ESRF	**Experiment title:** X-ray magnetic circular dichroism at the rare earth $L_{2,3}$ edges in ferromagnetic rare earth nitrides	**Experiment number**: HE-3437
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Shifts: 18	**Local contact(s)**: Fabrice Wilhelm	*Received at ESRF:*

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Report:

Origin of the proposal

This $L_{2,3}$ XMCD experiment aimed to clarify the magnetic state of semiconducting rare earth nitrides RN. Our focus in 2010 was on the peculiar compounds SmN and EuN, retaining GdN as a reference. The study complemented a previous XMCD experiment was carried out in 2009 with the ESRF beamline ID08 at the rare earth $M_{4,5}$ edges. The L edge spectra performed under the 2010 ID12 experiments access the induced 5*d* moment so that the XMCD signal probes the polarisation in that orbital. The exchange mechanism in these compounds is however still a matter of debate, so a comparison across the RE series, with data on both RE edges, provides important insight into the exchange process. Moments on the RE 5*d* and N 2*p* (also proposed ID08 experiment in 2009) shells are particularly important in this regard.

SmN is ferromagnetic with a near-zero moment due to the cancellation of the 4*f* spin and orbital moments for Sm^{3+}, as is demonstrated on the XMCD spectra at low temperature.

In **EuN** the Eu^{3+} total angular momentum is zero (J=0), for which an applied field induces Van Vleck paramagnetic behaviour. However magnetisation measurements on EuN show in addition a Curie-like paramagnetic behaviour at very low temperature. The $M_{4,5}$ XMCD data from 2009 had already revealed the coexistence of both Eu^{3+} and Eu^{2+} components. The Eu^{3+} 4*f* contribution is temperature independent as expected, while the Eu^{2+} 4*f* contribution shows a Curie temperature dependence as seen also in magnetisation studies.

The analysis of the $L_{2,3}$ spectra needs modelling which includes band structure effects, due to the more extended 5d empty state orbitals. This part is carried out by V. Antonov, who already achieved such calculations for GdN.

Run of the experiments and main results

The films used for the study were thinner than the L-edge x-ray extinction length, permitting the use of total fluorescence yield. Full XAS and XMCD spectra were measured at different temperatures above and below the Curie temperature, in the maximum applied field. Magnetisation loops were measured by scanning the field at a fixed energy corresponding to the maximum peak of the dichroic difference spectra.

A first experiment was run on a reference sample of ferromagnetic GdN, and a magnetisation loop was well reproduced at 7K in a maximum applied field of 6T by monitoring the field-dependent peak in the L_2 XMCD spectrum.

For **SmN** a large XMCD response was measured at 7K and 15K, while at 33K above the Curie temperature the paramagnetic signal dropped in intensity. Analysis of these spectra will be held back for further measurements.

The **EuN** results were supported by a calculation by V. Antonov, permitting the separation of both the XAS and the XMCD contributions from Eu^{2+} and Eu^{3+}. We found a Eu^{2+} signal that varied with temperature as expected for a paramagnetic ion. In contrast the Eu^{3+} signal showed both a temperature-independent van Vleck contribution and an additional signal that tracked the Eu^{2+} magnetisation. Clearly there is a strong Eu^{2+}-driven polarization of Eu^{3+} ions; the average Eu^{3+} 5*d* polarization below 10 K is more than doubled by the inter-ion *d-d* exchange from neighboring Eu^{2+} ions. Recalling that the *L*-edge XMCD spectra reflect the 5*d* polarization, the data indicate that an excess 5*d* polarization on Eu^{3+} ions is induced by polarized Eu^{2+} in the network and it is in the same sense as the polarization determined by the ferromagnetic intra-ionic *f -d* exchange on the Eu^{3+} ion itself. Not surprisingly, that *d*-orbital polarization is too weak to develop any $J = 1$ admixture on the Eu^{3+} ions, as is demonstrated by the lack of a temperature dependence in the trivalent XMCD signal at the *M* edge data collected on ID08 in 2009.

Publication

The complementary XAS/XMCD results from the four Eu edges, $M_{4,5}$ (ID08, HE2986) and $L_{2,3}$ (ID12, this report) generated an important paper identifying the magnetic state and inter-ion exchange in EuN.

Magnetic state of EuN: X-ray magnetic circular dichroism at the Eu $M_{4,5}$ and $L_{2,3}$ absorption edges, B. J. Ruck, H. J. Trodahl, J. H. Richter, J. C. Cezar, F.Wilhelm, A. Rogalev, V. N. Antonov, Binh Do Le, and C. Meyer, Phys. Rev. B **83**, 174404 (2011).

Abstract:

We report $M_{4,5}$- and $L_{2,3}$-edge x-ray magnetic circular dichroism (XMCD) studies of the ion-specific magnetic polarization on Eu in europium mononitride. The absorption spectra at both edges show a main signal originating from Eu^{3+} with a well-separated small (~2%) contribution from a Eu^{2+} valence state that we propose is associated with a small concentration of N vacancies. Magnetic polarization is observed in both 4*f* and 5*d* orbitals and for both valence states. The temperature and field dependence of the $M_{4,5}$-edge XMCD signal shows a van Vleck signature from Eu^{3+} (main EuN contribution) and a Brillouin paramagnetic signal from the small concentration of Eu^{2+} ions. In addition, the $L_{2,3}$ edge shows that the Eu^{3+} ions have 5*d* orbitals that are polarized both by intra-ion *f -d* exchange and by inter-ionic *d-d* exchange from neighboring Eu^{2+} ions. There is no evidence of magnetic order among the Eu ions at temperatures as low as 10 K.