



	Experiment title: Magnetic exchange coupling in RCo ₂ Pn ₂ (R = Eu, Pr; Pn = P, As) pnictides	Experiment number: HC-2989
Beamline: ID12	Date of experiment: from: 26/01/2018 to: 30/01/2018	Date of report: 27/02/2018
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Report:

This project is intended to investigate the rare-earth cobalt pnictides RCo₂Pn₂ (R=Eu, Pr; Pn = P, As) that show a distinct interplay between local and itinerant magnetism resulting in multiple magnetic transitions, highly sensitive to perturbations of electronic filling of the 3*d* subband and in the FM state more or less governed by a Stoner criterion. The character of the coupling between the local 4*f* and the itinerant 3*d* magnetic moments and the magnetic properties of rare-earth cobalt pnictides are often assumed to be mediated by the type and position of the pnictogen atom in the lattice and this project also aims to uncover the role of pnictogen (P, As) in the magnetic exchange.

In the focus of this work are two compounds, the first is the phosphide Pr_{0.8}Eu_{0.2}Co₂P₂ with T_C = 282 K. In this compound Eu introduced into the crystal structure of parent AFM phase PrCo₂P₂ experiences strong chemical compression that perturbs its oxidation state and the magnetism by altering the filling of the 3*d* subband. The second sample is the arsenide EuCo₂As₂ in which the similar effect is induced by the application of external pressure, thus the FM state with T_C ~ 125 K was confirmed for the high pressure phase (above 4.7 GPa). The lower T_C indicates that the coupling between cobalt and rare-earth magnetic moments in the arsenides might be enhanced compared to the phosphides. It was decided to explore directly the coupling between different sublattices in pnictides using the X-ray magnetic circular dichroism.

Due to the technical difficulties the beamtime was split in two parts, the first part is an ambient pressure field- and temperature-dependent XMCD on the sample Pr_{0.8}Eu_{0.2}Co₂P₂. The second is a high pressure XMCD on the arsenide EuCo₂As₂; it will be carried out later when the respective setup is ready at ID12 beamline. Here we report the results of the first part.

XMCD measurements on Pr_{0.8}Eu_{0.2}Co₂P₂ compound

XMCD measurements were carried out at ambient pressure on FM compound Pr_{0.8}Eu_{0.2}Co₂P₂, showing a nice example of rich mutual behavior of different magnetic sublattices. Polycrystalline powder was mixed with black C and pressed into pellet. The XMCD experiments were carried out in the 17 T end station, using Si photodiode as fluorescence detector.

For Pr_{0.8}Eu_{0.2}Co₂P₂, XMCD spectra were measured at Eu *L*₃ and *L*₂, Pr *L*₃ and *L*₂, Co *K* and P *K* edges at temperatures 2, 20, 100, 200 K and the magnetic fields up to 17 T. Sizable XMCD signals were detected at all absorption edges, including P *K* edge. Thus, the P states are polarized and might participate in the magnetic interaction.

Eu XANES and XMCD spectra at *L*_{2,3} absorption edges indicate an Eu mixed valence state. And while in the XANES the ratio of Eu²⁺ and Eu³⁺ contributions is closer to 3/2, in XMCD it is approximately 3/1, showing the difference of mechanism defining the oscillator strength in case of X-ray absorption and circular magnetic dichroism.

The field dependence of the XMCD was recorded for Eu *L*₃, Pr *L*₂, Co *K* and P *K* absorption edges, Fig.1. The field dependence of the Eu magnetization shows a trend similar to the FM state, however, strongly

reminiscent of superparamagnetism. After normalization to the same intensity, the field dependences of XMCD at the Eu^{2+} and Eu^{3+} peaks have quite similar shape. This tells us that the $\text{Eu } L_{2,3}$ XMCD at the peaks corresponding to both europium valence states really show the same thing: the polarization of $\text{Eu } 5d$ band, which must be induced by the $4f$ electrons of magnetic Eu^{2+} configuration.

At the same time the dependences for the Pr, Co and P sublattices show a more complex behavior, without saturation and highly similar. For this reason, we unite Pr, Co and P into one sublattice. It seems that the orientation of magnetic moment of this sublattice is defined by the competing influence of Eu (Eu^{2+} ions have a largest magnetic moment in the system) and external magnetic field. Apparently, depending on the field, the mutual magnetization orientation of Eu and Pr-Co-P sublattices can be parallel or antiparallel. The field strength at which switching between these two states occurs is ~ 1.5 T.

Another interesting observation is that the magnetization dependences of the Pr, Co and P start to twist in a peculiar way when increasing the magnetic field higher than 4 T. This effect was noticed only at the lowest temperature in experiment, 2 K, while at higher temperatures the shape of the curves is rather linear. The effect might be similar to the observed in the total magnetization curve for this sample at the low temperature. This allows us to assume that the magnetization of different sublattices in reality might be not collinear and in certain conditions undergoes the canting.

The differences between Eu and Pr-Co-P sublattices indicate that $\text{Pr}_{0.8}\text{Eu}_{0.2}\text{Co}_2\text{P}_2$ is not a simple FM at low temperatures and represents rather a strong ferrimagnetic or complex mutual superparamagnetic behavior. The XMCD results for $\text{Pr}_{0.8}\text{Eu}_{0.2}\text{Co}_2\text{P}_2$ are currently under careful investigation. The next part of allocated beamtime will be spent on the XMCD study of the compound EuCo_2As_2 at high pressure using diamond anvil cell. Similar set of field and temperature-dependent measurements at different absorption edges will be performed in order to characterize the mutual behavior of different magnetic sublattices in this arsenide. The comparison to between two cases with different pnictogen (P, As) will help greatly to understand the forces driving the magnetism in rare-earth cobalt pnictide ferrimagnets.

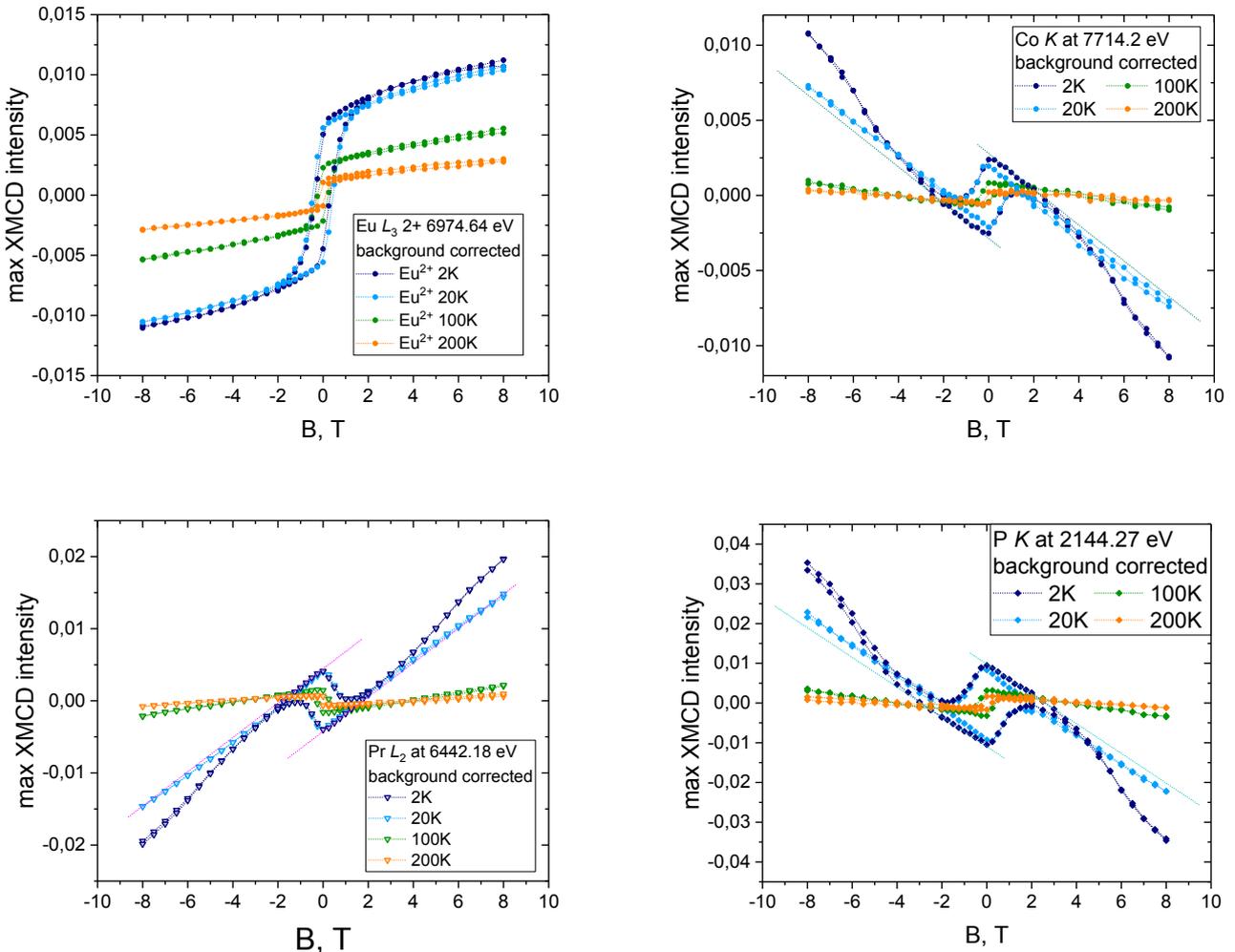


Fig 1. XMCD vs B curves for $\text{Pr}_{0.8}\text{Eu}_{0.2}\text{Co}_2\text{P}_2$ at different temperatures