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Specific heat of UNiAl in high magnetic fields

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Specific heat measurements on UNiAl in magnetic fields up to 20 T demonstrate that the Néel temperature (T_N = 19 K) is drastically reduced by magnetic fields $B \parallel c$-axis. AF ordering disappears for $B > 11.35$ T, the field of the metamagnetic transition. A step in γ versus B (to $\simeq 250$ mJ/mol K^2) observed just above the critical field separates the low-field increasing tendency from the high-field decrease. At 20 T, a γ-value smaller than the zero-field value of 163 mJ/mol K^2 is found.

UNiAl is one of many UTX compounds with the hexagonal ZrNiAl structure. It orders antiferromagnetically at $T_N \simeq 19$ K. The high γ-value of 163 mJ/mol K^2 [1] and the estimated ordered U magnetic moment of $0.5\mu_B$ [2] indicate the itinerant nature of 5f magnetism. One of the salient features of UNiAl is the sharp metamagnetic transition, located at about 11 T [1]. Due to the enormous magnetic anisotropy, the transition is observable only for the field applied along the c-axis (the easy-magnetization direction). Here, we report on specific-heat measurements on an UNiAl single crystal in magnetic fields up to 20 T.

The magnetic phase transition of UNiAl from the paramagnetic to the antiferromagnetic state is expressed in the C/T versus T plot (fig. 1) by a peak at 19 K. The peak position is not affected when the field is applied perpendicular to the c-axis, though the specific heat above the transition is slightly enhanced [1]. Dramatic effects are observed, however, if the magnetic field is applied parallel to the c-axis. The magnetic-transition anomaly is gradually shifted to lower temperatures and becomes more pronounced with increasing magnetic field. The Néel temperature displays the usual quadratic dependence on the applied field. In fields $B = 11$ and 11.25 T the peak moves below 10 K and becomes much sharper, reminiscent of a first-order transition. In agreement with this, the magnetization curves measured at temperatures below 10 K show a metamagnetic transition of the first-order type. Thus a tricritical point is suggested to be expected in the B–T phase diagram. In fields $B \geq 11.5$ T no transition appears any more, instead, a low-temperature upturn in C/T versus T is recorded. This feature is gradually suppressed with further increasing field.

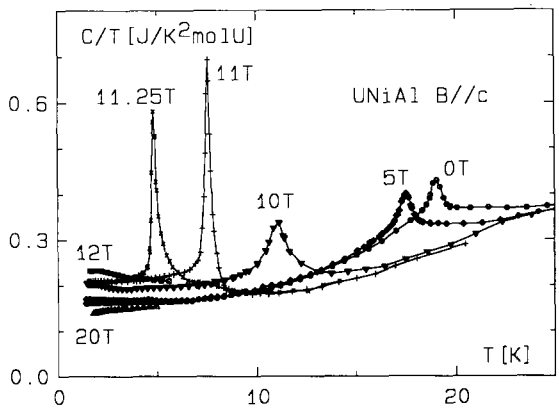

Fig. 1. Temperature dependence of the specific heat (C/T versus T plot) of UNiAl in various magnetic fields $B \parallel c$.

The effect of the magnetic field on the low-temperature part of the specific heat is illustrated in fig. 2. Since any extrapolation to zero temperature in the vicinity of a magnetic transition is somewhat doubtful, the C/T values at 2 K (instead of proper γ values) are plotted versus the applied magnetic field. Up to 11 T, C/T is increasing nearly quadratically with the field:

$$C/T = a + bB^2, \tag{1}$$

with $a = 162.0$ mJ/mol K^2 and $b = 4.1 \times 10^{-4}$ J/mol K^2 T^2. The smooth increase is followed by a sudden step between 11.25 and 11.5 T, and a subsequent gradual decrease. The value in 20 T (147 mJ/mol K^2) is already lower than that obtained in zero field. Similar effects have been found e.g. in U(Pt,Pd)$_3$ compounds [3].

The low-temperature susceptibility measured on a UNiAl single crystal [1] can also be described by a

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quadratic formula:

$$\chi = \chi(0) + cT^2, \tag{2}$$

with $\chi(0) = 140 \times 10^{-9}$ m^3/mol and $c = 2.8 \times 10^{-10}$ $m^3/mol\ K^2$. Taking into account this result, the field dependence of the specific-heat coefficient, γ, is qualitatively consistent with the thermodynamic relation [4]:

$$B(\partial^2\chi/\partial T^2) = \mu_0(\partial\gamma/\partial B), \tag{3}$$

where χ is the zero-field static susceptibility. The value of $b = 2.3 \times 10^{-4}$ $J/mol\ K^2\ T^2$, calculated by means of eq. (3) from the observed $\chi(T)$ dependence (2), is by a factor of two lower than the b value obtained from the specific-heat measurements. This discrepancy can be easily accounted for e.g. by a small experimental error ($< 1\%$) in the determination of γ. Also, it should be noted that eq. (3) is only valid if C/T remains a temperature-independent constant.

In the specific heat of UNiAl we probably deal with a complicated interplay of inter-site and on-site magnetic fluctuations. One of the directly accessible parameters, the field of the metamagnetic transition, presumably characterizes the strength of an AF coupling, i.e. also the energy scale of inter-site fluctuations. On the other hand, the on-site fluctuations persist in fields up to 50 T, preventing saturation in lower fields [1]. The degrees of freedom connected with the on-site fluctuations can lead to the high γ-value in zero field. The inter-site correlations are probably responsible for the high-temperature tail of the anomaly at T_N,

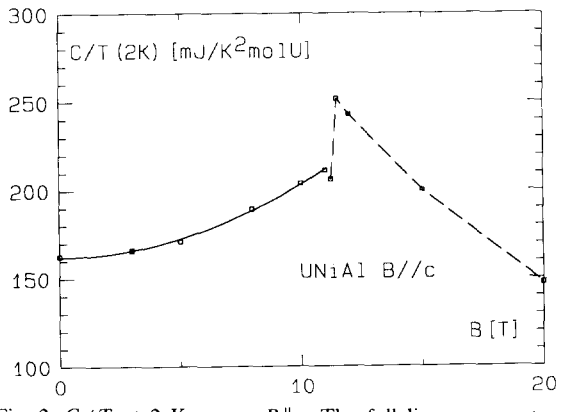

Fig. 2. C/T at 2 K versus $B \| c$. The full line represents a least-squares fit to $C/T = a + bB^2$ in the field interval 0–11 T. The dashed line is a guide to the eye.

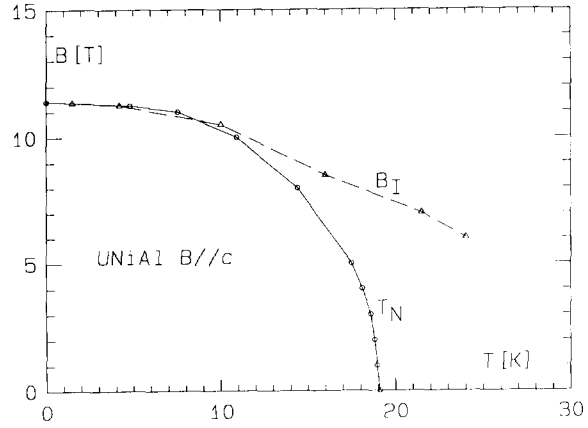

Fig. 3. Inflection points B_I of the magnetic isotherms and transition temperatures T_N versus magnetic field for $B \| c$. The lines are guides to the eye.

which is progressively suppressed by the magnetic field. The existence of inter-site antiferromagnetic spin fluctuations is documented also in the B–T magnetic phase diagram (fig. 3), where the branch obtained from inflection points of the S-shaped magnetization curves goes above the actual T_N values for $T > 10$ K.

The question remains whether the origin of the anomaly in γ at the metamagnetic transition is related to changes in the spectrum of fluctuations, or whether a description in terms of itinerant antiferromagnetism is more appropriate and the jump in γ must be seen as a manifestation of the closure of a gap at the Fermi energy.

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