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LETTER TO THE EDITOR

Frustration of magnetic order in GdAl_3 (and in CeAl_3 ?)

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Abstract. Electron spin resonance measurements of $4f^7$ Gd moments confirm the suggestions of susceptibility measurements that the onset of antiferromagnetism in GdAl_3 is strongly frustrated. This raises the possibility that the isostructural heavy fermion compound CeAl_3 fails to order magnetically, unlike CeAl_2 , because frustration has depressed the temperature of long-range ordering.

It is well known that in certain types of crystal lattices the existence of nearest-neighbour antiferromagnetic interactions between magnetic moments on lattice sites can frustrate the onset of long-range antiferromagnetic order. The most obvious examples are two-dimensional triangular lattices or face-centred cubic lattices where nearest neighbours of one moment are nearest neighbours of one another and receive competing instructions on the direction in which they should point. In MnO , for example, an average strength of the exchange interaction is indicated to be about 500 K from a value of that order for $|\theta|$ in a high-temperature fit of the susceptibility to the Curie–Weiss expression $C/(T-\theta)$. Since the final onset of antiferromagnetic order, in a structure governed by the relative strengths of second-neighbour and first-neighbour interactions, is at 100 K, long-range order is said to be frustrated in the temperature range $\sim 100\text{--}500$ K, and only short-range order is seen in neutron scattering experiments at room temperature.

The intermetallic compound GdAl_3 with the Ni_3Sn structure (DO_{19}) has a Néel temperature that is surprisingly low (17 K) compared with the value of $|\theta|$ (~ 90 K) obtained from fitting the Curie–Weiss law ($\chi = C/(T-\theta)$) to its susceptibility above 20 K (Buschow and Fast 1966). It is also low compared with the strength of interactions in its neighbouring ferromagnetic Laves phase GdAl_2 ($T_c \approx \theta \approx 170$ K).

Such large ratios of $|\theta|/T_N$ have been noted (Taylor and Coles 1975, Fisk *et al* 1971) in a number of other Gd intermetallic compounds, and have been shown to correlate with deviations from the $a+bT$ form of temperature dependence for the linewidth of the electron spin resonance of the well known S-state Gd $4f^7$ resonance. In all such compounds the linewidth passes through a minimum at a temperature somewhat above $|\theta|$, broadening rapidly as T is reduced towards T_N . This pattern of behaviour was ascribed to the frustration that can arise, as pointed out above, for antiferromagnetic interactions in some geometries.

Figure 1 shows the linewidth data for GdAl_3 at X band (9.23 GHz) in a sample grown from an aluminium melt. (The GdAl_4 phase forms only at temperatures below the 650°C

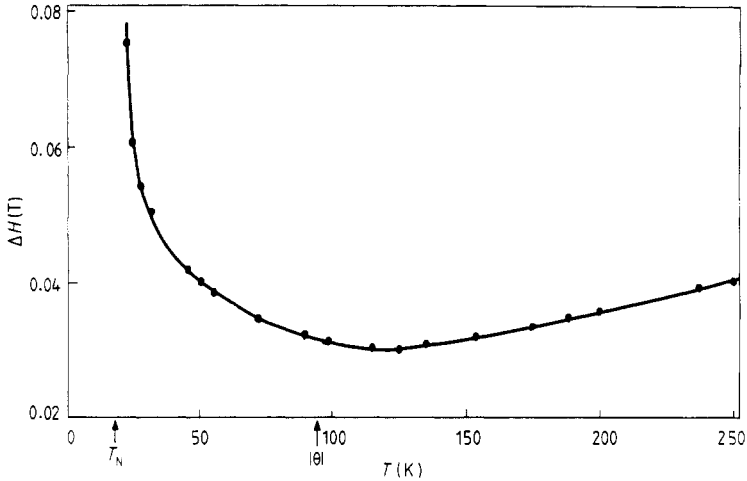


Figure 1. Linewidth at half maximum of the EPR signal from GdAl_3 at 9.23 GHz as a function of temperature.

eutectic and therefore does not prevent the growth of the XAl_3 phase here, as unfortunately the high peritectic temperature of $\text{Ce}_3\text{Al}_{11}$ prevents the growth of CeAl_3 by this technique.) At 300 K the g value of GdAl_3 was 2.00 ± 0.01 , remaining unchanged down to about 50 K, below which temperature it seems to increase, reaching 2.03 ± 0.03 at 30 K. However, the rapid increase in linewidth below 50 K, which has already begun below 100 K and which is firm evidence for frustration, makes the g value increasingly uncertain at lower temperatures.

The ESR results are thus in strong agreement with the suggestion that GdAl_3 possesses competing interactions which suppress long-range order from an expected onset temperature of ~ 100 K to the observed Néel temperature of 17 K. The apparent enhancement of μ_{eff} to $8.3 \mu_B$ may also be a consequence of short-range order effects below 100 K.

In the DO_{19} structure the Gd atoms, which form a hexagonal lattice, lie in close-packed planes in which they are separated from one another by Al atoms. The stacking of the planes is such that each atom has six Gd neighbours out of its plane at $\sim 4.4 \text{ \AA}$ and six other Gd neighbours within the plane at $\sim 6.3 \text{ \AA}$. For ferromagnetic interactions with the latter group the onset of order is clearly not frustrated, being simple ferromagnetic for ferromagnetic interactions with the former group and planar antiferromagnetic for antiferromagnetic interactions with them. If the 6.3 \AA interaction is antiferromagnetic, however, the result must be frustration whatever the sign of the 4.4 \AA interaction, and experiment indicates that this situation holds. It can be noted, without too much significance being attached to it, that for a free-electron Fermi surface with $k_F = 1.75 \times 10^8 \text{ cm}^{-1}$ both these separations correspond to negative values of the RKKY coupling, lying between the fifth and sixth and between the seventh and eighth zeros respectively, and give inevitable frustration of the in-plane interactions.

Current interest in the heavy fermion problem (Fisk *et al* 1986) makes these results of slightly more than passing interest, since CeAl_3 , isostructural with GdAl_3 , is one of the heavy fermion compounds that (unlike CeAl_2 in spite of its very similar value of μ_{eff}) fails to give a magnetically ordered ground state, but also fails (unlike CeCu_2Si_2) to become

superconducting. Thus, if the ratio $T_c(\text{GdAl}_2)/T_N(\text{GdAl}_3)$ is any guide, the long-range order that is found below 3.8 K in CeAl_2 could not be expected above about 0.35 K in CeAl_3 , by which temperature the Kondo compensation of the Ce moments is likely to have made a non-magnetic Kondo lattice more stable. The delicate question of how 'non-magnetic' this lattice has to be for a superconductive state to be stable is still not clear. It must also be borne in mind that the ordering of CeAl_2 is of an incommensurate character, and its Néel temperature may not be a good guide to what one would expect of CeAl_3 in the absence of frustration, especially if the Fermi surface geometry that leads to its characteristic ordering has been reconstructed (relative to that of GdAl_2) by its own f-conduction band hybridisation.

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