Experimental report

11/02/2017

Proposal: 5-32-792 **Council:** 4/2014

Title: Diffuse Neutron Scattering Study of the spin correlations in the first spinel spin ice CdEr2Se4

Research area: Physics

This proposal is a new proposal

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Samples: 114CdEr2Se4

Instrument	Requested days	Allocated days	From	To
D20	3	3	23/06/2015	26/06/2015
D7	5	5	14/10/2015	19/10/2015

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 diffuse neutron scattering to investigate the static spin correlations in CdEr2Se4. Our experiment will reveal the temperature evolution of the spin correlation function and provide solid evidence of the spin ice behavior in CdEr2Se4.

As was scheduled in the proposal, we investigated the short-range spin correlations in the spin ice candidate $CdEr_2Se_4$ [1] using the non-polarized neutron diffractometer D20 and the polarized neutron diffractometer D7. For the D20 measurements, 9 g of $^{114}CdEr_2Se_4$ powder was filled into an annular-shaped copper can with out/inner diamters of 6/10 mm, and the setup with 2.41 Å incoming neutron wavelength was used. For the D7 measurements, 5.26 g of $^{114}CdEr_2Se_4$ powder was filled into an annular shaped copper can with outer/inner diameters of 15/14 mm and the setup of 4.8 Å incoming neutron wavelength was used.

Fig. 1a presents the typical D20 results measured at 0.5, 1.5, 4.0, and 8.0 K. Measurement at 20 K has been subtracted as the background. As can be seen from the plot, broad peaks around 0.6 and 1.4 Å start to form below 8 K, with their intensities growing with decreasing temperatures. Such a feature is compatible with what is normally observed for classical spin ices [2] and is a strong indication for the ice correlations in $CdEr_2Se_4$

Near the base temperature of 0.5 K, a series of sharp peaks are also observed. As is detailed in Fig. 1b, the temperature evolution of these sharp peaks are different from the broad diffuse features: intensities of the sharp peaks saturate at temperatures below 0.8 K while the broad peaks from diffuse scattering continues their growing. Thus the origin of the sharp peaks should be different from the main feature. Combined with the synchrotron diffraction data, we can attribute the sharp peaks observed in Fig. 1b to the long-range magnetic order of impurity erbium selenium Er_xSe_y

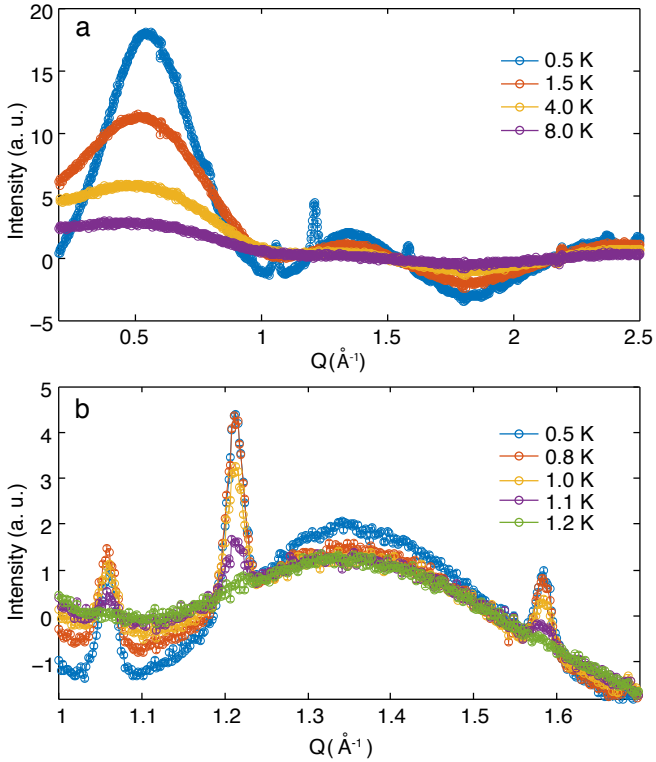

Fig. 1 (a) Non-polarized neutron diffuse scattering results for $CdEr_2Se_4$ measured on D20 at 0.5, 1.5, 4, and 8 K. The 20 K measurement has been subtracted as the background. **(b)** Detailed temperature dependence of the intensities in the region of 1-1.7 Å in between 0.5 and 1.2 K.

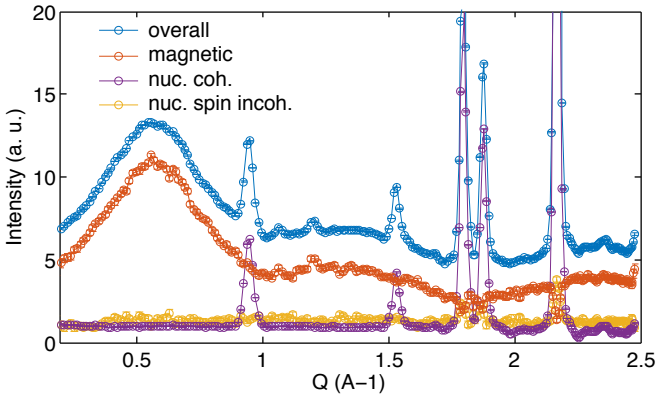

Fig. 2 xyz polarization analysis for the diffuse scattering of $CdEr_2Se_4$ measured at 70 mK on D7, showing the separation of the magnetic scattering component from the nuclear coherent/isotope incoherent scattering and the nuclear spin incoherent scatterings.

Fig. 2 presents the xyz polarization analysis results for $CdEr_2Se_4$ measured at 70 mK. The magnetic scattering has been separated from the nuclear coherent/isotope incoherent scatterings and the nuclear spin incoherent scatterings using the 10-points method. Similar to the results of non-polarized neutrons shown in Fig. 1a, broad diffuse peaks can be observed around 0.6 and 1.4 Å, which suggests the short-range ice correlations in this system.

Reverse Monte Carlo simulations are performed with the spinvert program to fit the D7 data, and the results are shown in Fig. 3a. One advantage of the reverse Monte Carlo simulation is its ability to recover the diffuse scattering pattern for single crystals without any intermediate microscopic model. Fig. 3b presents the spin correlations in the (hhl) plane calculated from the average of 10 independent reverse Monte Carlo simulation runs. The overall pattern looks very similar to that of the dipolar spin ice [2]. Therefore, our reverse Monte Carlo simulation results corroborate the ice correlations in $CdEr_2Se_4$.

As a summary, our D7 and D20 experiment successfully reveals the ice-correlations in $CdEr_2Se_4$. Combined with the crystal electric field results obtained from IN4 and IN6, we establish $CdEr_2Se_4$ as the first spinel that realizes the spin ice state.

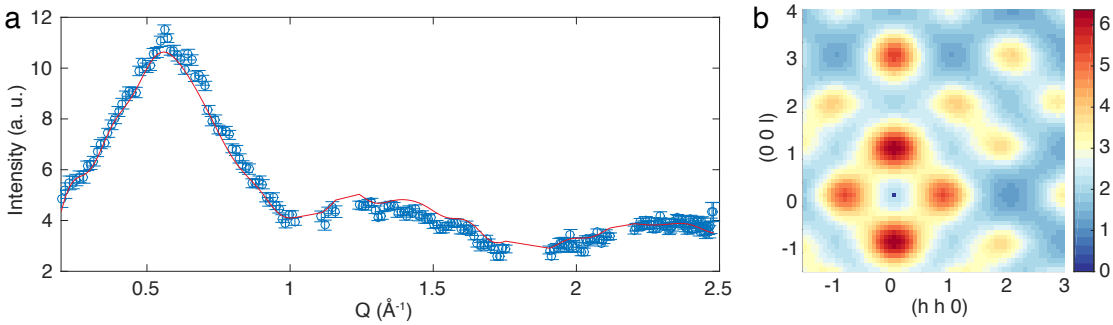

Fig. 3 Mean-field calculation results for the magnetic scattering data measured on D7. Data at T = 0.07, 0.5, 1.5, 5, and 20 K are shifted by 9, 18, 27, 36, and 45, respectively. Blue circles are the experimental data. Red lines are the mean-field calculation results.

[1] J. Lago et al., PRL 104, 247203 (2010)
[2] S. T. Bramwell et al., Nat. 294, 1495 (2001)