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Second order Zeeman interaction and ferroquadrupolar order in $TmVO_4$

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$TmVO_4$ exhibits ferroquadrupolar order of the Tm 4f electronic orbitals at low temperatures, and is a model system for Ising nematicity. A magnetic field oriented along the *c*-axis constitutes a transverse effective field for the quadrupolar order parameter, continuously tuning the system to a quantum phase transition as the field is increased from zero. In contrast, in-plane magnetic fields couple to the order parameter only at second order, such that orienting along the primary axes of the quadrupole order results in an effective longitudinal field, whereas orienting at 45 degrees results in a second effective transverse field. Nuclear magnetic Resonance (NMR) studies of these effects are hampered by increased linewidth of the spectra due to inhomogeneous demagnetizing fields but can be minimized by cutting the sample to an ellipsoidal shape using a Xe plasma FIB.

Previous NMR measurements of $TmVO_4$ identified a scaling between the spin–lattice relaxation rate and the shear elastic stiffness constant, c_{66}, suggesting that the ^{51}V ($I = 7/2$) nuclear spins couple to the Tm orbitals through the electric field gradient (EFG), giving rise to a quadrupolar relaxation channel [1]. However, the spectra were significantly broadened by inhomogeneous demagnetization fields and the anisotropic *g*-factor of the Tm ground state doublet. In order to better discern the spectra and relaxation mechanisms at play, we reshaped a single crystal of $TmVO_4$ to an ellipsoidal shape, with a homogeneous demagnetization field (see Fig. 1). We utilized a Xe^{2+} plasma focused ion beam (FIB) by Thermo Fisher Scientific with a 30 kV, 1 μA beam to cut our sample with the long-axis along the *c*-axis of the crystal. Sample damage from the beam is only expected on the surface within a depth of 30–40 nm and energy dispersive X-ray analysis (EDX) of a test surface verifies the unchanged composition of $TmVO_4$ below. The final sample diameter is 0.4 mm and the length of 1.3 mm require a total cutting time in excess of 25 h of each side. The magnetic broadening was dramatically reduced

in the FIB crystal, such that each of the seven peaks separated by the quadrupolar splitting are clearly resolved. The ability to resolve all seven peaks is important because it enables us to extract details of the magnetic and quadrupolar contributions to the spin–lattice relaxation rate that would otherwise be inaccessible, as discussed above in the section on Spin-lattice relation [2].

[1] Z. Wang, I. Vinograd et al.; *Anisotropic nematic fluctuations above the ferroquadrupolar transition in $TmVO_4$; Phys. Rev. B* **104**, 205137 (**2021)**.

[2] I. Vinograd, K. R. Shirer et al; *Second order Zeeman interaction and ferroquadrupolar order in $TmVO_4$; npj Quantum Materials* **7**, 68 (**2022)**.

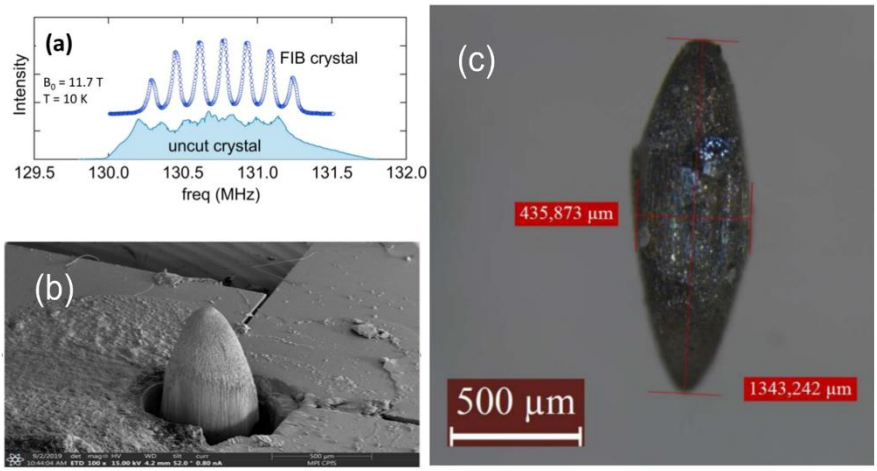

Fig. 1: (a) ^{51}V-NMR spectrum with and without ellipsoidal crystal shape. (b) Scanning electron microscopy (SEM) scan of the sample during the FIB process. (c) Crystal after FIB. Al and C are deposited on the sample surface layer during the FIB processing, but do not contribute to the NMR signal.