Experimental Report

21/01/2014

Proposal:	**4-03-1698**	**Council:**	10/2012
Title:	Detailed measurements of the crystalline electric field of SrHo2O4		

This proposal is continuation of: 4-01-1223

Researh Area: Physics

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Samples: SrHo2O4

Instrument	Req. Days	All. Days	From	To
IN5	4	4	25/02/2013	01/03/2013

Abstract:

We propose a continuation experiment to measure the crystalline electric field of SrHo2O4 using the IN5 spectrometer in order to map out the low energy part of the spectrum with sufficient resolution. SrHo2O4 belongs to a family of compounds with the formula SrLn2O4, where Ln = Lanthanide. The triangular chains of Ln ions that run along the c-axis make the SrLn2O4 compounds suitable candidates to exhibit geometric frustration. Recent collaboration with a theoretical group has allowed us to start understanding the rather complicated crystal field scheme of one member of this family, SrHo2O4. So far we have made some measurements (using the IN4 spectrometer) of the temperature dependence of the higher energy crystal field levels for this material down to 1.6 K. We would like to now extend this study to measure the changes down to dilution temperatures and with an improved energy resolution in the 0 to 5 meV range using the IN5 instrument. These measurements would further the theoretical understanding of SrHo2O4.

$SrHo_2O_4$ is a geometrically frustrated magnet because the Ho^{3+} ions are arranged in a network of triangles and hexagons, and are coupled with antiferromagnetic exchange interactions. The crystal structure, space group *Pnam*, allows for the presence of two inequivalent sites of the rare earth ions in a unit cell [1]. In zero field, measurements on a single crystal sample of $SrHo_2O_4$ [2] revealed that there are two distinct types of magnetic short-range order, which can be inferred from the neutron diffraction patterns, arising from the two different sites for the Ho^{3+} ions. In the $(hk0)$ plane diffuse magnetic scattering appears around the $\mathbf{k} = 0$ positions below 0.7 K. Whereas rods of scattering intensity seen at much higher temperatures in both the $(h0l)$ and $(0kl)$ planes in reciprocal space at $Q = (00\frac{1}{2})$ and symmetry related positions suggest that the second type of short-range order present in this material is one-dimensional. From the *XYZ*-polarised neutron measurements on D7 it can be inferred that the spins that participate in the first (second) stucture are antiferromagnetically coupled and are pointing along the c-direction (b-direction).

In order to try and understand the highly anisotropic behaviour of $SrHo_2O_4$, inelastic neutron scattering was used to probe the crystal field and cooperative excitations in order to determine the nature of the interactions and the local anisotropy. The CEF of $SrHo_2O_4$ is very complicated due to the presence of 4 Ho^{3+} ions on each of the two crystallographically inequivalent positions in a unit cell. Since Holmium is a non-Kramers ion, and the symmetry of the sites is quite low, a large number of CEF levels are expected. It was reasoned that the CEF levels associated with the one-dimensional short-range ordered system will shift up by the exchange-interaction energy as the temperature is lowered, while the position of the CEF levels that correspond to the $\mathbf{k} = 0$ short-range ordered structure would not change their position with temperature. By carefully studying the changes in CEF as a function of temperature it should possible to resolve some of the complex outstanding issues about the CEF for this material, and would explain why previous investigations have been insufficient to fully solve the CEF of $SrHo_2O_4$. Our previous experiment on IN4 has established that several of the CEF levels shift their position to higher energies with decreasing temperature.

The data for lower energies were collected using the IN5 instrument, with the lowest temperatures reached using a dilution refrigerator, in order to map out the temperature dependence of the CEF and see what changes occur as the temperature is lowered beyond the second short-range ordering temperature of ∼0.7 K. The some of the data are shown in Figs. $1-2$, and it is obvious that there are a large number of low energy CEF modes, a lot of which have a pronounced temperature dependence (*i.e.* some CEF levels do shift in energy with decreasing temperature). Another important observation about the CEF excitations in $SrHo_2O_4$ is that, as the temperature is lowered between 20 K and 1.6 K (the temperatures between which the one-dimensional scattering becomes strongly correlated), some of the CEF excitations become dispersive along Q. The minimum of the dispersion for this level corresponds to the location of the $(00\frac{1}{2})$ peak observed in the elastic scattering data.

The thermal occupation of the CEF levels changes, but from the temperature dependence it is possible to work out which inelastic features are enhanced by temperature and hence compensate for this in any CEF analysis. Also a new thermally excited mode at 0.12 meV, which may be related to transitions between CEF levels, only appears above 0.7 K as shown in Fig. 2, and thus can be excluded from the CEF scheme. At the lowest temperature of 0.05 K, the positions of the CEF levels in the $SrHo_2O_4$ system derived from inelastic neutron scattering measurements are 0.75, 0.94, 1.06, 1.53, 1.91, 2.71 and 3.53 meV; and several more possible levels at 0.55, 1.15 and 3.85 meV (these are small, and from powder data alone it is difficult to judge whether these shoulders are peaks in their own right). The detailed coverage of the temperature dependence of the inelastic spectrum has been used to separate out which CEF levels belong to which Ho^{3+} subsystem, and now it may be possible to try to fit the the CEF parameters for each subsystem separately. Currently work in underway on a theoretical interpretation of the CEF scheme for this material and the anomalous Q dependence behaviour that develops for some of the CEF levels, as well as the fitting of the contributions to the CEF from the two different Ho^{3+} sites separately.

[1] H. Karunadasa, Q. Huang, B. G. Ueland, J. W. Lynn, P. Schiffer, K. A. Regan, and R. J. Cava, Phys. Rev. B **71**, 144414 (2005).

[2] O. Young, A. R. Wildes, P. Manuel, B. Ouladdiaf, D. D. Khalyavin, G. Balakrishnan, and O. A. Petrenko, Phys. Rev. B **88**, 024411 (2013).

[3] O. Young, L. C. Chapon, and O. A. Petrenko, J. Phys.: Conf. Ser. **391**, 012081 (2012).

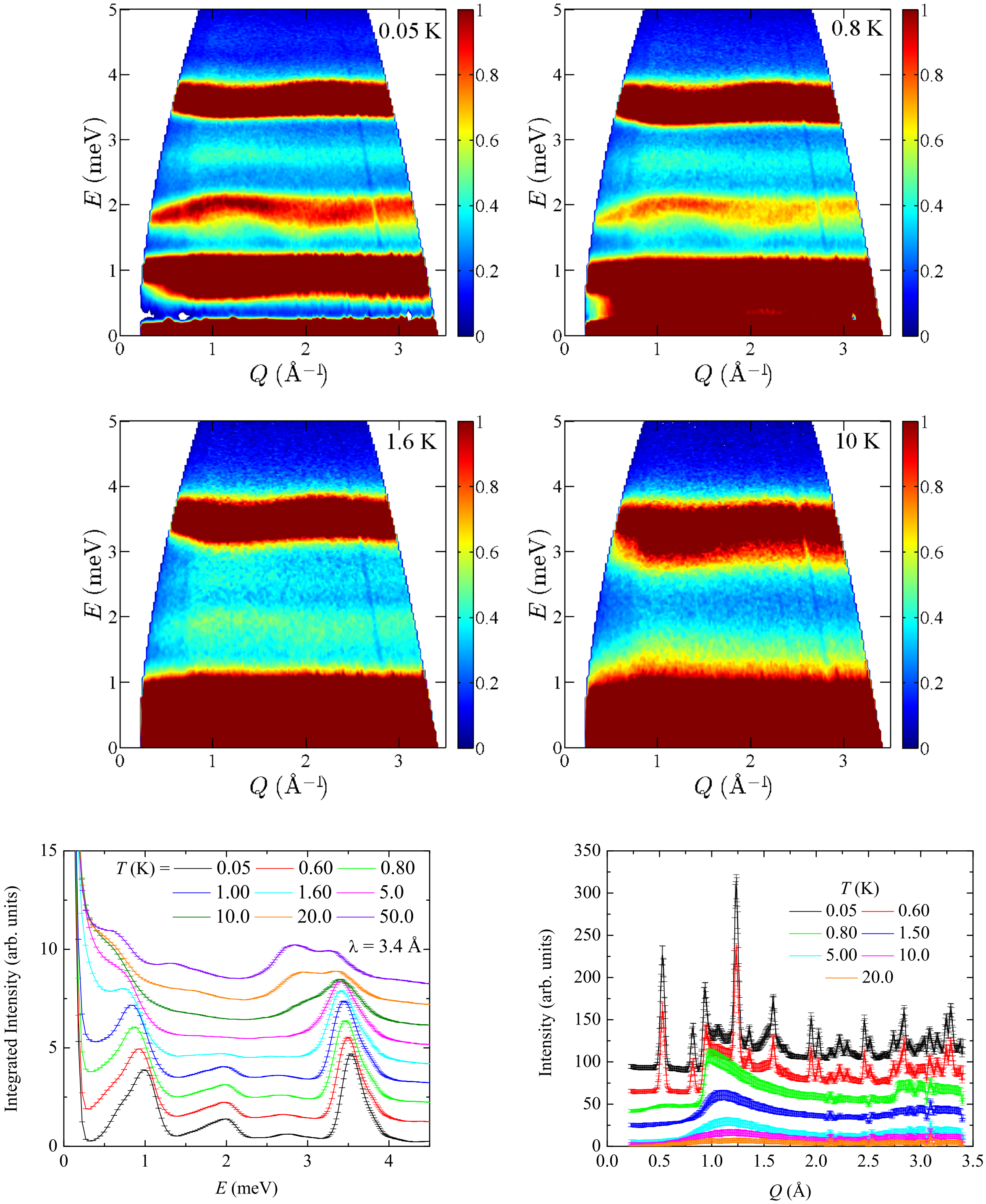

Figure 1: (Top, Middle) $S(Q,\omega)$ for $\lambda = 3.4$ Å at several temperatures. Note that not only do some of the CEF levels shift to higher energy with decreasing temperature, but there is also a change in the *dispersion* of some of the levels as well, for example the 3.53 meV crystal field level. (Bottom, left) Temperature dependence of the inelastic scattering from $SrHo_2O_4$, constructed from integrating over all of the $S(Q,\omega)$ data collected using the IN5 spectrometer for $\lambda = 3.4$ Å. The curves have been offset for clarity. (Bottom, right) Temperature evolution of the magnetic scattering of $SrHo_2O_4$ (curves have been offset for clarity), isolated by subtracting a 50 K background from the low temperature runs. Below 0.7 K resolution-limited **k** = 0 peaks can be seen which are directly comparable to the data collected using the GEM instrument on a powder sample of $SrHo_2O_4$ [3].

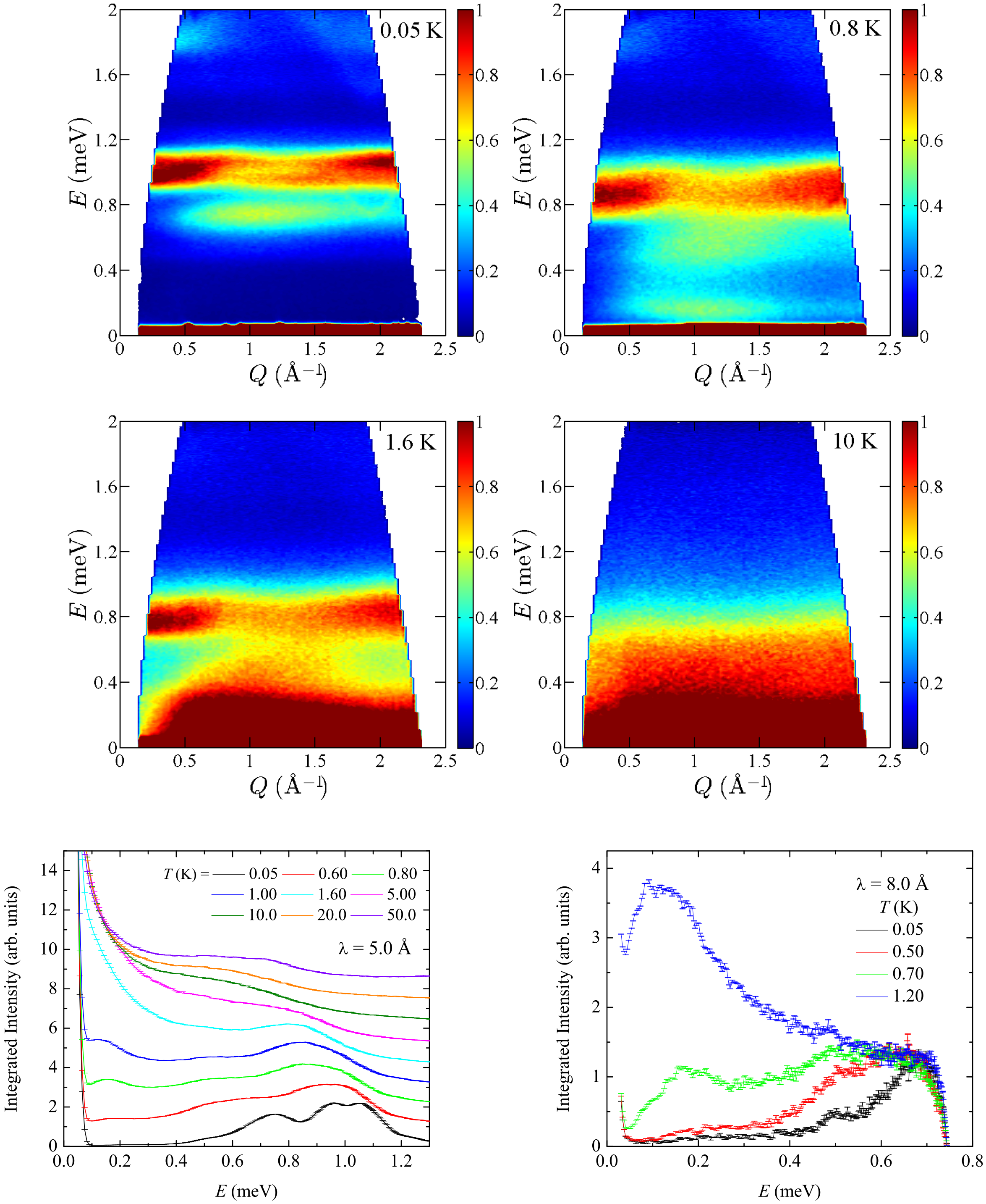

Figure 2: (Top, Middle) $S(Q,\omega)$ for $\lambda = 5.0$ Å at several temperatures. Again, the CEF levels shift and become dispersive upon changing the temperature, and new thermally excited features (that are not present at the lowest temperature) are also seen only above 0.7 K - the temperature at which the diffuse scattering is seen at $\mathbf{k} = 0$ positions. The data have been collected using the IN5 instrument. (Bottom, left) Temperature dependence of the inelastic scattering from $SrHo_2O_4$, constructed from integrating over all of the $S(Q,\omega)$ data collected using the IN5 spectrometer, for $\lambda = 5.0$ Å, the datasets for different temperatures have been offset for clarity. (Bottom, right) Data collected with $\lambda = 8.0$ Å to look at the thermally excited feature.