Experimental report

20/11/2018

Proposal: **5-31-2537** **Council:** 4/2017

Title: Frustrated magnetism in Eu-based intermetallic systems

Research area: Physics

This proposal is a new proposal

Main proposer: **Diego Gaspar FRANCO**

Experimental team: Gabriel Julio CUELLO
Diego Gaspar FRANCO
Silvia SEIRO

Local contacts: Gabriel Julio CUELLO
Henry FISCHER

Samples: EuPtSi
EuPtGe
EuIr2P2

Instrument	Requested days	Allocated days	From	To
D4	10	6	12/06/2018	18/06/2018

Abstract:

Intermetallic compounds EuIr2P2, EuPtSi and EuPtGe order antiferromagnetically at TN=5, 4.1 and 3.3 K, respectively, although positive Curie-Weiss temperatures indicate predominantly ferromagnetic interactions. In addition, substantial magnetic fluctuations subsist well above TN, as attested by specific heat measurements on all three compounds. These observations point to a certain degree of magnetic frustration as a consequence of competing magnetic interactions, and in the case of the equiatomic compounds, favored by the particular lattice geometry. Furthermore, in all three materials, the absence of inversion symmetry in the crystal structure favors chiral magnetism. Neutron powder diffraction will allow us to determine the magnetic structure of these intermetallic compounds, and characterize its temperature evolution. This is an essential step to understanding the interplay between the spin lattice geometry, the chirality of the crystal structure and the competition of different exchange interactions in these systems.

Experimental report for Proposal nº 5-31-2537: Frustrated magnetism in Eu-based intermetallic systems.

Frustration in magnetic systems is a topic of strong current interest, since it can result in very unusual ground states with intriguing properties, as e.g. spin liquids, spin ice, or magnetic monopole-like excitations. The vast majority of studies on frustration in magnetic systems have been performed on insulating spin systems. In contrast, frustrated metallic systems remain largely unexplored, although interesting effects may be expected from the interplay between itinerant electrons and frustrated magnetic degrees of freedom. Besides, the presence of competing magnetic interactions, typical in frustrated systems, in a chiral magnet can result in the formation of a skyrmion lattice for certain temperature and magnetic field regions, as for example in MnSi [1].

We proposed to study the magnetic structure of three Eu-based intermetallic compounds: EuPtSi, EuPtGe[2]and $EuIr_2P_2$. These materials crystallize in a chiral space group, $P2_13$ for the 111 materials and $P3_221$ for the 122 compound. Previous bulk magnetization and specific heat measurements show unusual magnetic states and important frustration effects. Besides, an A-phase was found recently in the field – temperature phase diagram in EuPtGe, similar to the skyrmion system MnSi [3].

We performed the measurements on the D4 diffractometer, which allows the use of short wavelengths, essential in our samples due to the presence of the strong neutron absorbing element europium. Several Intensity vs. Q (or 2Θ) scans were measured at 0.6958 Å for EuPtGe and $EuIr_2P_2$ polycrystalline samples, mounted on vanadium cans. Besides, we also measured a nickel sample, the empty cryostat, a

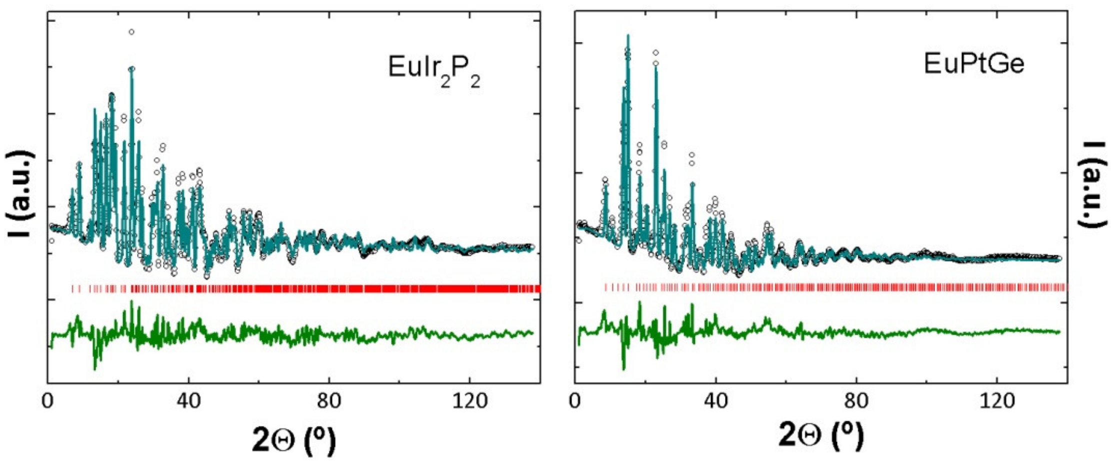

Figure 1. Room temperature Rietveld refinements for $EuIr_2P_2$ and EuPtGe samples. Observed (black circles), calculated (dark cyan), difference (green bottom line) and Bragg positions (red vertical bars)

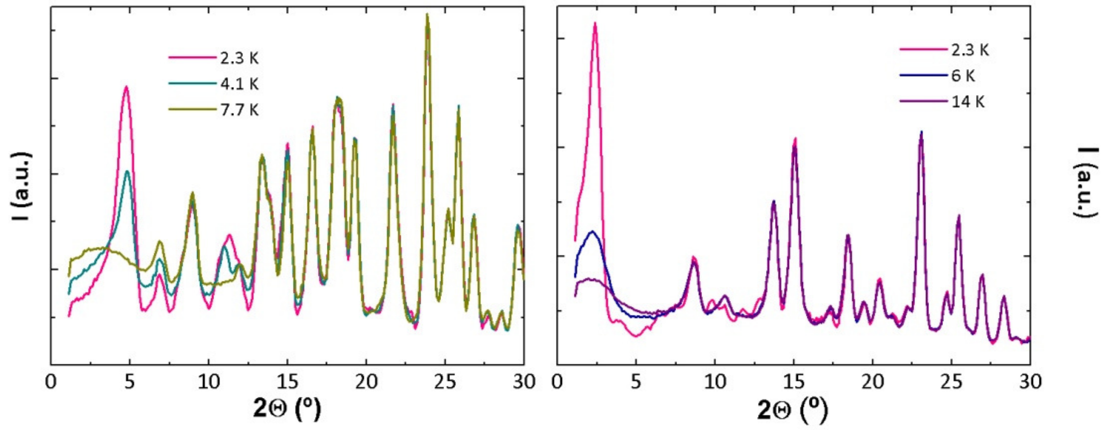

Figure 2. Selected low temperature diffractograms for $EuIr_2P_2$ (left) and EuPtGe (right) samples, showing the appearance of magnetic peaks at low angles.

vanadium sample and a ^{10}B sample, in order to determine the instrument wavelength and to make the background subtraction and correction analysis. This is fundamental to get a reliable magnetic pair distribution function (mPDF). The sample EuPtSi was not measured because the approved measurement beam time was not long enough.

Room temperature diffractograms on both samples confirm the cubic $P2_13$ and trigonal $P3_221$ crystal structures for EuPtGe and $EuIr_2P_2$ compounds respectively, Figure 1. No crystal structural transition is observed down to the lowest measured temperature (2.3 K), but one has to take into account that D4 is a low resolution diffractometer, so a structural transition cannot be completely ruled out.

We focus now on the low temperature data. Previous measurements show that $EuIr_2P_2$ presents an antiferromagnetic transition at 5.2 K and a second transition at 2.9 K. On the other hand, EuPtGe presents a first-order-like antiferromagnetic transition at 3.2 K. We show in Figure 2 some selected diffractograms above the two transitions (7.7 K), in between (4.1 K) and below the lowest transition (2.3 K) for $EuIr_2P_2$, and above the transition (14 and 6 K) and below the transition (2.3 K) for EuPtGe.

Some common features are observed for both samples. First, a reduction in the background below 2Θ ~ 15º is observed when cooling down, together with the appearance of additional sharp peaks. This is consistent with paramagnetic scattering disappearing in the long-range magnetically-ordered state. For $EuIr_2P_2$two and three magnetic peaks are observed at 4.1 K and 2.3 K respectively, indicating some change in the magnetic structure between these two temperatures. Preliminary analysis of these peaks indicate a magnetic propagation vector (1/4 1/4 1/8) for 4.1 K and (0 1/4 1/4) at 2.3 K. For EuPtGe several peaks could be identified, which gives k = (1/3 0 0).

Some additional feature to mention is the presence of a broad hump above the transition temperatures, even up to 40-50 K in the EuPtGe case. This hump evolves to

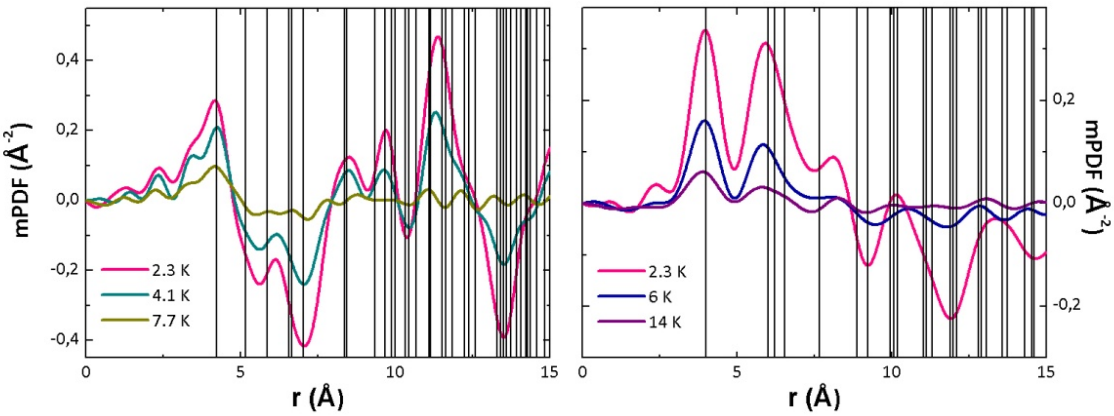

Figure 3. Magnetic pair distribution functions (mPDF) for $EuIr_2P_2$ (left) and EuPtGe (right) at several temperatures. The black vertical lines indicate Eu-Eu distances.

the first magnetic peak upon cooling. This large scattering in the paramagnetic regime could be an indication of short-range magnetic correlations. Long-range magnetic order occurs only at low temperatures, possibly as a result of frustration fostered by geometry and/or competing magnetic interactions.

Finally, we show in Figure 3 the magnetic pair distribution function (mPDF) obtained from the I_{mag} vs 2Θ data through a Fourier transform, where I_{mag} is the magnetic scattering. In order to obtain I_{mag} we subtracted the nuclear and paramagnetic contributions, taking as a reference the 45 K measurement. mPDF is a recently developed method to analyze long range magnetic structure but also short range correlations [4]. Positive peaks indicate ferromagnetic coupled ion pairs while negative peaks correspond to antiferromagnetically coupled ion pairs. Since susceptibility measurements indicate antiferromagnetic ordering [2], the striking result here is the sign of the observed first peak, which correlates with the first Eu-Eu distance for both materials. Contrary to the expected negative peak for an antiferromagnet, we get a clearly positive signal, which indicates a predominantly ferromagnetic nearest neighbor interaction. This suggests a complex non collinear magnetic structure. Further analysis of the data is underway in order to obtain a full description of the magnetic structure.

References.

[1] S. Mühlbauer, et al., Science **323** (2009) 915-919.

[2] D. Franco, et al,.Physical Review B **96** (2017) 014401.

[3] M. Kakihana, et al., J. of the Physical Society of Japan **87** (2018) 023701.

[4] B. Frandsen, et al., ActaCryst. A**70** (2014) 3.