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Electronic properties of UNiAl in high magnetic fields

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The ternary compound UNiAl, crystallizing in the ZrNiAl-type structure, is an itinerant antiferromagnet with magnetic moments of less than $0.8\mu_B$ per U atom and with a high γ value of 164 mJ/mol K^2 . The anisotropic response of the electronic properties to magnetic field is closely connected with the huge uniaxial magnetic anisotropy, observed not only in the magnetically ordered state, but also in the paramagnetic region. The origin of this anisotropy can be found in a strongly anisotropic hybridization. The magnetic ordering temperature, $T_N = 19.3 \text{ K}$, is reduced upon applying the field along the c axis of the hexagonal structure, which is the easy-magnetization direction. Antiferromagnetic ordering is suppressed by magnetic fields higher than the critical field of the metamagnetic transition, which is 11.35 T at 1.4 K . The electronic contribution to the specific heat is gradually enhanced by the magnetic field up to the transition, where a γ value of about 260 mJ/mol K^2 is recorded. In higher fields, the γ coefficient is gradually suppressed even below the zero-field value and reaches 143 mJ/mol K^2 at 20 T . The magnetic field applied in the basal plane has a negligible effect on both T_N and γ . The pronounced anisotropy is also found in the low-temperature electrical resistivity, which is considerably reduced in fields above the metamagnetic transition. Strong indications of anisotropic magnetic fluctuations can be traced in a number of experimental findings on pure UNiAl and also on Y-, Co-, or Fe-substituted samples, in which a fast decay of magnetic ordering is observed.

I. INTRODUCTION

UNiAl is one of the numerous UTX compounds (T = late transition metal, X = p metal, like Al, Ga, In, Sn, Sb) that are known to crystallize in the hexagonal ZrNiAl type of structure¹⁻⁵ (space group $P6_3/m$), which is schematically shown in Fig. 1. It is built up of two types of basal-plane layers, containing U-Ni and Ni-Al, alternating along the c axis. Each uranium atom has four nearest U neighbors within the U-Ni layer. The interuranium spacing $d_{U-U} = 349 \text{ pm}$, is related to the lattice parameter a (673.3 pm). The U-Ni layers are separated by the lattice parameter c (403.3 pm).

The magnetic properties of the UTX compounds³⁻⁷ vary from enhanced Pauli paramagnetism in UFeX through spin-fluctuation behavior and metamagnetism in URu(Al, Ga) and UCoAl to the ordering of magnetic moments growing in size up to $1.5\mu_B/\text{f.u.}$ and predominantly originating from the U atoms. UNiAl is one of the few antiferromagnetic compounds in this family.

UTX

Fe_2P (ZrNiAl) - type

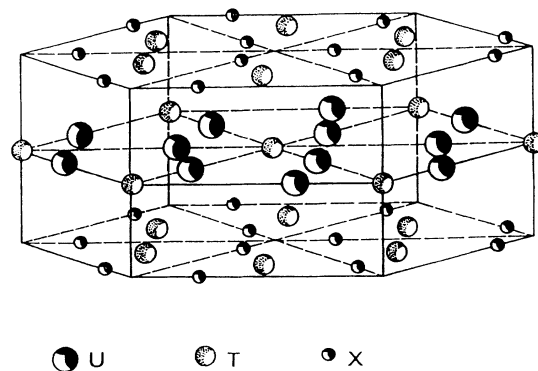


FIG. 1. Schematic picture of the UNiAl crystal structure.

The main features in the systematics of the ground-state properties in the UTX compounds can be summarized as follows. The tendency to magnetic ordering, and also the size of the uranium magnetic moments, increases with filling of the d band of the transition metal, as well as with increasing atomic volume of the X element, i.e., in the sequence Al, Ga, In, or Sn.⁷ These observations are consistent with the dominating tendencies of the $5f$ -ligand hybridization,⁸ which is an important mechanism causing delocalization of $5f$ electrons and thus disturbing the formation of uranium magnetic moments:

(a) The $5f$ - d hybridization is reduced with increasing difference in energies of uranium $5f$ and ligand d states. This comes into effect when moving towards the right end of the transition-metal series in which a more populated d band is pushed down in energy, while the $5f$ band of uranium remains pinned in the vicinity of E_F .

(b) The $5f$ - p hybridization with X ligands is weakened with increasing volume of the X atom (i.e., when moving down in the p -element column of the periodic table), which reduces the overlap of $5f$ and p wave functions.

These tendencies are confirmed for the whole UTAI series by self-consistent energy-band calculations,⁹ which also show that the Ni $3d$ band in UNiAl is shifted below E_F and does not contribute to the net magnetic moment.

The essential features of UNiAl, an antiferromagnetic ground state with a Néel temperature T_N of 19 K, a metamagnetic transition with a critical field B_C of about 11 T, and an enhanced value for the γ coefficient of the low-temperature specific heat of 164 mJ/mol K², were reported in early studies of polycrystalline samples.^{10,11} The last feature suggests an itinerant character of the magnetism, which can account for the small estimated magnetic moment of approximately $0.5\mu_B$ /f.u. A huge aniaxial magnetocrystalline anisotropy, which seems to be a common characteristic of all UTX compounds with the ZrNiAl type of structure, was found in a study of the bulk properties of single-crystalline samples.¹²

A preliminary analysis of neutron-diffraction experiments has revealed the following features of magnetic ordering in UNiAl.¹³ Indications for short-range order within the basal plane are found around 26 K (the temperature of a susceptibility maximum), whereas three-dimensional long-range antiferromagnetic order appears below 19.5 K. The magnetic phase transition at this temperature is of second order. The magnetic structure at 4.2 K is collinear, with uranium magnetic moments oriented along the c axis. It can be characterized by a propagation vector $\mathbf{q}=(0.1,0.1,0.5)$. The maximum value of the uranium magnetic moment is not precisely determined: it was estimated as $0.5\mu_B$, which is compatible with the value obtained from magnetization measurements above the metamagnetic transition.

In this paper, we review results of the studies of magnetic properties, resistivity, magnetoresistance, the magnetocaloric effect, and specific heat of UNiAl single crystals, with a special emphasis on the behavior in high magnetic fields. In Sec. VII, we also briefly summarize results of Y, Co, and Ni substitutions in UNiAl. The discussion in Sec. VIII leads us to a tentative model, with the anisotropic character of the spin fluctuations a key ingredient.

II. EXPERIMENT

Single crystals of UNiAl were grown by a modified Czochralski technique¹⁴ from a stoichiometric melt with a pulling speed of about 8 mm/h. The first crystal grown from a polycrystalline seed showed a mosaic structure in the x-ray Laue pattern, and therefore single-crystalline seeds were used in further trials. To reduce the mosaicity of the final product, the seed was tilted from the easy-growth direction by an angle of 15°. In this way, we obtained a crystal of about 30 mm length and a maximum diameter of 6 mm. The top and lower end were inspected by microprobe analysis. The detected composition $U_{33.3(4)}Ni_{35.9(4)}Al_{30.8(3)}$ agrees with the stoichiometric composition UNiAl within the experimental uncertainty. Al, the lightest element in the compound, is a limiting factor in the accuracy of the determination of the composition. Small inclusions of uranium oxide were detected on the crystal surface by the microprobe.

For the purpose of magnetic measurements with magnetic fields along various crystallographic directions, a spherical sample with 3 mm diameter was glued into a Stycast cube. The magnetic susceptibility was measured by means of a pendulum magnetometer in the temperature range 4.2–300 K in fields up to 1.3 T. Magnetization measurements at helium temperatures in magnetic fields up to 35 T were performed in the High-Field Installation of the University of Amsterdam¹⁵ by means of an induction technique.

Bar-shaped samples (typical dimension $4 \times 0.2 \times 0.3$ mm³), cut parallel and perpendicular to the c axis, were used for electrical-resistivity measurements with the current parallel and perpendicular to the c axis. Since the contacts were glued by silver paint on the rather small bars, the uncertainty in the absolute values of the specific resistivity may reach 10%. The resistivity in zero magnetic field was measured in the temperature interval 0.3–300 K using a standard four-point ac method. Magnetoresistance studies at 1.5, 4.2, and 77 K in magnetic fields up to 35 T were done also in the Amsterdam High-Field Installation. The resistance was measured using a dc technique. As the sample is immersed in the cryogenic liquid in this experimental setup, higher currents can be applied without any danger of heating. In the case of UNiAl, currents up to 200 mA were used. More accurate magnetoresistance measurements were performed in a superconducting solenoid in the low-field range ($B < 14$ T) at 4.2 K (sample immersed in liquid He).

The specific heat was measured on a single crystal of about 2 g by a standard semiadiabatic method with the calorimeter inserted in a superconducting coil providing fields up to 5 T. Measurements in fields up to 20 T were performed on the same sample at the Nijmegen High-Field Laboratory using a Bitter magnet.

III. MAGNETIC MEASUREMENTS

The temperature dependence of the magnetic susceptibility of UNiAl, measured with the magnetic field parallel and perpendicular to the c axis, is shown in Fig. 2. The susceptibility χ^{\parallel} (measured with $B \parallel c$) exhibits a pronounced and round maximum around 25 K. This tem-

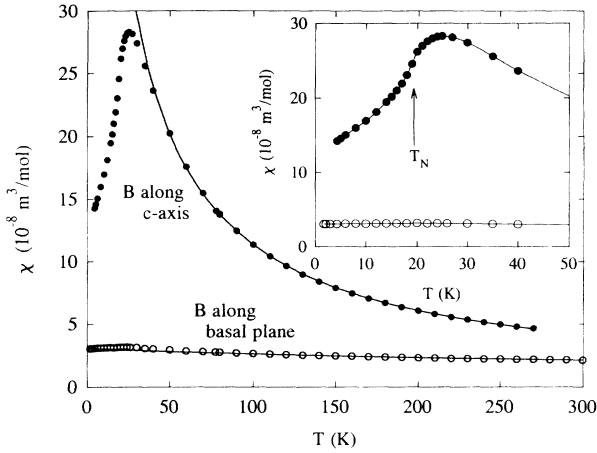


FIG. 2. Temperature dependence of the magnetic susceptibility of the UNiAl single crystal for magnetic field oriented along the c axis and along the ab plane. Lines represent fits described in text. The low-temperature part with arrow indicating the ordering temperature is displayed in the inset.

perature, however, does not coincide with the transition temperature determined from the specific heat ($T_N = 19$ K).¹² Apparently, the magnetic phase transition can be better associated with the maximum in $\partial(\chi T)/\partial T$, as was argued by Fisher¹⁶ and Fedders and Martin.¹⁷ Below 19 K, the c -axis susceptibility shows a quadratic temperature dependence,

$$\chi^{\parallel}(T) = \chi^{\parallel}(0) + \lambda_{\chi} T^2, \quad (1)$$

with $\lambda_{\chi} = 2.8 \times 10^{-10} \text{ m}^3/\text{K}^2 \text{ mol}$ and $\chi^{\parallel}(0) = 14 \times 10^{-8} \text{ m}^3/\text{mol}$, while above 40 K it can be fitted with a modified Curie-Weiss (MCW) law,

$$\chi = \frac{C}{(T - \Theta_p)} + \chi_0, \quad (2)$$

with $\mu_{\text{eff}} = 2.85 \mu_B/U$ atom ($C = \mu_0 N_U \mu_{\text{eff}}^2 / 3k_B$), $\Theta_p = -13$ K, and a small temperature-independent term χ_0 of $1 \times 10^{-9} \text{ m}^3/\text{mol}$. The susceptibility in the basal plane (χ^{\perp}) is much smaller and only weakly temperature dependent. Nevertheless, a tentative analysis by fitting to Eq. (2) with parameters $\mu_{\text{eff}} = 2.25 \mu_B/f.u.$, $\Theta_p = -380$ K, and $\chi_0 \cong 1 \times 10^{-8} \text{ m}^3/\text{mol}$ is applicable in the temperature range 120–300 K. We are aware that a considerable error can be introduced even by a small grain misalignment or a slight misorientation during sample mounting. Both can lead to an admixture of the easy-axis susceptibility, which can distort the data for the hard-magnetization axis especially in the low-temperature range, where the ratio $\chi^{\parallel}/\chi^{\perp}$ is large. This can explain the deviation from the MCW law, which was observed for χ^{\perp} below $T \approx 120$ K. The difference $\Delta\Theta_p \approx -370$ K, between the paramagnetic Curie temperatures obtained for the susceptibility parallel and perpendicular to the c axis, provides an approximate measurement of the anisotropy energy in the paramagnetic range.

The magnetization curves measured at 1.5 and 4.2 K

are displayed in Fig. 3. When the field is applied along the c axis, a metamagnetic transition is observed around 11 T. In fields above the transition, the magnetization slowly saturates, yielding a value of $1.22 \mu_B/f.u.$ in 35 T. The transition becomes more abrupt on cooling to 1.5 K, and the midpoint B_c is shifted from 11.25 T at 4.2 K to 11.35 T at 1.5 K. The small hysteresis of about 0.1 T is similar for both temperatures. The high-field magnetization values are slightly higher at 1.5 than at 4.2 K, especially in the range just above B_c . Measurements on oriented powders of UNiAl, well accounting for the easy-axis magnetization, were performed at 4.2 K in the high-field facility at Osaka University in magnetic fields up to 50 T. They show that the easy-axis magnetization tends to saturate at $1.3 \mu_B/f.u.$ ¹² The slow saturation of the magnetization can be an indication of the itinerant character of the $5f$ magnetism in UNiAl. However, we cannot be conclusive in this respect, because the magnetic structure is not known above B_c and we cannot exclude further transitions in fields above 50 T.

At elevated temperatures the transition is progressively smeared out¹² while its midpoint is somewhat shifted towards lower fields. A remnant of the S shape with inflection around $B = 7$ T is still observable at 24 K, i.e., above T_N , in the range of the susceptibility maximum. This is consistent with the general observation¹⁸ that compounds showing a susceptibility maximum have S-shaped magnetic isotherms below T_{max} .

The basal-plane magnetization is very small and increases linearly with the field, yielding only $0.14 \mu_B/f.u.$ in 35 T. These results demonstrate a huge uniaxial anisotropy of UNiAl with an effective anisotropy field far exceeding the maximum applied field. If one tries to estimate the magnitude of the anisotropy field as the magnetic field where the extrapolated magnetizations $M^{\perp}(B)$ and $M^{\parallel}(B)$ intersect each other, values of several hundred tesla are obtained.

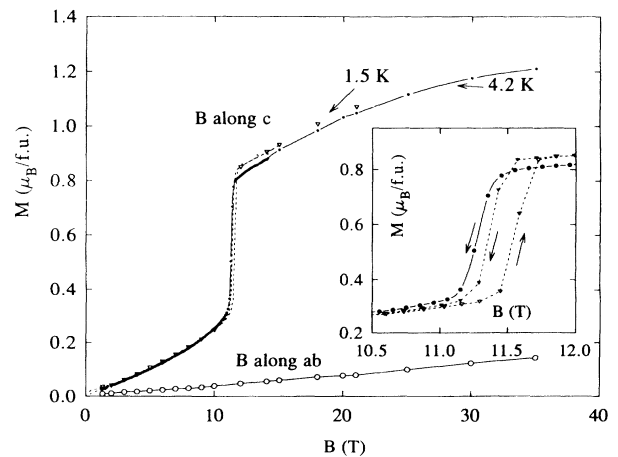


FIG. 3. Magnetization curves of UNiAl single crystal for $B \parallel c$ at 1.5 K (∇) and 4.2 K (\bullet) and $B \perp c$ at 4.2 K (\circ). The inset shows in detail the metamagnetic transition including hysteresis for the 1.5-K measurement. The arrows indicate the direction of field sweeps. Lines are guides to the eye.

IV. TRANSPORT PROPERTIES

The resistivity of UNiAl (Fig. 4) shows little anisotropy at temperatures above 100 K and is nearly temperature independent up to room temperature, where $\rho(300\text{ K}) \approx 150\ \mu\Omega\text{ cm}$. When decreasing the temperature below 100 K, the $\rho^{\parallel}(T)$ and $\rho^{\perp}(T)$ curves (for $i\parallel c$ and $i\perp c$, respectively) progressively deviate from one another. Below 30 K, a negative $\partial\rho/\partial T$ gradually evolves and a maximum is found at 19 K for $i\parallel c$. For $i\perp c$, a less pronounced maximum was found at 15 K. At lower temperatures, ρ^{\parallel} decreases with decreasing temperatures to a low-temperature limit for $\rho^{\parallel}(0.3\text{ K})$ of $85\ \mu\Omega\text{ cm}$, which gives the ratio $\rho^{\parallel}(0.3\text{ K})/\rho^{\parallel}(300\text{ K})$ of 0.57, whereas $\rho^{\perp}(0.3\text{ K}) = 135\ \mu\Omega\text{ cm}$ and the ratio $\rho^{\perp}(0.3\text{ K})/\rho^{\perp}(300\text{ K})$ is 0.90 for the basal plane. The relatively large values of the low-temperature resistivity ρ_0 could be taken as an indication of poor sample quality. However, as we shall see below, a substantial portion of ρ_0 is related to the magnetic state of the sample.

Figure 5 shows the results of an analysis of the low-temperature behavior in terms of possible power laws. We found a T^2 behavior for $\rho^{\perp}(T)$ in the range 0.7–4.5 K. Writing $\rho(T) = \rho_0 + aT^2$ we obtained $a = 5.49 \times 10^{-2}\ \mu\Omega\text{ cm K}^{-2}$ and $\rho_0 = 135.3\ \mu\Omega\text{ cm}$. At very low temperatures, the measured values deviate slightly positively from the quadratic law. On the other hand, the quadratic law holds for ρ^{\parallel} only in a very limited temperature range (0.3–1.5 K). The fitting parameters are: $a = 1.38\ \mu\Omega\text{ cm K}^{-2}$, $\rho_0 = 84.8\ \mu\Omega\text{ cm}$. A much wider temperature interval is covered by the function $\rho = \rho_0 + bT^{5/3}$ with $\rho_0 = 84.56\ \mu\Omega\text{ cm}$ and $b = 1.68\ \mu\Omega\text{ cm K}^{-5/3}$, which is followed by the experimental data at least in the interval 0.3–3.5 K. In this range this type of temperature dependence is clearly distinguishable from those involving T^2 or $T^{3/2}$.

We have performed magnetoresistance measurements of both $\rho^{\parallel}(B)$ and $\rho^{\perp}(B)$ with B parallel to the c axis and of $\rho^{\perp}(B)$ also with B in the ab plane. In the latter case

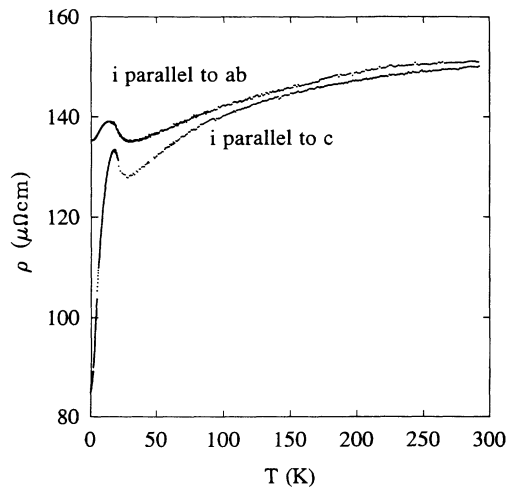


FIG. 4. Temperature dependence of the electrical resistivity of the UNiAl single crystal for $i\parallel c$ (lower curve) and $i\perp c$ (upper curve).

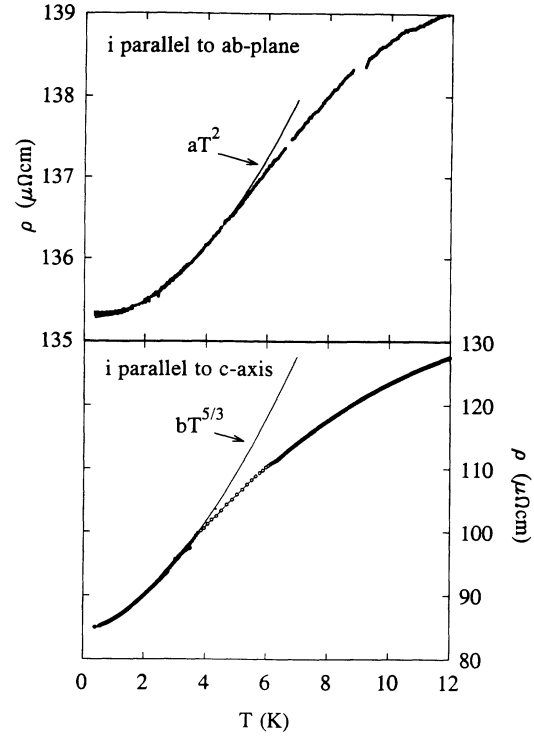


FIG. 5. Low-temperature details of $\rho(T)$ dependencies with lines representing fits described in the text.

(field perpendicular to the easy-magnetization direction), no noticeable field effect is recorded within the relative accuracy of 1%. The similarity of responses of ρ and M to magnetic fields is also seen in the sudden drop of $\rho(B)$ found at the critical field of the metamagnetic transition (Fig. 6). At 4.2 K, ρ^{\parallel} first slightly decreases with B to a shallow minimum at $B = 9\text{--}10\text{ T}$, and then increases again, reaching nearly the zero-field value in fields just below the transition. Then ρ^{\parallel} is reduced abruptly by $\approx 50\ \mu\Omega\text{ cm}$, and in the high-field range it displays a weakly saturating tendency analogous to the $M(B)$ behavior. A value of ρ^{\parallel} of about $20\ \mu\Omega\text{ cm}$ is recorded in the high-field limit. In the range 11–14 T, the results of the pulsed-field and quasistatic-field measurements display a noticeable disagreement. The reason may be in rather slow relaxation phenomena, traced in the time dependence of ρ during the field pulse in this range. Due to the limited accuracy of the pulsed-field measurement, these transient phenomena have not been studied yet systematically.

At $T = 1.5\text{ K}$ only the pulsed-field measurements were performed. The results are similar to the experiment at $T = 4.2\text{ K}$ in the quasistatic field, i.e., the transition is much sharper than seen at 4.2 K in the pulsed fields. The absolute value of the drop in ρ^{\parallel} at the transition remains approximately the same, but the values saturate faster in high fields. One should note that the difference between $\rho^{\parallel}(B, 4.2\text{ K})$ and $\rho^{\parallel}(B, 1.5\text{ K})$ above B_c is much more pronounced than the corresponding difference in $M^{\parallel}(B)$. Comparing approximate absolute values of $\rho^{\parallel}(B, 4.2\text{ K})$ and $\rho^{\parallel}(B, 1.5\text{ K})$, which are practically equal in the high-

field limit, we can see that the initial steep increase of $\rho^{\parallel}(T)$ in zero field, which is maintained still in a limited field range above B_c , is progressively suppressed in high magnetic field. The maximum in $\rho(B)$ followed by a drop at B_c is quite common in antiferromagnets. In the simple Néel model with two sublattices, the dominating effect is the loss of periodicity due to the reversal of moments residing in the sublattice with antiparallel orientation.¹⁹ Similarly, an increase of ρ up to the metamagnetic transition is expected also for band antiferromagnets.²⁰

For i in the ab plane, $\rho^{\perp}(B)$, a slow monotonic decrease of the resistivity is observed in fields up to about 9 T in agreement with results of Schoenes *et al.*²¹ Such a behavior is pertinent to ferromagnets and, in general, can be accounted for by a suppression of magnetic fluctua-

tions. In materials close to the onset of magnetic order there are mostly fluctuations of the paramagnon type being suppressed.²² This behavior, which is qualitatively different from that for the other current direction, may be related to the (nearly) ferromagnetic coupling in the ab plane. However, ρ^{\perp} also then drops at the transition by about 50% and, in larger fields, it behaves qualitatively like $\rho^{\parallel}(B)$. A remarkable difference is, however, the weaker tendency to saturation. A comparison of measurements at 1.5 and 4.2 K shows again that the critical-field drop is more pronounced at lower temperature, but both curves join in fields above 25 T. Also here we see the discrepancy mentioned for ρ^{\parallel} . The magnetoresistance measured for ρ^{\parallel} at 77 K shows only a weak (2%–3%) decrease of resistance in $B = 35$ T.

Schoenes *et al.*²¹ studied also the Hall effect in UNiAl. It shows qualitatively almost the same temperature dependence as the magnetic susceptibility, from which it can be concluded that the Hall effect in UNiAl is dominated by magnetic contributions, i.e., by the anomalous part. The value of the normal part, $R_0 = -1.33 \times 10^{-3}$, has been determined by the decomposition of the total Hall effect for $B \parallel c$. This corresponds, in a one-band model, to a conduction-electron concentration of 0.77 per formula unit. A normalization of the anomalous part of the Hall effect with the magnetic susceptibility shows that the Fermi surface is reconstructed below T_N , but the effect is much less pronounced than in URu_2Si_2 .²³

V. SPECIFIC HEAT

The temperature dependences of the specific heat measured in various magnetic fields ($B \parallel c$) up to 20 T are seen in Fig. 7. The most prominent feature is the peak connected with the Néel temperature. When applying the field, this peak is progressively shifted from $T_N = 19.3$ K ($B = 0$) to lower temperatures, which is a tendency expected in an antiferromagnet. In magnetic fields of 11 and 11.25 T, the peak becomes apparently narrower and sharper resembling a first-order transition. Finally, the

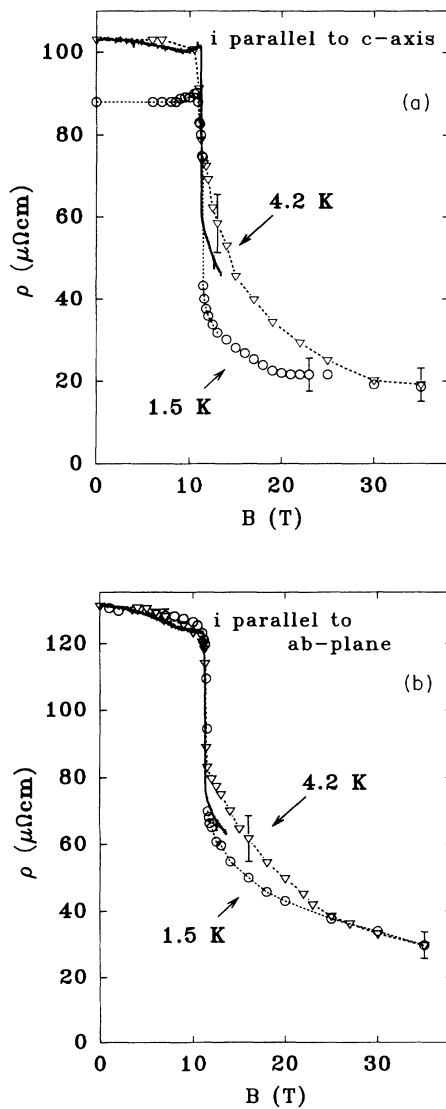


FIG. 6. Magnetic-field dependence of the electrical resistivity of the UNiAl single crystal in pulsed fields at 4.2 K (∇) and 1.5 K (\circ) for $B \parallel c$, (a) with $i \parallel c$, and (b) with $i \perp c$. Solid lines represent in both cases the measurements performed at 4.2 K in quasistatic fields. The error bars indicate the absolute accuracy of ρ in the pulsed field; the dashed lines are guides to the eye.

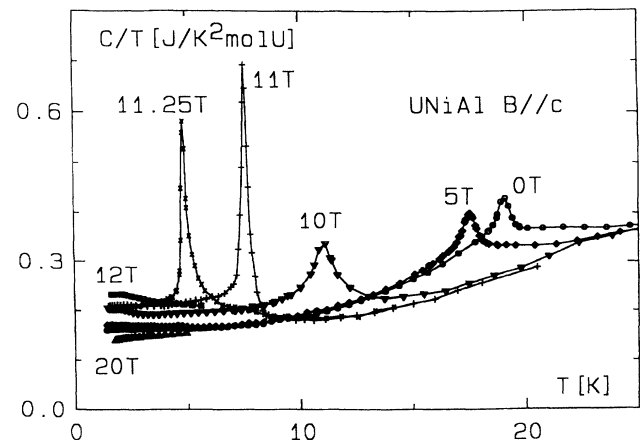


FIG. 7. Temperature dependence of the specific heat of UNiAl (C/T vs T) in various selected magnetic fields applied along the c axis. The lines are guides to the eye.

anomaly disappears in higher magnetic fields ($B \geq 11.5$ T) indicating suppression of the antiferromagnetic order. Comparison of the zero-field data with those measured in field shows that in zero field there can be a non-negligible part of the magnetic entropy located above T_N and this part is gradually removed with increasing field. In contrast to the field along the c axis, the ordering temperature of UNiAl is not affected by a magnetic field applied perpendicular to it,¹² but the related peak becomes somewhat enhanced and broadened. Moreover, the shoulder in C/T vs T above T_N is slightly enhanced by a field applied in the basal plane.

The B - T diagram in Fig. 8 displays in one plot the temperature dependence of the metamagnetic transition field B_I , determined from the inflection point of the magnetic isotherms presented by Havela *et al.*,¹² and the field dependence of T_N taken as the temperature of the maximum in the C/T -vs- T curve in various magnetic fields for $B \parallel c$. The Néel temperature exhibits the usual quadratic dependence on the applied field. The two curves in Fig. 8, $B_I(T)$ and $B(T_N)$, coincide at low temperatures, where the sharp peaks (suggesting first-order transitions) in the specific heat occur. At higher temperatures, the field of the metamagnetic transition, which is gradually smeared out but still visible at 24 K (i.e., above T_N), decreases monotonically to a value of 6 T. These observations corroborate the idea about antiferromagnetic (AF) correlations dominating the behavior of the magnetic system in a limited temperature range above the actual T_N in the part of the diagram where the two curves deviate from one another ($T > 10$ K), while the magnetic phase transition at T_N is of the second-order type. One should realize that a magnetic phase diagram containing a tricritical point, which separates the low-temperature part of the B - T line between antiferromagnetic and field-aligned paramagnetic phase from the high-temperature one, is rather normal in a broad class of mean-field local-moment models.²⁴ This means that the metamagnetic transition at $T=0$ K is usually a first-order transition, while the Néel point in 0 T is usually a second-order transition.

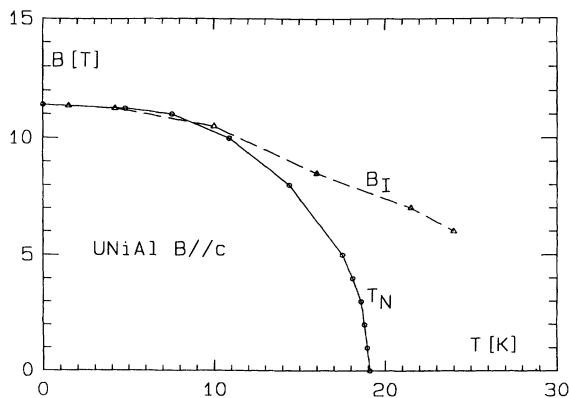


FIG. 8. B - T diagram displaying inflection points B_I of magnetic isotherms vs T and ordering temperatures T_N vs magnetic field for $B \parallel c$.

Besides the gradual suppression of T_N , the magnetic field applied along the c axis also strongly affects the low-temperature part of the C/T -vs- T dependence (Fig. 9). An upturn, which develops gradually in the temperature range below 10 K, can be followed to fields larger than the critical field of the suppression of antiferromagnetism (up to 15 T). Only in the highest applied field (20 T) is it not observed any more. In order to obtain values for the γ coefficient in field we have to find an acceptable way of extrapolating C/T to $T=0$ K. Although theoretical reasons for applying this dependence are disputable (see Sec. VIII), we tried to apply the commonly used formula

$$C/T = \gamma + \beta^* T^2 + \delta T^2 \ln T, \quad (3)$$

which describes well the experimental data in the temperature range up to 5–10 K. The fitting parameters are summarized in Table I. Applying tentatively a spin-fluctuation (paramagnon) model,^{25,26} the experimental coefficient of the quadratic term β^* can be related to the coefficient of the lattice contribution β by the expression

$$\beta^* = \beta - \delta \ln T_{SF}, \quad (4)$$

where T_{SF} represents the characteristic temperature of spin fluctuations. Provided β is known, we can derive the field dependence of T_{SF} . For this purpose we tried several ways of estimating β . One method is to take β of the isostructural nonmagnetic compound YNiAl. Another method is to estimate the slope of C/T vs T^2 in UNiAl in the temperature range above the low-temperature upturn and below the significant contribution of the magnetic entropy in the vicinity of T_N . This temperature

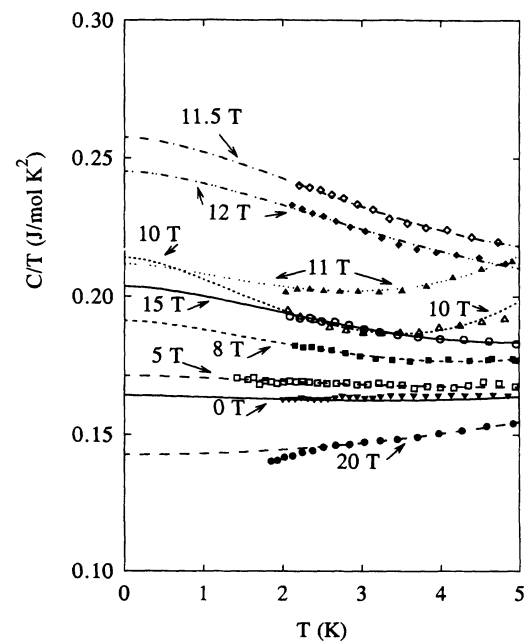


FIG. 9. Low-temperature detail of the specific heat of UNiAl in various magnetic fields. The lines represent fits to Eq. (3) with parameters given in Table I.

TABLE I. Parameters obtained from the low-temperature C/T -vs- T data obtained on UNiAl in various magnetic fields applied along the c axis. The data were fitted to formula (3), values of T_{SF} were obtained supposing various β values: I, 0.000 488 J/mol K⁴ (from UNiAl in 20 T); II, 0.000 396 J/mol K⁴ (from UNiAl in 0 T, temperature range 8–12 K); and III, 0.000 186 4 (from YNiAl).

B (T)	γ (J/mol K ²)	β^* (J/mol K ⁴)	δ (J/mol K ⁴ lnK)	T_{SF}^{I}	$T_{\text{SF}}^{\text{II}}$ (K)	$T_{\text{SF}}^{\text{III}}$
0	0.1643	−0.000 60	0.000 363	20.2	15.7	8.8
5	0.1714	−0.000 75	0.000 368	30.7	22.6	12.8
8	0.1912	−0.003 15	0.001 62	9.5	8.9	7.8
10	0.2142	−0.007 88	0.004 51	6.4	6.3	6.0
11	0.2118	−0.003 64	0.002 34	5.8	5.6	5.1
11.5	0.2576	−0.005 38	0.002 37	11.9	11.5	10.5
12	0.2451	−0.004 26	0.001 78	14.3	13.6	12.1
15	0.2037	−0.003 52	0.001 68	10.8	10.3	9.1
20	0.1426	0.000 488				

range is, however, rather narrow (7–11 K). The third method we have applied was to take β from the C/T measurements of UNiAl in 20 T, in the field where magnetic fluctuations are suppressed, at least to such an extent that no upturn was observed in the C/T -vs- T plot (C/T is linear in T^2). From the large spread of T_{SF} values obtained using somewhat different β values we can see that this procedure is very sensitive and the results should therefore be taken with caution. An intrinsic uncertainty can originate from the fact that theoretically an anisotropy in the paramagnon excitations can appear (see discussion), which means that there can be two different T_{SF} values.

From Fig. 10 we can see a progressive increase of γ up to $B = 10$ T. The point at 11 T is apparently affected by the proximity of the phase transition to the temperature range from where the extrapolation was performed (see Fig. 7). Therefore, we cannot decide whether there is a true discontinuity or whether $\gamma(B)$ increases smoothly up to its maximum value of approximately 260 mJ/mol K² found at 12 T. Above the transition field, γ decreases, reaching at 20 T a value 143 mJ/mol K², which is already lower than that obtained in zero field.

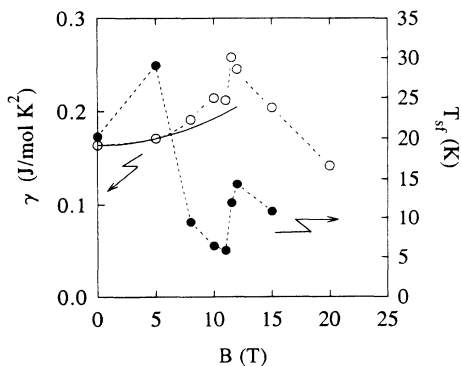


FIG. 10. Development of extrapolated γ values of the UNiAl single crystal in fields applied along the c axis (\circ). The solid line represents the quadratic fit to the initial increase of $\gamma(B)$. \bullet symbols are the corresponding values of T_{SF} . Dashed lines are guides to the eye.

It is interesting to check whether the field dependence of the specific-heat coefficient γ is consistent with the second derivative of the low-temperature susceptibility, i.e., whether it obeys the thermodynamic Maxwell relation²⁷

$$B(\partial^2\chi/\partial T^2) = \mu_0(\partial\gamma/\partial B), \quad (5)$$

where χ is the zero-field static susceptibility.

We have fitted the initial increase of $\gamma(B)$ to a quadratic dependence,

$$\gamma = \gamma_0 + \kappa_C B^2, \quad (6)$$

with $\gamma_0 = 164$ mJ/mol K² and $\kappa_C = 2.8 \times 10^{-4}$ J/mol K² T². The agreement of the latter parameter with the value of 2.3×10^{-4} J/mol K² T², calculated by means of (5) from the $\chi(T)$ dependence, is rather good considering the large experimental inaccuracy in the determination of κ_C .

VI. MAGNETOCALORIC EFFECT

Measurements of the magnetocaloric effect (adiabatic heating or cooling of the sample due to field variations) confirm the first-order character of the metamagnetic transition at low temperatures, because an abrupt step of the temperature is recorded at the critical field B_c (Fig. 11). The effect was measured by monitoring the temperature of the adiabatically mounted sample while sweeping the field at different rates. The hysteresis of the transition is about 0.1 T, i.e., similar to the value obtained from magnetization measurements. As follows from the Clausius-Clapeyron equation for the case of negative $\partial B_c/\partial T$, the high-field phase has higher entropy at the same temperature than does the antiferromagnetic phase. Therefore one would expect a heating effect when crossing B_c from above, which is indeed observed. However, the curve is not retraced with increasing field and we can even obtain the opposite step (i.e., also heating) depending on the sweeping rate. Although this effect was not followed systematically under different conditions, it is clear that some heating mechanism depending on the sweeping rate is at work. At certain conditions we can reach even a cancellation of the “intrinsic” magneto-

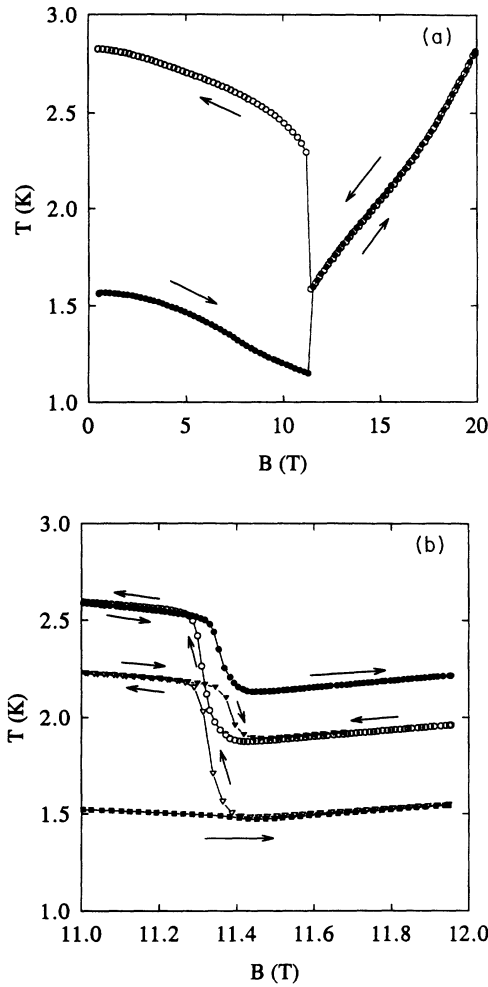


FIG. 11. Temperature of an adiabatically mounted sample of UNiAl vs magnetic field applied parallel to c . Decreasing field is marked by open symbols, increasing field by solid symbols. The sweeping rate was 4 T/min in (a) and 0.2 T/min in (b).

caloric effect by this additional effect. This effect, especially its possible relation to magnetostriction effects, deserves further study.

The change of sign of the slope of the $T(B)$ curves at the transition ($\partial T/\partial B < 0$ for $B < B_c$ and $\partial T/\partial B > 0$ for $B > B_c$, see Fig. 11) can be understood on the basis of the thermodynamic relation:

$$\left\{ \frac{\partial T}{\partial B_e} \right\}_{S,p} = - \frac{T}{C_{B,p}} \left\{ \frac{\partial M}{\partial T} \right\}_{B,p}, \quad (7)$$

where $C_{B,p}$ is the specific heat in a given field and B_e the external magnetic field. As can be derived from the temperature development of the magnetic isotherms,¹² at low temperatures the derivative $\partial M/\partial T$ changes sign from positive for $B < B_e$ to negative for $B > B_e$ and therefore $\partial T/\partial B_e$ should undergo the opposite change of sign.

VII. Y, Co, AND Fe SUBSTITUTIONS IN UNiAl

As shown already in earlier papers,^{28,29} substitution of Y for U leads to a very fast breakdown of the long-range

magnetic order in UNiAl. Upon substitution of a mere 5% of U by Y the specific-heat anomaly connected with T_N disappears. Nevertheless, the susceptibility displays a broad maximum, gradually shifted to lower T , which is observable up to 10% Y. Correspondingly a metamagnetic transition can be observed at approximately the same field as in pure UNiAl. The magnitude of the magnetization change at the transition is, however, rapidly reduced and practically linear $M(B)$ curves are found for 20% and 40% Y. The most remarkable effect is the gradual development of a low-temperature upturn in the C/T -vs- T curves, which can also be well accounted for by formula (3). The parameters obtained, using the same analysis as for pure UNiAl, are shown in Fig. 12. The values for the γ coefficient reach a maximum around 20% Y, which means in a sample displaying no metamagnetic transition, but also for a sample with 40% Y a noticeable C/T upturn is still found. We should note that these upturns remain practically unaffected by magnetic fields up to 5 T, applied on polycrystalline samples. A crystal-structure analysis showed a noticeable lattice expansion in the a direction (1.9% for 40% Y), whereas the lattice constant c shrinks (by 2.4% for 40% Y).

Substitution of Ni by Co or Fe has a different impact on the lattice parameters. Whereas the parameter a remains practically unchanged, c is reduced, but to a lesser extent than for Y substitution (1.2% for 40% Co or 20% Fe). A gradual decay of magnetism (which is not unexpected because UCoAl is a band metamagnet³⁰ and UFeAl an exchange-enhanced Pauli paramagnet³¹) is apparent in the temperature dependencies of the specific heat. The anomaly related to T_N shifts to lower T and is finally embedded in the low-temperature upturn in C/T , which gradually develops. This tendency can be followed to concentrations of 20% Co or 10% Fe, where a γ value of approximately 250 mJ/mol K² is recorded in both cases. For the Co case, a decrease of γ was observed for 40% substitution. Co and Fe substitutions also lead to a suppression of the field-induced transition in $M(B)$, measured at 4.2 K, but in a different way from Y substitutions. The initial slope of $M(B)$ increases and the positive curvature is converted into a negative one with increasing concentration of the substituting element. The S

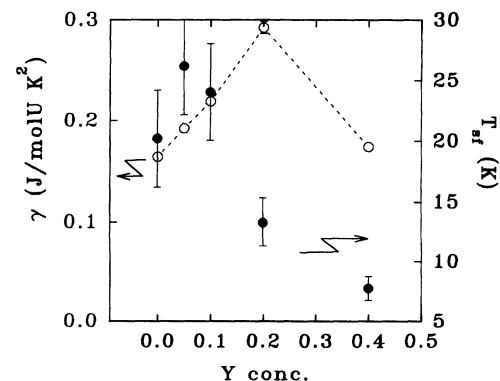


FIG. 12. Concentration dependence of extrapolated γ values and T_{SF} in the system (U,Y)NiAl.

shape disappears between 10% and 20% Co or 5% and 10% Fe. The absolute values of the high-field magnetization are reduced upon substitution, but much less than for the Y case. Apart from the delocalization of U moments, this tendency indicates the dominant role of ferromagnetic correlations along the c axis favored by Co or Fe substitutions. This situation contrasts with the Y substitution, where antiferromagnetic correlations persist even in the paramagnetic regime.

VIII. DISCUSSION

Results of magnetic, electrical, and specific-heat measurements are consistent with antiferromagnetic ordering in UNiAl below $T_N = 19$ K. In all cases, a very strong magnetic anisotropy, persisting to the paramagnetic state, was observed. The magnetic response is concentrated in the c direction, yielding uniaxial anisotropy. The effective anisotropy fields are estimated as several hundred tesla. It is, however, difficult to imagine that using a hypothetical field (oriented in the basal plane) larger than the anisotropy field we could simply turn the moments out of the easy-magnetization direction. The close connection of the geometry of the crystal lattice and magnetic anisotropy, which is generally valid for all UTX compounds with the ZrNiAl type of structure, suggests that the direction of magnetic moments is determined by general features of bonding symmetry. In particular, models based on the notion of hybridization-mediated anisotropic exchange^{32,33} predict the orientation of magnetic moments perpendicular to high-coordination directions (planes), and this prediction was found valid also for UTX compounds crystallizing in other structure types.³⁴ In UNiAl, similar to other UTX compounds of the ZrNiAl-type family, the high coordination of nearest U neighbors in the U-Ni basal plane, together with a strong hybridization of the $5f$ states of U with the $3d$ states of Ni within the same plane, is a probable source of the rigid orientation of magnetic moments perpendicular to the plane. Any attempt to rotate the magnetic moments from this direction (e.g., by magnetic field) would lead, in the framework of these models, to a breaking of the bonds within the U-Ni layers. Thus, the binding energies (the difference of binding energies for different structure types) can provide the energy scale for the effective fields of the magnetic anisotropy (typically of the order of 100 meV).

Other important features influencing the properties of UNiAl are the magnetic fluctuations. The origin of high γ values is traditionally attributed to on-site spin fluctuations related to the Kondo effect or to similar many-body phenomenon affecting the magnitude of the local magnetic moments. They can cause a substantial enhancement of the effective mass m^* , because their magnitude is large in the whole Brillouin zone. Besides those there are also the intersite magnetic fluctuations with low characteristic energies, which can lead to formation of the low-temperature upturn in C/T .³⁵ We can still classify the latter effects into ferromagnetic spin fluctuations (paramagnons) with ($q \approx 0$) and antiferromagnetic spin fluctuations with the wave vector q concentrated around

a finite Q , which is the propagation vector of the antiferromagnetic structure. The antiferromagnetic spin fluctuations can have presumably a better possibility for a large γ enhancement than the ferromagnetic ones, which are large only in a limited volume around the center of the Brillouin zone.³⁶ The existence of a $T^3 \ln T$ term in the $C(T)$ dependence was first derived for ferromagnetic spin fluctuations,^{25,37} but such a term appears also in a more general Kondo-lattice model leading to a heavy Fermi liquid.³⁸ On the other hand, pure antiferromagnetic spin fluctuations, although giving rise to a γ enhancement,³⁹ do not lead to this logarithmic term due to a different critical behavior of the ferromagnetic and antiferromagnetic spin fluctuations.⁴⁰ These theories have been developed mainly for nearly ferro- or antiferromagnetic materials. UNiAl, although magnetically ordered, is close to the onset of magnetism. This is demonstrated by the low value for the U moments. The observed $\mu_U \approx 0.5 \mu_B$ is not only much lower than the free f^2 - or f^3 -ion values, which is in fact rather normal for most of the U intermetallics, but also compared to other ordered materials with the ZrNiAl structure, where moments of $1 \mu_B - 1.5 \mu_B$ per U are typical. For example, UNiGa displays moments of $1.35 \mu_B / U$.⁴¹ The reason for such different magnetic properties of UNiGa and UNiAl can be found in the $5f$ hybridization with p states, which should be generally weaker for Ga than for Al. This tendency was indeed also found for other UAl and UTGa compounds.⁶ The other indication that UNiAl is close to the onset of magnetic order is a strong sensitivity to doping, illustrated by the Y substitution, and also the small entropy change (estimated¹² as $0.14 \times R \ln 2$) associated with the magnetic phase transition. The other indirect evidence comes from photoelectron spectroscopy. Both valence-band and core-level U $4f$ spectra display only weak features, which can be attributed to $5f$ localization or electron-electron correlations in the $5f$ band.⁴²

The character of the intersite coupling in UNiAl gives a possibility of both antiferromagnetic spin fluctuations around $Q = (0, 0, 1/2)$ and ferromagnetic (or nearly ferromagnetic) ones based on correlations in the basal plane. The antiferromagnetic correlations are probably the origin of the broad maximum in $\chi^{\parallel}(T)$ above T_N . Their breaking in fields around 10 T is demonstrated by the S shape of the $M^{\parallel}(B)$ dependencies. Furthermore, the short-range AF ordering was measured directly by neutron-diffraction experiments.¹³

For direct information about magnetic fluctuations in the magnetically ordered state, inelastic neutron-scattering experiments are highly desirable. However, the progressive increase of γ with field applied along the c axis and especially the maximum in $\gamma(B)$ coinciding practically with the field of the metamagnetic transition give a clear impression that a substantial portion of γ is related to intersite spin fluctuations. There is a certain analogy, for example, with the system CeRu₂Si₂,⁴³ which has a nonmagnetic ground state, but for which AF correlations are demonstrated by the neutron-scattering technique. A maximum of γ is also found here at the metamagnetic transition around 8 T, where these correlations are suppressed.⁴⁴ On the other hand, the width of

the quasielastic line, reflecting a local-moment instability (on-site spin fluctuations) is nearly field independent.

The enhancement of γ up to a maximum where the antiferromagnetic correlations are presumably suppressed, and the development of the $T^3 \ln T$ term, could point to a dominant role of ferromagnetic spin fluctuations. They can gradually evolve with increasing field from antiferromagnetic spin fluctuations, but in the high-field limit they have to be suppressed again due to the increasing Zeeman spinning of the spin-up and spin-down levels. The latter effect can account for the decrease of γ above the metamagnetic transition. The alternative framework for understanding the depression of γ in high fields, which was used in Ref. 44, is the field effect on the Kondo resonance.

The analogy of UNiAl with CeRu₂Si₂ is, however, not complete. For the latter compound, which is driven to the ordered antiferromagnetic state by small substitutions of La, no γ enhancement in field was found.⁴⁵ This system, however, also demonstrates the possibility of coexistence of long-range order and short-range dynamic antiferromagnetic correlations, indicated by neutron scattering.⁴⁶ As to other materials, high-field specific-heat data are available for the U(Pt,Pd)₃ system, where a similar enhancement of γ around the metamagnetic transition was found for both UPt₃ and U(Pt_{0.95}Pd_{0.05})₃.⁴⁷⁻⁴⁹ The discussion in Ref. 48 introduces also the possibility of field-enhanced ferromagnetic spin fluctuations in essentially antiferromagnetically correlated systems.

Complementary information can be extracted from studies on substituted UNiAl. A qualitatively similar monotonic increase of γ and a gradually developing $T^3 \ln T$ term are observed up to concentrations of 20% Y despite the fact that AF correlations are indicated by a metamagnetic transition and a maximum in $\chi(T)$ only for 5% and 10% Y. This means that similar enhancement effects occur also in the compound with a paramagnetic ground state and that ferromagnetic spin fluctuations, induced here not by external field but by substitution, can be a plausible source of the γ enhancement. On the other hand, a precipitous drop of γ was found in the pseudoternary system UNiAl_{1-x}Ga_x in the concentration range between 20% and 40% Ga.⁵⁰ The fact that the U magnetic moments (derived from bulk magnetization in 35 T) increase rapidly when increasing Ga concentration through this range²² can point to the importance of on-site spin fluctuations. Such fluctuations can be responsible for a considerable portion of the (zero-field) γ in UNiAl and cause the U-moment reduction.

A variety of different intersite magnetic fluctuations in the antiferromagnetic state is demonstrated by the anisotropy of the electrical resistance and its field dependence. When inspecting the low-temperature $\rho(T)$ behavior for i in the basal plane, we find that the value of 0.055 $\mu\Omega \text{ cm K}^{-2}$ for the coefficient a of the quadratic term gives the ratio $a/\gamma^2 = 2 \times 10^{-6} \mu\Omega \text{ cm mol}^2 \text{ K}^2/\text{mJ}^2$, which fits between transition metal values⁵¹ and heavy-fermion values,⁵² with the characteristic ratio of $a/\gamma^2 = 0.4 \times 10^{-6} \mu\Omega \text{ cm mol}^2 \text{ K}^2/\text{mJ}^2$ and $1 \times 10^{-5} \mu\Omega \text{ cm mol}^2 \text{ K}^2/\text{mJ}^2$, respectively. However, the value found for i along the c axis ($a = 1.38 \mu\Omega \text{ cm K}^{-2}$) is by a

factor of 5 too high compared even with the heavy-fermion systematics for a given γ value. This situation points again to the presence of spin fluctuations based on correlations along the c axis. Theories of itinerant antiferromagnets lead to a divergence of the a coefficient around the onset of long-range ordering together with a progressive restriction of the validity of the T^2 law to a smaller temperature range. Instead of this, a lower exponent ($\frac{3}{2}$ or $\frac{5}{3}$) characterizes the $\rho(T)$ dependence over a more extended temperature range.⁵³ Nevertheless, a similar behavior with a $\frac{5}{3}$ exponent was predicted⁵⁴ and observed⁵⁵ for compounds with ferromagnetic spin fluctuations. Thus, the $\rho(T)$ behavior only does not give us a clear-cut distinction between ferromagnetic and antiferromagnetic fluctuations, but the importance of fluctuations, originating in the AF correlations along c , is evident.

Here, we have not studied the field dependence of a , but the ratio of the high-field values at $T = 1.5$ and 4.2 K points to a much less steep initial increase in the high-field limit. However, we can compare this situation with UNiGa, which has also an AF ground state and the metamagnetic transition can be achieved in fields below 1 T. Also here the value for a of $2.4 \times 10^{-1} \mu\Omega \text{ cm K}^{-2}$ for the c -axis resistance is too high for a compound with $\gamma = 59 \text{ mJ/mol K}^2$, but this value is reduced by a factor of 5 above the transition.⁵⁶ The remarkable fact is that the magnetic moments observed by neutron diffraction are of the same size below and above the metamagnetic transition and also the γ value stays invariable. This means that the a - γ^2 relation can be violated.

Large resistance changes connected with the metamagnetic transitions in antiferromagnets are usually attributed to the suppression of AF fluctuations and to the disappearance of magnetic superzones when the AF ordering is suppressed by a magnetic field.^{20,57} The case of UNiGa (with presumably more stable magnetic moments), where the resistance along the c axis drops at the metamagnetic transition by 87%, also shows how large negative magnetoresistance effects can be related to a rearrangement of magnetic structure. This would mean that effects like decoupling of conduction electrons and magnetic moments (field-induced breakdown of the Kondo interaction) need not be important. We are, however, in doubt about the main mechanism of the large negative magnetoresistance. The observed Fermi-surface reconstruction²¹ due to a new periodicity can at least contribute to the large drop of ρ in UNiAl, because the field-induced breakdown of the antiferromagnetic correlations along the c axis can lead to a suppression of the gapping of part of the Fermi surface. Another reason can be found in spin-dependent scattering effects.

Adding all these results together we can try to construct a tentative model based on the existence of ferromagnetic spin fluctuations within the basal planes and antiferromagnetic fluctuations connected with a weaker antiferromagnetic interplane coupling, the energy of which is given by the field of the metamagnetic transition. The intersite fluctuations can contribute to the relatively high γ value in the ground state, provided they survive in the ordered state. Another source of the high γ

can be the on-site spin fluctuations.

The field dependence of γ can be related to the breaking of the AF correlations, enabling thus the population of low-energy (low- T_{SF}) ferromagnetic spin fluctuations (paramagnons), which are based on coupling along the c axis, in the critical region around B_c . The insensitivity of the resistance to these low- q excitations causes the fact that the most prominent magnetoresistance feature is the large drop at B_c .

If we speculate even further, we can relate the increase of γ for Y-, Co-, and Fe-substituted UNiAl to a reduction of the strength of the in-plane magnetic coupling, which leads to paramagnonlike in-plane fluctuations, and/or to a gradually prevailing ferromagnetic interplanar coupling allowing a population of the c -axis paramagnons in zero field for the Co and Fe substitutions. This assumption is corroborated by the observed tendency to ferromagnetism for compounds with transition metals with a reduced number of d electrons.⁷ However, also here the role of local spin fluctuations can be important, and a real

analysis cannot be done on the basis of bulk measurements only. The very fast suppression of long-range magnetic order for any of these substitutions shows unambiguously that the electronic structure in UNiAl is very close to the onset of a magnetic instability, and any small increase of hybridization and possibly also the loss of supporting Coulomb interactions due to dilution by Y returns the system to the nonmagnetic ground state.

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