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Specific heat of the antiferro / ferro-magnet NpGa₃

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The actinide compounds AnX₃, where X is an element from Group IIIA or IVA, crystallize in the cubic AuCu₃ structure. They are characterized by an actinide-actinide interatomic distance far above the Hill limit, therefore 5f–ligand hybridization is the main mechanism responsible for the delocalization of the 5f electrons. The systematic of this hybridization has been well demonstrated for the UX₃ compounds, which either do not order magnetically (X=Al, Si, Ge, Sn) or exhibit antiferromagnetism (X=Ga, In, Tl, Pb). It was concluded that the 5f–ligand hybridization increases as one moves up a column of the Periodic Table or moves from a Group IIIA element to a Group IVA element [1,2].

Although data on corresponding Np intermetallics are much less documented, a similar trend was noticed. However, it is clear that the Np-based compounds are more “magnetic” than their uranium analogues, which is consistent with the general picture that the hybridization decreases as one substitutes a heavier actinide. Indeed, all NpX₃ compounds (X=Al, Ga, In, Sn) order magnetically at the exception of NpGe₃ and NpSi₃ [3,4,5].

Several UX₃ systems (X=Al, Ga, In, Sn, Pb) present enhanced specific heat at low temperature, in particular USn₃ (γ = 170 mJ mol⁻¹ K⁻²) and UPb₃ (γ = 110 mJ mol⁻¹ K⁻²). In the neptunium analogues, only NpSn₃ (γ = 88 mJ mol⁻¹ K⁻²), NpIn₃ (γ = 72 mJ mol⁻¹ K⁻²) and NpGe₃ (γ = 34 mJ mol⁻¹ K⁻²) have been investigated [6].

In the present study, we focus on the specific heat properties of NpGa₃. It is worth noticing that UGa₃ is the only UX₃ compound considered to be itinerant that displays magnetic order [2]. NpGa₃ exhibits antiferromagnetic ordering ($k = (\frac{1}{2} \frac{1}{2} \frac{1}{2})$) below T_N ≈ 65 K, but ferromagnetic order with an ordered moment $\approx 1.5 \mu_B$ is stabilized below T_C ≈ 50 K [7]. The high pressure behavior of NpGa₃ indicates a weak delocalization of the 5f electrons [8].

Figure 1 shows the temperature dependence of the specific heat in NpGa₃. The room-temperature value corresponds to the Dulong-Petit limit (≈ 100 J mol⁻¹ K⁻²). Going down in temperature, we observe a lambda-type anomaly at 66 K that corresponds to the onset of antiferromagnetic ordering. At 51 K, an intense and narrow peak emerges from the curve, indicating the magnetic phase transition from antiferromagnetism to ferromagnetism. This sharp peak reveals the first-order nature of the transition, as previously pointed out by the thermal variation of the magnetic moment measured by Mössbauer spectroscopy and neutron diffraction [7]. At low-temperature, the extraction of the electronic specific heat is delicate, due to the magnetic contribution and the presence of an upturn, but the Sommerfeld coefficient can be roughly estimated to ~ 40 mJ mol⁻¹ K⁻², which is smaller than the γ values reported for the other ordered NpX₃ compounds and for UGa₃. This observation is consistent with the narrow-band picture suggested by high-pressure experiments [8]. Experiments in magnetic fields up to 14 T have also been performed and will be presented. The antiferromagnetic phase is destroyed above 4 T. The magnetic phase diagram was precisely rebuilt and essentially confirms the previously established one.

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