to selectively probe anisotropic muonium. The principal idea is to resonantly drive MQ spin transitions, for which both the electron and the muon spin flip simultaneously (Fig. 1a). In the high-field limit of the electron spin, such MQ microwave excitations are only possible in presence of hyperfine anisotropy, as demonstrated recently for bond-centered muonium in crystalline Si 1.
Here we extend the approach to systems with random orientation. The first system is $SrTiO_3$, where a polaronic muonium state has been reported with TF-μSR on single crystals. Using MQ spectroscopy, the presence of this state can also be detected in a powder sample of $SrTiO_3$ and manifests as a resonance curve recorded in LF mode (Fig. 1b). The second system is the catalytic zeolite framework titanium silicalite (TS-1). A MQ resonance curve was detected successfully at 260 K, which confirms the formation of anisotropic, weakly-coupled muonium and holds promise for further μSR studies that focus on catalytic reactions with TS-1.
Besides these results, we outline extensions of the technique, namely MQ experiments at higher fields and microwave frequencies that have the potential to complement ALC μSR on muoniated radicals.

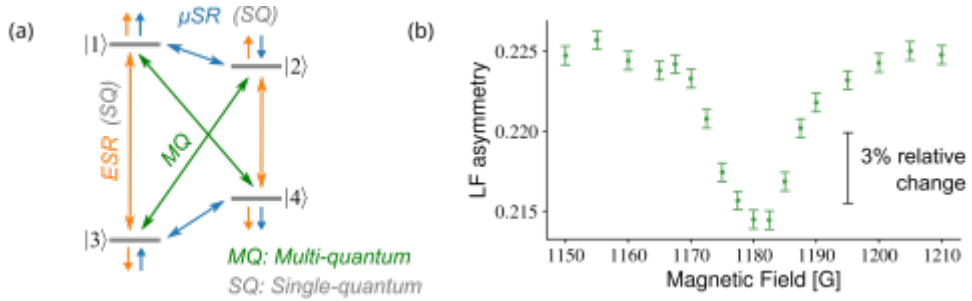

Figure 1: (a) Energy levels for MQ spectroscopy. (b) MQ spectrum or $SrTiO_3$ at 17 K with 3.335 GHz drive.

1 A. Doll et al., arXiv:2503.24023 (2025).

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