# Experimental report

29/09/2021

**Proposal:** **4-06-11** **Council:** 10/2019

**Title:** Study of a Novel Series of Isostructural Heterometallic 3d-4f Single Molecule Magnets with INS

**Research area:** Physics

**This proposal is a new proposal**


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**Samples:** [AlIII2ErIII2(µ3-OH)2(pmide)2(p-Me-PhCO2)6]·2MeCN
[FeIII2ErIII2(µ3-OH)2(pmide)2(p-Me-PhCO2)6]·2MeCN

| Instrument | Requested days | Allocated days | From | To |
| --- | --- | --- | --- | --- |
| IN6-SHARP | 6 | 3 | 14/09/2020 | 17/09/2020 |
| IN5 | 3 | 0 | | |


**Abstract:**

A major goal in tailoring single molecule magnets (SMMs) is to raise their blocking temperature, below which magnetic hysteresis on the molecular scale is observed, to technologically relevant temperatures. A crucial factor in the game is the magnetic anisotropy, and molecules containing both 3d and 4f metal ions are attractive candidates in this context.

Understanding the magnetic anisotropy due to the Lanthanide ions in such systems is however challenging. In a recent breakthrough, a new family of isostructual M2Ln2 butterfly compounds were synthesized, which for the first time allows us to replace the 3d Fe ions by diamagnetic Al ions in isostructural 3d-4f-SMMs, and therefore unprecedented means to study the Lanthanide's anisotropy in such SMMs. Here we propose to measure the Er-containing members of this new family on the instrument IN5 or IN6.

# 4-06-11 Study of a Novel Series of Isostructural Heterometallic 3d-4f Single Molecule Magnets with INS

**Motivation:**

The aim of this study is to complete the INS-dataset of a novel series of single-molecule-magnets (SMMs) to understand the interplay of magnetic anisotropy and exchange coupling between magnetic metal ions in SMMs, and their impact on their magnetic properties. This series of heterometallic SMMs includes both transition metal and rare earth ions. The rare earth ions have typically a very low symmetry ligand structure and therefore requires complicated magnetic models with many parameters (e.g. Stevens parameters) [4]. In the studied SMMs the rare earth $Er^{III}$ ions can be chemically exchanged by nonmagnetic $Y^{III}$ ions, which allowed us to directly measure the magnetism of the transition metal ions without the effects due to magnetic rare earth ions. In addition, and for the first time, the studied series of SMMs also permitted to chemically replace the transition metal ions, which thus allowed us to also directly measure the magnetism of the rare earth ions in the relevant low symmetry ligand field environment [1,2].

Our first experiment on this unique series of heterometallic SMMs provides us with insights into the role of the 4f electrons of $Dy^{III}$ ions for the magnetism [5]. With completing the dataset of this series of SMMs it is possible to investigate the difference of the effect of the $Dy^{III}$ ions and the $Er^{III}$ ions on the magnetism in this SMMs. Due to the fact that in [5] the studied SMM $Fe_2Y_2$ unfortunately turned out to be poly-phase, this compound is studied again.

**Experiment:**

In the experiments we recorded INS data on non-deuterated powder samples of the three isostructural compounds $Fe_2Y_2$, $Fe_2Er_2$ and $Al_2Er_2$ [3] $[M^{III}{}_2Ln^{III}{}_2(\mu_3\text{-}OH)_2(pmide)_2(p\text{-}Me\text{-}PhCO_2)_6]\cdot 2MeC$, where $Ln^{III}$ was either $Y^{III}$ or $Er^{III}$ and $M^{III}$ was either $Fe^{III}$ or $Al^{III}$, respectively (the samples were provided by the group of Prof. Annie K. Powell, University Karlsruhe, Germany). The sample weight of $Fe_2Y_2$ was 0.85g, of $Fe_2Er_2$ 0.74g and of $Al_2Er_2$ 1.06g. From the three compounds it is possible to distinguish the effects of the $Ln^{III}$-$Ln^{III}$ interactions, the interactions of the $Ln^{III}$ ions with the $M^{III}$ ions, and the effect of the $M^{III}$-$M^{III}$ interactions. Different neutron wavelengths and several temperatures were chosen to explore the excitations in these compounds.

For $Fe_2Er_2$ data was recorded at 4.1 Å (1.5 K, 5 K, 15 K and 30 K), 5.1 Å (1.5 K, 8 K, 15 K, 30 K and 50 K) (the 4.1 Å data are shown in Figure 1) and quite many magnetic transitions in the energy range between 1.3 and 2.6 meV were observed.

The $Al_2Er_2$ compound was measured at 5.1 Å (1.5 K and 30 K). The data at low temperatures provides a nearly perfect background for the $Fe_2Er_2$ measurement. Hints for an $Er^{III}$-$Er^{III}$ interaction were not observed (Figure 2Figure 2).

For $Fe_2Y_2$ data was taken at 4.1 Å (1.5 K, 50 K and 70 K).

The recorded data are of excellent quality and provide us a detailed view on the magnetic excitations in these compounds. IN6 was very suitable for the study and performed excellently. Within the allocated time, which was a bit too short, it was not possible to measure the $Al_2Er_2$ at 4.1 Å which would give us a better insight on the background, e.g. on the energies around 3.5 to 4 meV. This could be due to the remote access (due to lockdown and Covid19). This beamtime permitted us to record the important data. We also wish to emphasize the excellent support by the local contact.

Detailed analyses and quantum simulations to reveal the energy scheme of the $Fe_2Er_2$ and the $Fe_2Y_2$ compounds are underway.

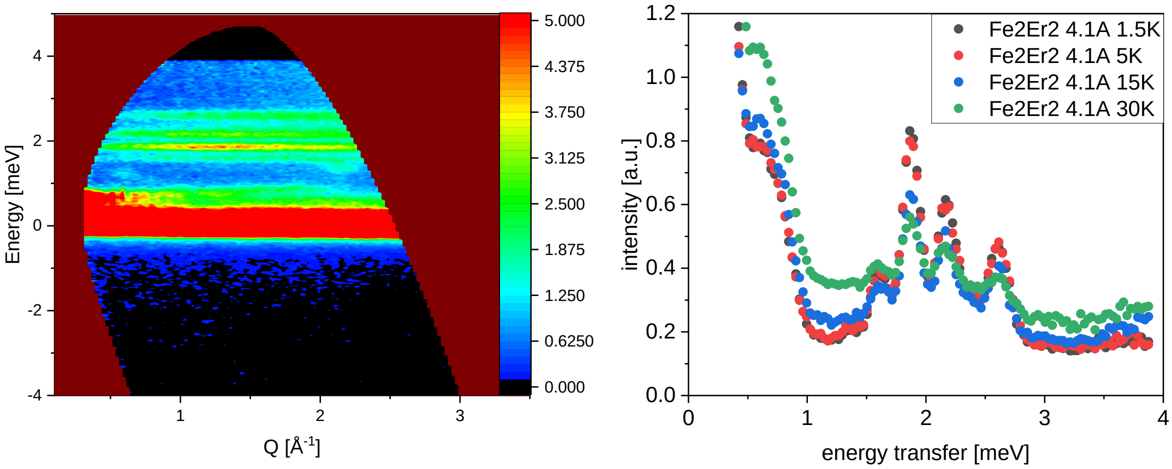


*Figure 1: $Fe_2Er_2$: S(q,w)-plot for 4.1 Å at 1.5 K (left) and temperature dependence of peaks (right)*

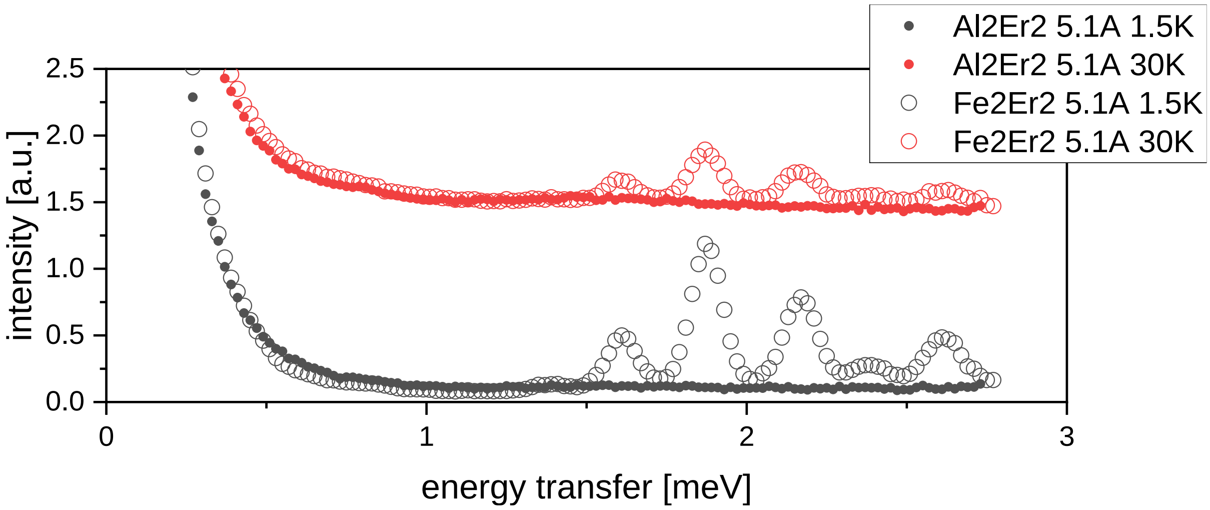


*Figure 2: Data of $Fe_2Er_2$ and $Al_2Er_2$ at 5.1 Å at temperatures 1.5 K and 30 K*


References:

[1] Y. Peng, V. Mereacre, C. E. Anson, A. K. Powell, ACS Omega 2018, **3**, 6360-6368 (2018)

[2] Y. Peng, V. Meracre, C.E. Anson, A.K. Powell, Phys. Chem. Chem. Phys., **18** 21469 (2016)

[3] A. Furrer, O. Waldmann, Rev. Mod. Phys. **85,** 367 (2013)

[4] K. Prša, J. Nehrkorn, J.F. Corbey, W.J. Evans, S. Demir, J.R. Long, T. Guidi, O. Waldmann, Magnetochemistry, **2**, 45 (2016)

[5] J.Mutschler, O.Waldmann, ILL - IN5 - Proposal 4-04-9 (2019)