

**Experiment title:**Impact of Aluminium on the magnetic properties of  $RFe_2$  and  $RCo_2$  through the modification of the electronic structure**Experiment****number:****25-01-676**

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The exact nature of the induced magnetic moments remains an open key problem into the understanding of the magnetic interactions in many magnetic systems. This is the case of the spin-polarization of the 5d states of the rare-earth in R-T intermetallic systems (where R denotes a lanthanide and T a 3d transition metal). The Laves phase compounds  $RFe_2$  and  $RAI_2$  exhibit ferromagnetic ordering. However, while the  $RAI_2$  systems show ferromagnetism (FM) below 150 K, the magnetic ordering temperature varies between 550 and 650 K through the  $RFe_2$  series. It is thought that the FM order in the  $RAI_2$  compounds is exclusively due to the rare-earth ions within a RKKY interaction framework. By contrast, the magnetism of the  $RFe_2$  results from the competition of the localized 4f magnetism of the rare-earth and the itinerant magnetism of the transition metal. The exact nature of the R-T interaction is not well accounted a priori by any theoretical model and only phenomenological descriptions addressing the critical role of the R(5d)-T(3d) hybridization have been reported. The problem of characterizing the 5d states and of determining the exact nature of the R-T interaction becomes critical in the case of the  $R(Fe_{1-x}Al_x)_2$  compounds. In this series we found systems evolving from pure RKKY grounds like the  $RAI_2$  series to pure Campbell's-like magnetism ( $RFe_2$ ) as a function of the Fe-Al dilution. However, while the magnetic characterization of the rare earth R(5d) states has been considered as fundamental into determining the magnetic properties of the intermetallic R-Fe systems, the same R(5d) states are no considered when the R-Al systems are studied.

This scenario indicates that the magnetism of the 5d states is nor marginal but crucial into determining the magnetic properties of many magnetic systems regarding both basic and applied magnetism. This experiment was aimed to get a

deeper insight on the exact nature of the induced magnetic moments at the delocalised states. In particular, we face the problem of determining the mechanism that induces the magnetic moment on the 5d states when two different localized moments are present in the same material, as in the case of R-Fe intermetallic compounds. Aimed to shed light on this debate, we have tailored a new series of compounds in which the polarization of these 5d states due to the rare-earth and to the transition-metal is varied in a controlled way. In the case of  $(\text{Ho}_x\text{Lu}_{1-x})\text{F}_2$  the magnetization of the rare-earth sublattice is progressively depleted by increasing the substitution of Ho by Lu. Therefore, the polarization of the Lu 5d states due to the Fe 3d ones is maintained fixed, while that of the Ho 4f states decreases. By contrast, in the case of  $(\text{Ho}_{0.5}\text{Lu}_{0.5})(\text{Fe}_{1-x}\text{Al}_x)_2$  the polarization of the Lu 5d states due to Ho is fixed, while that due to the transition metal is modified by substituting Fe for the non-magnetic Al. We have performed a detailed x-ray absorption spectroscopy (XAS) study of these compounds at the Fe K-edge and at the  $L_1$  and  $L_3$ -edges of the rare-earth. Due to the element and shell selectivity of the XAS technique and the extreme sensitivity of the near-edge region of the absorption spectrum to the modification of the DOS, it is possible to disentangling the structural and electronic effects induced by the different substitutions.

Our results show that the effect of substitution on the magnetic properties of the systems is markedly different whether the magnetic rare-earth (R) is substituted by a non-magnetic one (Y, Lu), or if the transition metal is substituted by Al. The Lu substitution acts as a simple dilution effect of the rare-earth sublattice magnetization and it has little influence in the magnetic ordering temperatures. By contrast, when Al replaces Fe the ferromagnetic behaviour of the material is seriously affected leading to the breakdown of the long magnetic ordering and to the appearance of spin-glass behaviour (probably associated to the existence of magnetic clusters with different sizes and different magnetic responses). The present XAS study at different absorption edges provides the disentangling of the structural and electronic changes induced by the different substitutions. Hence, further work is planned to determine the way in which this substitution modifies the R(5d)-T(3d) hybridization which governs the R-T interaction and, consequently, the magnetic properties of the systems under study. The expected results should gain insight into the origin of induced magnetic polarization in the 5d states and to assess the importance of RKKY interactions and R(4f)-R(5d)-Fe(3d) hybridisation in mediating the magnetic coupling