Magnetic structures in substituted $ErCo_2$ and $HoCo_2$ systems

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The magnetocaloric effect is seen as an avenue of creating more energetically efficient cooling systems. Recently much effort has been put into applying the magnetocaloric effect for hydrogen liquefaction. This has been achieved by using the base systems of $ErCo_2$ and $HoCo_2$ and substituting in small amounts other elements into the Co positions. [1] In addition to successfully creating a range of materials which could cover the temperature range, it was also able to change the order of the magnetic transition. This was seemingly done by suppressing the structural transitions of the cubic to rhombohedral, for $ErCo_2$, or cubic to tetragonal, for $HoCo_2$, which occur concurrently with the magnetic transition. It is not known what happens to the magnetic structure, and understanding it is key to developing and harnessing these and similar systems.

Neutron diffraction measurements were conducted on two groups of samples with using HERMES diffractometer, the $ErCo_2$ based samples, and the $HoCo_2$ based samples. Measurements on the base $ErCo_2$ sample indicated it was single phased, and the magneto-structural cubic to rhombohedral transition was successfully identified (See Fig. 1). The simple ferrimagnetic ordering along the c-axis and the moment sizes, although slightly lower for Er moments, agreed with previous reports. The Fe substituted samples contained a small impurity of magnetic $ErCo_3$. Despite the presence of the impurity, the transition still occurred at the expected temperatures of the substituted samples. The transition, however, was still a cubic to rhombohedral with similar magnetic moments to the base $ErCo_2$ sample. The Ni substituted sample did not have any impurity phases, and exhibited the rhombohedral phase, but with heavily reduced magnetic moment and lower transition temperature. Curiously, the only sample to hint at a different magnetic structure was a new sample of $ErCo_2$, which exhibited an additional reflection only at the transition temperature on the first cooling.

In the $HoCo_2$ based samples the cubic to tetragonal transition is observed for all samples. The resolution was unfortunately too low to detect the second low temperature tetragonal to orthorhombic transition. The moments aligned along the c-axis as a simple ferrimagnet and the moment sizes were generally very similar to the Er based samples. It should be noted that inclusions of non-magnetic Al, while increasing the magnetic ordering temperature, took significantly longer to approach saturation, and achieved a lower magnetic moment.

While not conclusive on its own, the data collected from these experiments is vital to understanding the underlying mechanism for the change in the magnetic transition type. The hints uncovered, such as increased intensity at low Q suggesting spin fluctuations, or the potential intermediate magnetic structure in $ErCo_2$, help understanding the origin of the phase transitions.

[1] X. Tang *et al.*, Nat. Commun. **13**, 1817 (2022).

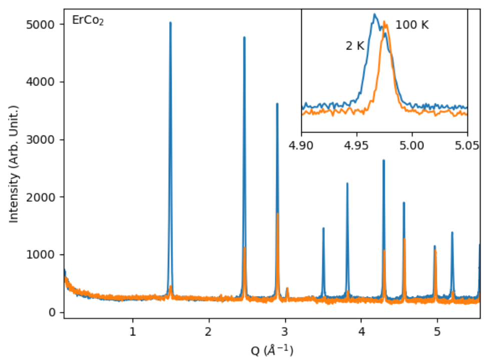

Fig. 1. Comparison of the diffraction patterns of $ErCo_2$ at 100 K (orange) and 2 K (blue). The inset highlights the broadening arising from the rhombohedral structure.