Experimental report

28/10/2015

Proposal: **4-01-1378** **Council:** 4/2014

Title: Investigation of the crystal fieldscheme of the first spinel spin ice $CdEr_2Se_4$

Research area: Physics

This proposal is a new proposal

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Samples: 114CdEr2Se4
Tb2Hf2O7

Instrument	Requested days	Allocated days	From	To
IN4	4	4	17/11/2014	21/11/2014
IN6	4	4	24/11/2014	28/11/2014

Abstract:

CdEr2Se4 was recently proposed to be the first spinel that possesses a spin ice state. Although specific heat and susceptibility measurements support this picture, till now no microscopic evidence has been provided. Here we propose to use inelastic neutron scattering to investigate the crystal field transitions in CdEr2Se4. Our experiment will verify the Ising character of Er^{3+} spins, which is crucial for spin ice behavior. Additionally we will measure quasielastic scattering, to develop understanding of possible spin relaxation mechanisms relevant to the hopping of emergent magnetic monopoles at low temperature (if such things exist in CdEr2Se4).

As was scheduled in the proposal, we investigated the crystal electric field (CEF) scheme in the first spinel spin ice candidate $CdEr_2Se_4$ on inelastic neutron spectrometers IN4 and IN6. Altogether 9 g of ^{114}Cd isotope-enriched $^{114}CdEr_2Se_4$ sample was put into an annular aluminum can with outer/inner diameter of 12/8 mm. For the IN4 measurements, the incoming neutron wavelengths were 1.14 and 2.4 Å, which cover the positive energy transfer side up to 38 meV. For the IN6 measurements, 5.1 and 5.9 Å incoming neutron wavelengths were employed, providing a high-resolution for E < 10 meV at the negative transfer side. Temperatures of 2, 25, 50, and 100 K were chosen to map the evolution of the CEF transitions.

Fig.1 shows the Q-E map for the CEF transitions measured at T = 2 K with 1.14 and 2.4 Å incoming neutron wavelength. Several flat transitions can be observed. From the Q-dependence of the transition intensity, we can deduce the origin of the transition. For example, the intensity of the transition at about 18 meV increases with Q, revealing its phonon origin. While some other transitions at about 10 and 28 meV can be fitted very well by the magnetic form factor of Er^{3+} ions and therefore can be attributed to the CEF transitions.

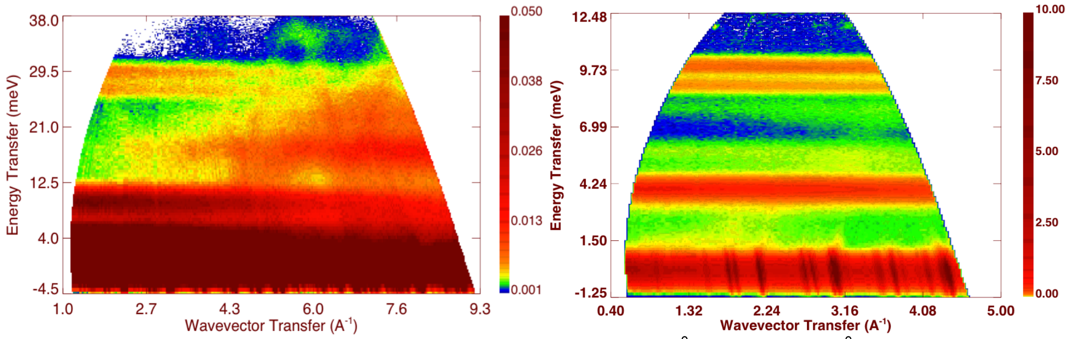

Fig. 1 Q-E map measured on IN4 at T = 2 K with (a) 1.14 Å and (b) 2.4 Å incoming neutron wavelength.

The Q-E map shown in Fig.2 was measured on IN6 at 25 K for the negative energy transfer side. Different from the positive energy transfer side, the negative side relies on the anti-stokes CEF transitions and therefore can only be observed at a high temperature where the excited states are populated.

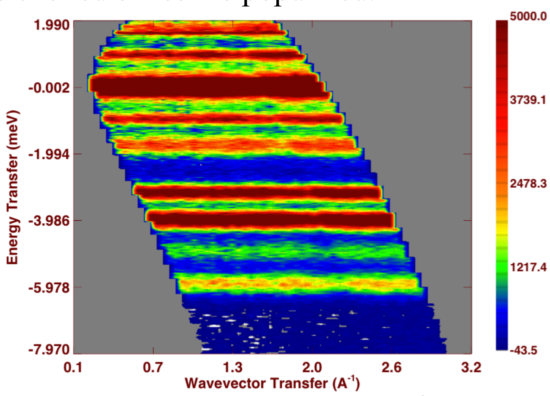

Fig. 2 Q-E map measured on IN6 at T = 25 K with 5.1 Å incoming neutron wavelength

From these measurements, altogether we could resolve 6 transitions from the CEF ground state (see Fig. 3). By fitting these transitions using the McPhase package, the ground state is found to be:

Ground state doublet: $|\pm\rangle = \pm 0.9081|\pm 15/2\rangle + 0.3845|\pm 9/2\rangle \pm 0.1495|\pm 3/2\rangle$
$+0.0725|\mp 3/2\rangle \pm 0.0038|\mp 9/2\rangle$

g-tensor: $g_{xx} = 0, g_{yy} = 0, g_{zz} = 16.5020$

which shows an obvious Ising character. In this way, we provide the first microscopic evidence that the Er^{3+} manifest Ising character in $CdEr_2Se_4$, which is the prerequisite for the realization of spin ice.

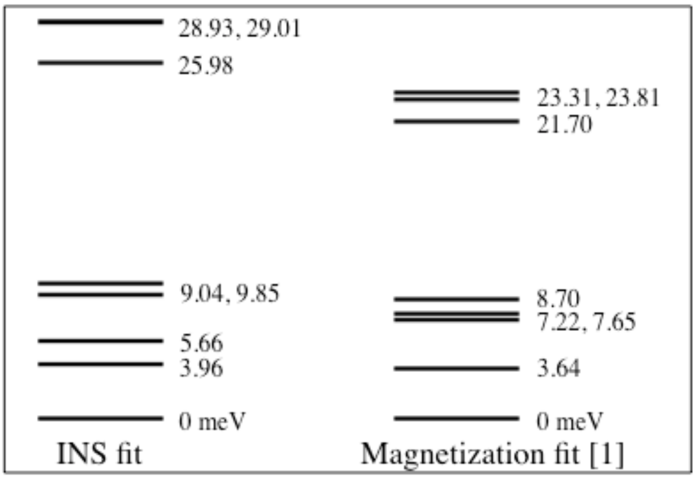

Fig. 3 CEF scheme from our inelastic neutron scattering study. Also shown is the previous CEF scheme extracted from the fit to magnetization data.

As a summary, we successfully measured the CEF transitions on IN4 and IN6 at ILL. By fitting to the temperature-dependent data, we extracted the CEF ground state. The extracted ground state manifests a strong Ising character and thus provides evidence that $CdEr_2Se_4$ satisfies the prerequisite for hosting the spin ice state.