Crystalline electric field excitations in $NdRuSn_3$

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Quantum spin liquid states arising from magnetic frustration have been explored in various materials. In the present study, we focus on the *Ln*$RuSn_3$ (*Ln* = lanthanide), which is expected to exhibit a new spin liquid state.

Ln$RuSn_3$ crystallizes in a cubic structure with space group $Pm\bar{3}n$, as shown in Fig. 1 [1]. The *Ln* ions, represented by large spheres, occupy two Wyckoff sites. The nearest-neighboring *Ln* ions (6d site) form rod-like lattices. These rods, oriented along the fourfold axes of the cubic unit cell, interact with each other through the third-neighbor triangular lattice. Additionally, the *Ln* site at the corners and the body center of the unit cell (2a site) coordinate to the *Ln* ions on the rods via second-neighbor interactions.

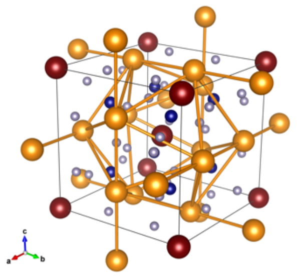

Fig. 1. Crystal structure of *Ln*$RuSn_3$, drawn using the VESTA software [2]. Large brown and orange, middle blue, and small grey circles are Nd, Ru, and Sn atoms, respectively.

The effective magnetic moment of $3.62\mu_B$/Nd in $NdRuSn_3$, evaluated from the Curie–Weiss law analysis for the high-temperature magnetic susceptibility data, is consistent with the Nd^{3+} $4f^3$ electron states. The Weiss temperature $\Theta_W = -11.1$ K indicates antiferromagnetic interactions. On the neutron powder diffraction experiments, performed at JRR-3 T1-1, no signs of magnetic order were observed even at 0.7 K, which is well below $|\Theta_W|$. The nonmagnetic ordering feature is consistent with the results in previous studies [3]. To understand the suppression of magnetic order, we studied crystalline electric field (CEF) splitting scheme of Nd ions using an inelastic neutron scattering (INS) technique, performed at the triple-axis spectrometer PONTA (5G). The final neutron energy at 14.7 meV was chosen. Sample temperature was controlled using a helium-gas closed cryostat.

Figure 2 shows INS spectra measured at the scattering vector magnitude Q = 1.5 Å^{-1} (red marks) and 3 Å^{-1} (blue marks) for $NdRuSn_3$ at 6 K. The CEF split levels are five doublets at the 6d sites ($\bar{4}$m.2) and one doublet with two quartets for the 2a site (m$\bar{3}$.). Therefore, the number of low-temperature excitation peaks are six in total. The data measured at Q = 3 Å^{-1} are fitted by six Gaussian peaks, as shown by red solid line in Fig. 2. This result supports the magnetic susceptibility increase with decreasing temperature owing to the magnetic ground state at the Nd ions. Consequently, the fact of no magnetic ordering indicates a signature of magnetic frustration effect.

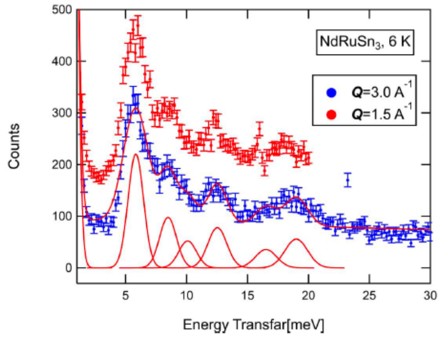

Fig. 2. INS spectra at fixed Q = 1.5 Å^{-1} (red marks) and 3 Å^{-1} (blue marks) of $NdRuSn_3$. Red solid line is a fitted result to the Q = 3 Å^{-1} data.

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[1] Binod K Rai et al., J. Phys.: Condens. Matter **34**, 273002 (2022). [2] K. Momma and F. Izumi, J. Appl. Crystal. **41**, 653 (2008). [3] T. Fukuhara et al., J. Phys.: Condens. Matter **3**, 8917 (1991).