Magnetic properties of $PuGa_3$

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Over the past two years researchers have discovered a wide variety of materials that not only lose their electrical resistance at low temperatures but open debates about the long thought on superconductivity and magnetism to be incompatible. With the discovery of superconductivity coexisting with ferromagnetism in UGe_2 under pressure [1] and at ambient pressure in URhGe [2], the recent announcement of superconductivity with unexpected critical parameters in plutonium-based materials $PuTGa_5$ [3,4], 5*f* materials seem to be a promising field for magnetically mediated superconductivity.

From the crystallographic point of view, there are strong family ties between these systems. In fact for all the compounds mentioned, except URhGe, the common building unit is the well-known cubic $AuCu_3$-type. Moreover, the binary $CeIn_3$ itself [5] was recently found to become superconducting at 250mK under 2GPa. Within the 1:1:5 ternary compounds superconductivity is also reported in cerium-based materials $CeMIn_5$, M = Co, Rh and Ir [6].

In this context, we started to (re) investigate the Pu-Ga binary system, and some preliminary results on $PuGa_3$ are reported here.

The binary phase diagram was reported in 1964 [7] showing two polymorhic transitions for $PuGa_3$. One at 400°C and another one at 922°C, whereas only two crystal structure determinations [7,8] were reported.

X-ray powder analyses of our $PuGa_3$ samples reveal that:

- annealed sample shows a hexagonal Ni_3Sn structure type (s.g. $P6_3/mmc$) with lattice parameters a = 0.6314(1) nm and c = 0.4523(1) nm. This structure is known as the close packed hexagonal structure DO19 with layering a-b-a-b plans;
- the as-cast and the water-quenched from 1020°C samples, were successfully refined with the rhombohedral (s.g. R-3m) structure reported in the literature[8]. The unit cell parameters thus obtained in the hexagonal setting are a = 0.6191(1) nm and c = 2.8089(1) nm.

In both cases, the X-ray powder and single crystal analyses confirm the crystal structures reported the literature [7,8]. However no magnetic measurements were reported on these two phases, although low temperature specific heat measurement of the DO19 phase were performed revealing a rather large γ value of 225 mJ mol^{-1} K^{-2} [9].

We investigated the magnetic properties of these phases by SQUID magnetometry.

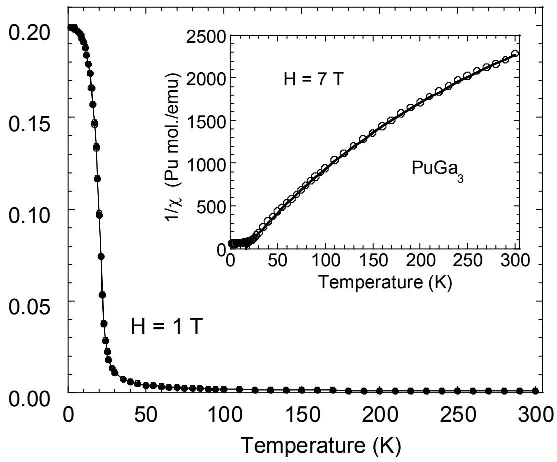

Fig. 1. Zero-field cooled (ZFC, open circles) and Field-cooled (FC, full circles) magnetization of $PuGa_3$ versus temperature at H=1T and inverse susceptibility at H=7T (circles, in insert) fitted by a modified Curie-Weiss law (dashed line).

Concerning the high temperature $PuGa_3$ phase, i.e. the rhombohedral structure type, the magnetization curve shows a ferromagnetic transition at $T_C \approx 20$ K (fig.

1). The zero-field-cooled and field-cooled curves superpose almost perfectly, even at relatively low magnetic fields, indicating that little energy is required to break the magnetic domains.

Figure 2 clearly shows that the magnetization is already close to saturation ($\mu_{sat} \approx 0.21\ \mu_B$) for fields as low as H = 0.25 T. From the hysteresis loop, a weak coercitive field $H_c \approx -0.08$ T is inferred.

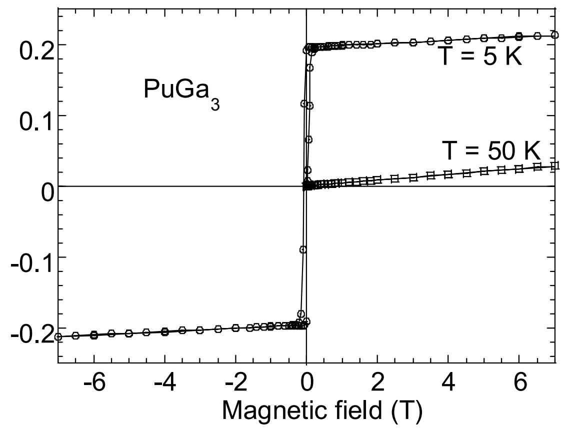

Fig. 2. Magnetization of $PuGa_3$ versus magnetic field at 5 K and 50 K.

In the paramagnetic state, the susceptibility obeys a modified Curie-Weiss law. The paramagnetic Curie temperature $\theta_p \approx 15.6$ K is positive as expected for a ferromagnet and close to the ordering temperature. The effective moment $\mu_{eff} \approx 0.77\ \mu_B$ corresponds to the value expected for Pu^{3+} (configuration $5f^5$) in Russel-Saunders coupling. The term $\chi_0 \approx 175.10^{-6}$ emu/mole is relatively modest, suggesting a limited hybridization of the 5f orbitals.

On the other hand, preliminary magnetic measurements of the DO19 phase suggest a quite different magnetic phase diagram with an antiferromagnetic transition at $T_N = 25$ K. The measurements is underway and will be presented at the conference.

As a conclusion, the two polymorphic phases of $PuGa_3$ were confirmed and obtained, and their magnetic property investigations started. The expected cubic $AuCu_3$ type phase for $PuGa_3$ was not observed in the present study. The study of the lower polymorphic transition reported at 420 °C is in progress. If such a phase could be obtained, it would be of very high interest to strengthen the family ties between the new class of superconducting 5*f* materials.

Acknowledgements
The high purity Pu metals required for the fabrication of the compounds was made available through a loan agreement between Lawrence Livermore National Laboratory and ITU, in the frame of a collaboration involving LLNL, Los Alamos National Laboratory and the US Department of Energy

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