Experimental report

15/10/2024

Proposal: **4-04-523**

Council: 4/2023

Title: Magneto-elastic coupling in the ¿Faraday-rotator¿ KTb3F10

Research area: Physics

This proposal is a new proposal

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Samples: KTb3F10

Instrument	Requested days	Allocated days	From	To
IN8	8	8	30/10/2023	07/11/2023

Abstract:

In recent years, the study of the coupling between the 4f electronic density and lattice degrees of freedom has aroused a lot of attention. Such interactions are suspected to play a key role in a number of issues, ranging from highly frustrated magnetism to the design of smart materials in optics. Preliminary single-crystal neutron diffraction experiments performed on the Faraday rotator KTb3F10 have revealed a decrease of the intensity on several Bragg reflections in a magnetic field up to 2 T, anomaly which can be interpreted as originating from a magneto-elastic coupling. Such a coupling would imply changes in the phonon spectrum, as well as in the crystal electric field energy levels, when a magnetic field is applied. Inelastic neutron scattering is the best tool to access S(Q,E) for both structural and magnetic excitations, and we are therefore requesting beamtime on the thermal spectrometer IN8, whose energy range is ideally suited for this study. Experiments will be carried between 2 and 300 K, for H = 0-6T.

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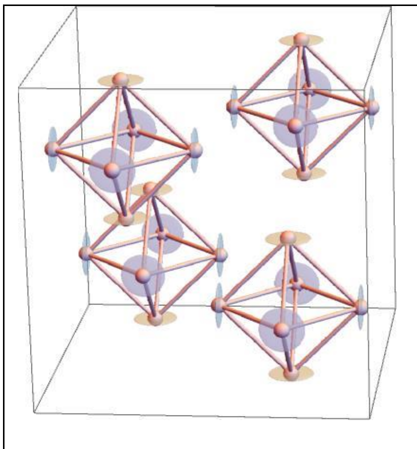
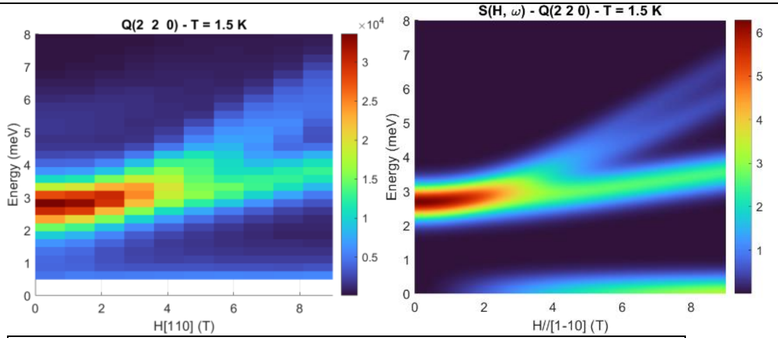
KTb_3F_{10} (Figure 1) is an interesting model system to investigate the so-called "induced magnetism". In such systems, the rare earth CEF ground state is a singlet, below a doublet located at the energy Δ. Using previous neutron data combined with point charge calculations, we could indeed confirm that the Tb^{3+} CEF scheme in KTb_3F_{10} indeed consists in such a singlet ground state $|O\rangle = |J=6, J_z=0\rangle$ below an excited doublet $|\pm\rangle = |J=6, J_z=\pm1\rangle$ located at Δ = 2.7 meV.

In this experiment on IN8, we were interested in the field evolution of both the CEF modes and lattice excitations. IN8 was operated under standard conditions, with SI111/PG002 kf =2.662 Å^{-1}. The KTb_3F_{10} crystal was aligned with (001) and (110) in the horizontal plane, and inserted into an aluminum sample holder attached to the stick of the 10T cryomagnet.

We first measured the field dependence of the CEF mode, as illustrated in Figure 2. With increasing the field, the mode at $\Delta = 2.7$ meV level splits into several modes, an evolution well captured by our CEF calculations.





CEF parameters (meV)				
B_0^2	B_0^4	B_4^4	B_0^6	B_4^6
-84.382	-74.124	23.573	11.345	0.144

Figure 1: Crystalline structure of KTb_3F_{10}. For simplicity, only the octahedra formed by nearest neigbors Tb ions are shown. The disks feature the anisotropy planes of the induced magnetization. Cubic *Fm-3m* space group, see also Ref [2].

Figure 2: (left) KTb_3F_{10} inelastic neutron scattering measurements at (220) and 1.5 K as a function of the applied field along (1-10). (IN8@ILL, k_F = 2.662 Å) and (right) calculated inelastic neutron scattering cross section. CEF parameters are given in the table.

Next, we heated up to 200K to better observe the phonon modes around polarized along (110) (Q=660), longitudinal and dispersing along (HH0) and transverse, dispersing along (00L). Measuring the same phonon dispersion at low temperature proved quite difficult because of the strong decrease of the Bose factor, but also because of the large increase of the Boltzmann factor, which very significantly increases the magnetic response of the transition at Δ. We could sort this out by making Q scans at constant energy rather than energy scans at constant Q. To the accuracy of this experiment, the phonon dispersion does not evolve both with H and T.

We eventually mapped out the region close to (220) and (660) up to 15 meV. In addition to the splitting described above, another mode close to 10 meV rises up (see Figure 3). According to our CEF calculations, it originates from an 8 meV mode (in zero field) whose intensity increases with increasing H. Along (22L), this mode is apparently dispersing, as illustrated in Figure 4: the bottom of the band is at (220) and (222), while the top is reached at (221) and (22-1). The bandwidth of this dispersion seemingly depends on the intensity of the field (not shown here). Strikingly, the fact that a CEF level can be so widely dispersed is troubling in itself. What's more, these data suggest that the CEF mode seems to be pushed upwards by the rising branch of the transverse acoustic phonon (propagating along (00L) and polarized along (HH0)). This branch is not observed in Figure 4 because the phonon cross section is too weak around this Q but also because the intensities of the low energy CEF modes are very strong. The dispersion measured at 200 K is reported in Figure 4 using dashed lines. Finally, among other unexplained phenomena, the intensity of the CEF mode at 5 meV experiences some significant variation at the crossing point with the same acoustic branch.

These features and their H dependence (that we recorded during our beamtime on IN8) is currently under analysis.

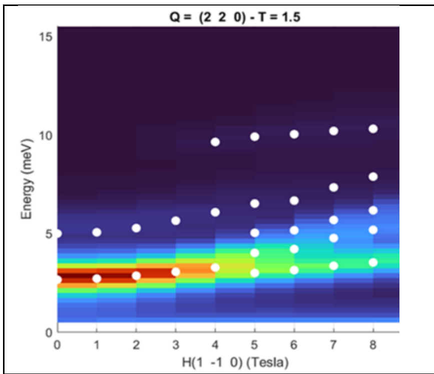


Fig. 3 : measurements at (220) and 1.5 K as a function of the applied field along (1-10).

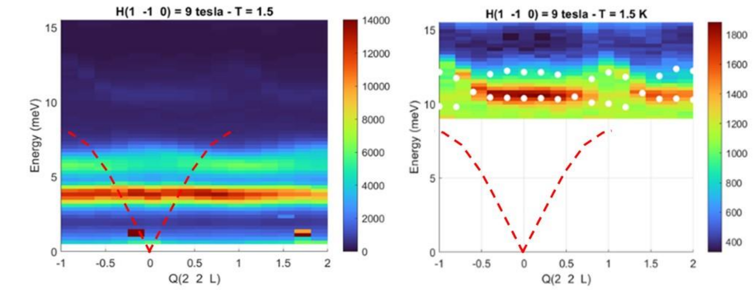
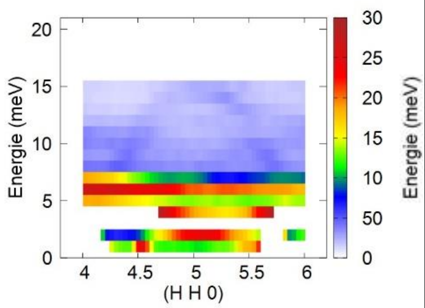
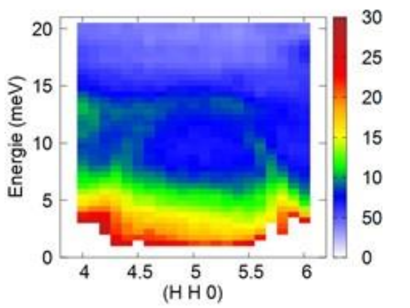


Fig. 4: scans along (22L) at 1.5 K at H=9 T, showing the dispersion of the CEF lines The two panels are identical but displayed with different scales to better highlight the high energy mode close to 10 meV.





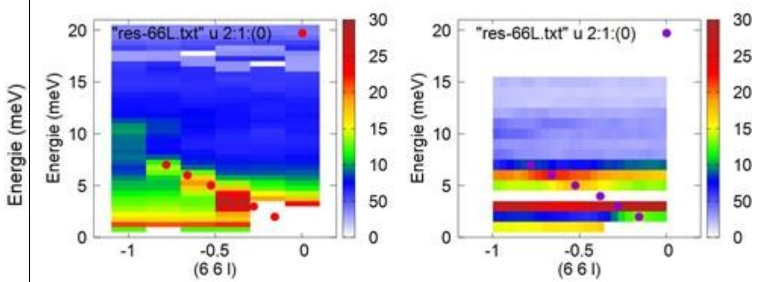


Fig 5: maps showing the Longitudinal (left) and Transverse (above) phonons dispersing along (HH0) and (00L) at 2 temperatures, 1.5 and 200K. The red dots indicate the fit of the phonon transverse dispersion.