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Electronic properties of U_2Cu_9Al

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Abstract. We report on the electronic properties of U_2Cu_9Al, which crystallizes in a hexagonal structure with space group $P6_3/mmc$. Anomalies in the temperature dependences of the transport, magnetic and thermal properties indicate that two magnetic transitions occur at ~9.5 K and ~31 K, the lower transition being of antiferromagnetic type. The assertion that the ground state of U_2Cu_9Al is antiferromagnetic is corroborated by magnetization measurements, which reveal a slight S shape in the M versus B curve with no indication of saturation in fields up to 18 T at 2.2 K. The electrical resistivity increases with lowering temperature, reaching a value of 284 $\mu\Omega$ cm at 0.26 K. Magnetoresistance studies at low temperatures show that a large part of the zero-field resistance is due to antiferromagnetic correlations, and a resistivity drop of ~30% is observed upon the application of a field of 18 T at 2 K. Another striking feature is the enhanced Sommerfeld coefficient γ of ~210 mJ mol_U^{-1} K^{-2}, extracted from extrapolation of the specific-heat data to $T = 0$ K, which we report here for the first time. Therefore, U_2Cu_9Al can be described as a moderately enhanced heavy-fermion system.

1. Introduction

In the last two decades a number of heavy-fermion materials have been identified and studied extensively. These materials contain 4f or 5f elements and they show peculiar properties at low temperatures, which can very often be explained within the context of the Fermi-liquid (FL) theory [1]. In the limit $T \to 0$, this theory yields a large temperature-independent contribution to the specific heat, aT^2-type behaviour of the electrical resistivity and a substantial contribution to the temperature-independent magnetic susceptibility χ_0. The physics of heavy-fermion compounds can be interpreted in terms of large renormalized effective electronic masses which occur as a consequence of either screening effects of a localized magnetic moment due to the Kondo effect or magnetic correlation effects in itinerant systems. The heavy-fermion behaviour is very often found at the borderline between local-moment

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and itinerant magnetism or between magnetically ordered and disordered systems. This is also the region where deviations from behaviours predicted by FL theory often occur. This is referred to as non-Fermi-liquid (NFL) behaviour and one of the hallmarks of NFL behaviour is a divergence in the low-temperature specific heat divided by the temperature, C/T, which behaves as $-\ln(T/T_0)$. Other properties may also exhibit divergences at low temperatures. NFL behaviour was found in a number of dilute 4f or 5f systems, where a certain type of disorder is introduced into the structure. In general, NFL compounds fall into two categories: (a) systems with the f-ion sublattice diluted with non-magnetic atoms [2–4]; and (b) systems in which the f-ion sublattice is not diluted but the surrounding is changed by substitutions [5, 6]. NFL behaviour is also the subject of several theoretical investigations which include the electrical or magnetic Kondo effect [7, 8], the distribution of Kondo temperatures [9], fluctuations in the proximity of a zero-temperature quantum phase transition [10, 11] or an instability in low-dimensional systems with diverging susceptibility at $q = 2k_F$ [12]. Recently, de Andrade *et al* [13] suggested that the NFL properties of disordered f-electron systems can be described in terms of a theory based on Griffith's singularities.

Recently, a detailed study of single-crystal and polycrystalline $UCu_{5-x}Al_x$ compounds revealed that the low-temperature magnetic, thermal and transport properties exhibit NFL behaviour [14]. Here, we report on the bulk electronic properties of another member of the U–Cu–Al family, namely U_2Cu_9Al. To the best of our knowledge, no electronic properties of this compound have been reported on since Blazina and Ban identified the crystallographic structure 25 years ago [15].

2. Sample preparation and characterization

A polycrystalline sample of U_2Cu_9Al has been prepared by arc melting stoichiometric amounts of the constituent elements. The phase purity and the structural parameters were verified by means of x-ray diffraction. For this measurement, U_2Cu_9Al was ground under an argon atmosphere and mixed with a small amount of fine silicon powder which we have used as an internal calibration standard. We used Cu Kα radiation, and the diffraction pattern was analysed by means of the Rietveld-refinement program FULLPROF [16], which allows simultaneous refinement of multiple phases. X-ray powder diffraction showed for a freshly powdered specimen virtually no extra phases, thus indicating the content of foreign phases to be below the sensitivity limit of this method, i.e. 3–5%. However, an x-ray pattern obtained for the same powder ten months later revealed that disproportionation occurred over time and a substantial amount of U_2Cu_2Al and Cu could be identified. In the light of our observation that polished surfaces of specimens of U–Cu–Al acquire Cu coloration in a few hours, it

Table 1. The refined structural parameters of U_2Cu_9Al at $T = 293$ K.

Space group: $P6_3/mmc$			
Atom	Site	Position parameters	
U	2(a)	0 0 0	
Cu_1	4(f)	1/3 2/3 z_{Cu_1}	$z_{Cu_1} = 0.1028$ (35)
Cu_2/Al	6(h)	x_{Cu_2} $2x_{Cu_2}$ 1/4	$x_{Cu_2} = 0.4422$ (32)
Lattice parameters		R-factors	
$a = 491.35 \pm 0.11$ pm		$R_p = 2.55\%$	$\chi^2 = 6.1$
$c = 909.42 \pm 0.47$ pm		$R_{wp} = 4.10\%$	

is tempting to suggest that the disproportionation observed in U_2Cu_9Al may stem from the large surface area produced by grinding. Therefore, we believe that the results presented are intrinsic properties of U_2Cu_9Al. For U_2Cu_9Al, our refinement indicates the hexagonal space group $P6_3/mmc$ with structural parameters which agree well with literature values [15], as given in table 1. The structure consists of two distinct planes. One contains U atoms at 2(a) sites and a fraction of Cu atoms at 4(f) sites shifted out from the U plane, while the other plane consists of statistically distributed Al and the rest of the Cu atoms at the 6(h) positions. Note that our analysis clearly rules out Al at 4(f) sites. Since U_2Cu_9Al is another compound of the U–Cu–Al system, it is useful to compare its crystal structure with that of $UCu_{5-x}Al_x$ compounds, which are hexagonal too. The latter compounds crystallize in the $CaCu_5$ type of structure, in contrast to U_2Cu_9Al which crystallizes in a structure closely related to the $MgZn_2$-type structure. In both systems one finds statistically distributed Cu and Al atoms in planes containing no U atoms. However, in $UCu_{5-x}Al_x$ compounds there is no out-of-plane shift of Cu atoms from U–Cu planes.

3. Results

We measured the temperature dependence of the magnetic susceptibility $\chi = M/H$ and the magnetization curves between 2 K and 300 K in fields up to 5 T using a Quantum Design SQUID magnetometer. For these measurements, a finely powdered sample with grains fixed in a random orientation with diamagnetic glue was used. At around 31 K the magnetic susceptibility exhibits an anomaly which is indicative of magnetic order. A second anomaly occurs at around 9.5 K. While the anomaly at higher temperature does not exhibit any sizable field dependence, the lower anomaly shifts towards lower temperatures as a function of the field. The shift amounts to -0.26 K T^{-1} between 1 and 5 T, suggesting that at low temperatures U_2Cu_9Al has an antiferromagnetic (AF) ground state. Below 9.5 K the magnetic susceptibility exhibits a progressively larger dependence on the magnetic field, manifested as a splitting of the susceptibility curves (figure 1) for different fields. The $1/\chi$ versus T dependence above 175 K is linear and obeys a Curie–Weiss (CW) law (figure 2) with an effective magnetic moment of

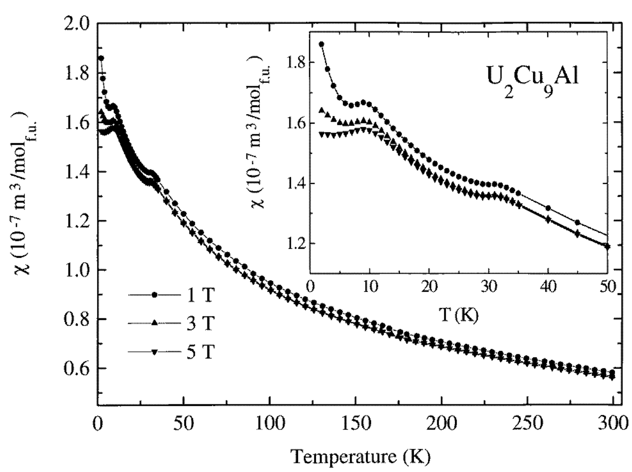

Figure 1. The temperature dependences of the magnetic susceptibility of U_2Cu_9Al in an M/H versus T representation measured in different magnetic fields. The low-temperature detail is shown in the inset.

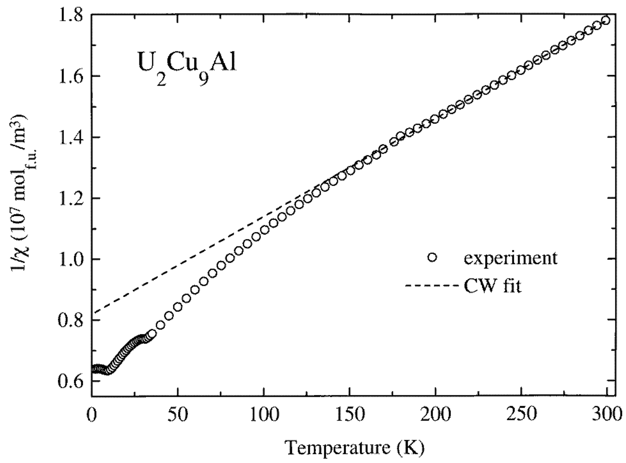

Figure 2. The temperature dependence of the inverse magnetic susceptibility of U_2Cu_9Al together with the best fits to a Curie–Weiss (CW) law denoted by a broken line.

$\mu_{eff} = 3.15 \pm 0.01\ \mu_B$/U and a paramagnetic Curie temperature of $\theta_P = -255.3 \pm 1.0$ K.

At low temperatures, below the second anomaly at 9.5 K, the χ versus T curves measured at low fields show a strong upturn, showing no tendency to saturation which is a hallmark of FL systems, suggesting that there is a NFL state. However, this upturn is gradually suppressed by the applied field (in higher fields, χ approaches a constant value χ_0 as T decreases, recovering its ordinary FL dependence), suggesting—rather than NFL behaviour—that some kinds of magnetic fluctuation are occurring in U_2Cu_9Al. Therefore, unlike for the case for the $UCu_{5-x}Al_x$ compounds [14, 17], we conclude from the temperature dependence of the magnetic susceptibility measured at low fields that no decisive evidence of NFL behaviour in U_2Cu_9Al has been found.

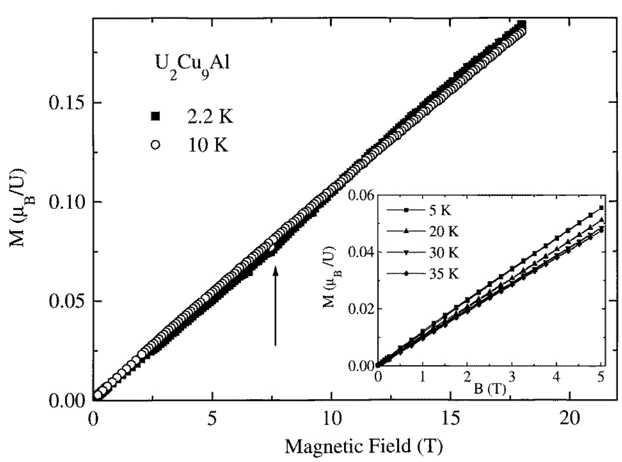

Figure 3. The field dependences of the high-field magnetization of polycrystalline U_2Cu_9Al at 2.2 K and 10 K. The field dependences of the low-field magnetization curves of U_2Cu_9Al measured at various temperatures for fixed powder up to 5 T are shown in the inset.

Low-field magnetizations, measured at various temperatures, are shown in the inset of figure 3. The magnetic response is almost linear at all temperatures, although a slight bending can be discerned for the lowest temperatures. Furthermore, we find very low magnetization values even in fields of 5 T (0.056 μ_B/U at 5 K) which indicates that the antiferromagnetic state still persists. Therefore, we extended our studies up to 18 T using a vibrating-sample magnetometer in the 20 T superconducting magnet at the Pulsed Field Facility of the NHMFL at Los Alamos National Laboratory. The magnetization curves taken for a 69 mg sample chunk at 2.2 K and 10 K are shown in figure 3. We do not find any evidence for a transition in fields up to 18 T in the 10 K data, while a broad S-shaped transition at around 7.5 T is visible at 2.2 K. The magnetization gain due to this transition amounts to 0.01 μ_B/U between 7.5 T and 18 T for the 2.2 K measurement. This suggests that the magnitude of the moments involved in this transition is very small. The magnetization curves do not saturate up the highest fields applied in the present experiment. Although a broad S-shaped transition visible at low temperatures suggests that a forced ferromagnetic state with small U moments exists in U_2Cu_9Al above 7.5 T, we cannot exclude the possibility that some other metamagnetic transition might occur at even higher fields, leading to higher and possibly the full free-ion values of the U magnetic moments.

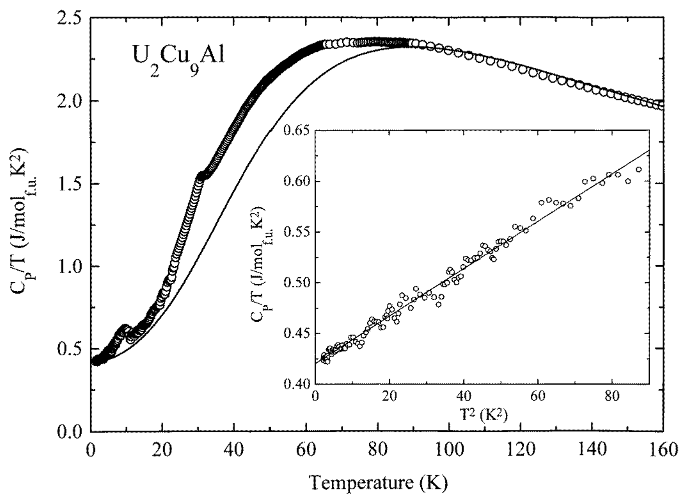

Figure 4. The temperature dependence of the specific heat in a C_p/T versus T representation for polycrystalline U_2Cu_9Al, together with the sum of the Debye function ($\theta_D = 320$ K) and the electronic term γ (full curve). In the inset, the low-temperature part of the specific heat in a C_p/T versus T^2 representation, together with the best fit (the line), is shown.

The specific heat was measured using two experimental set-ups by a standard semi-adiabatic method. The data taken in zero field in the temperature range 1.5–200 K are shown in figure 4. There are two clearly visible anomalies at around 9.5 and 31 K, which are indicative of magnetic phase transitions. The linear extrapolation of the low-temperature part of C_p/T versus T^2 to 0 K yields the low-temperature specific-heat coefficient $\gamma = 419 \pm 2.8$ mJ $\text{mol}_{\text{f.u.}}^{-1}$ K^{-2} (fitted up to 9 K—see the inset of figure 4). This value, which stands for two U atoms, is strongly enhanced with respect to that for simple metals and, therefore, U_2Cu_9Al can be classified as a moderately enhanced heavy-fermion system. In order to estimate the magnetic entropy associated with the magnetic phase transitions, we subtracted the contribution of the lattice using a Debye background with $\theta_D = 320$ K, which, in sum with the electronic term, agrees well with the experimental data in both the low- and high-temperature limits. By integrating C/T versus T up to 100 K, we obtain a value of $1.45R\ln 2$ for the magnetic entropy per U atom. This value suggests that 5f-electron states are rather itinerant in U_2Cu_9Al and/or that magnetic fluctuations play an important role.

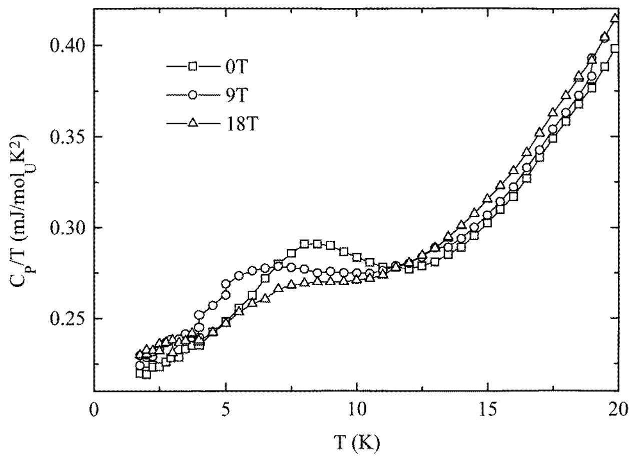

Figure 5. The low-temperature part of the specific heat of polycrystalline U_2Cu_9Al in a C_p/T versus T representation measured in zero field and at 9 and 18 T.

In figure 5, the low-temperature part of the specific heat of polycrystalline U_2Cu_9Al measured in zero field and in fields of 9 and 18 T produced by a superconducting magnet is shown. As can be seen, the low-temperature anomaly shifts with field first to lower temperatures (by -0.24 K T^{-1}, in good agreement with magnetic susceptibility measurements) and then to higher temperatures (in approximately the same, but positive, ratio). With increasing field the anomaly gets somewhat spread out. These results suggest that the ground state of U_2Cu_9Al is indeed AF and that above the S shape indicated by magnetization measurements, the lower transition behaves as a ferromagnetic transition. The striking fact is that the low-temperature specific-heat coefficient gets enhanced with increasing field. It amounts to 215.6 ± 0.4, 223.8 ± 0.7 and 228.4 ± 1.0 mJ mol_U^{-1} K^{-2} for 0, 9 and 18 T. The results obtained with the two set-ups agree well for the zero-field overlap region.

We see that the low-temperature part of the specific heat of U_2Cu_9Al exhibits a dependence typical for FL systems (i.e. C_p/T versus T^2 is a straight line—see the inset of figure 4). Although there exists a certain class of NFL materials (namely 3d antiferromagnets) for which C/T approaches a constant value [18] similarly to that of FL materials, we do not think that this scenario applies to our system in view of the expectation that only U atoms are magnetic in U_2Cu_9Al, with the d states filled.

The electrical resistivity was measured for a small bar-shaped sample by the standard AC four-point method in the 260 mK–290 K temperature range in zero field and in a field of 18 T. For the measurements below 2 K, we used a ^{3}He refrigerator. The electrical contacts were established using silver paint.

The temperature dependence of the electrical resistivity of U_2Cu_9Al is shown in figure 6. As can be seen, the zero-field curve is very unusual. Upon cooling from 290 K, it first increases with lowering temperature, reaches a maximum at around 60 K, and then starts to decrease. Around 32 K it again reaches a minimum, and it goes through another maximum and minimum at around 27 K and 16 K respectively. Finally, it increases again and reaches

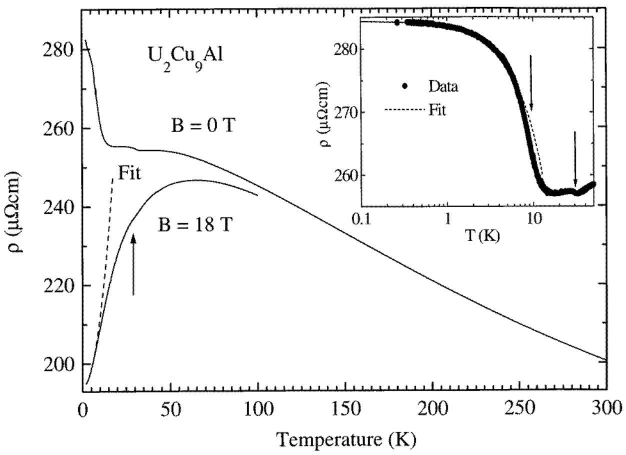

Figure 6. The temperature dependences of the electrical resistivity measured in zero field and in a field of 18 T. The arrow indicates a weak anomaly at around 31 K visible in the 18 T curve. The low-temperature part down to 0.26 K is displayed together with the best fit described in the text in a logarithmic representation in the inset and the transition temperatures determined from the specific heat are indicated by arrows.

a value of 284 $\mu\Omega$ cm at 260 mK. However, the absolute values of the electrical resistivity should be treated with caution, because the uncertainty in the geometrical factors can be as high as 10% due to internal microcracks. Clearly, no T^2-dependence is observed at low temperatures. Instead, the dependence can be approximated by $\rho(T) = \rho_0(1 - a(T/T_K)^n)$, which is used for the description of NFL systems [19]. The fitting parameters resulting from the fit between 0.26 K and 5 K are $\rho_0 = 284.4 \pm 0.1$ $\mu\Omega$ cm, $a = 0.130$, $T_K = 17.40$ K and $n = 1.33 \pm 0.01$. The magnetic phase transitions determined by means of the magnetic susceptibility and the specific heat are indicated by arrows in the inset of figure 6, where the low-temperature part of the electrical resistivity and the fit are shown. The transition temperatures can be clearly identified as the inflection points of the electrical resistivity upturns. The

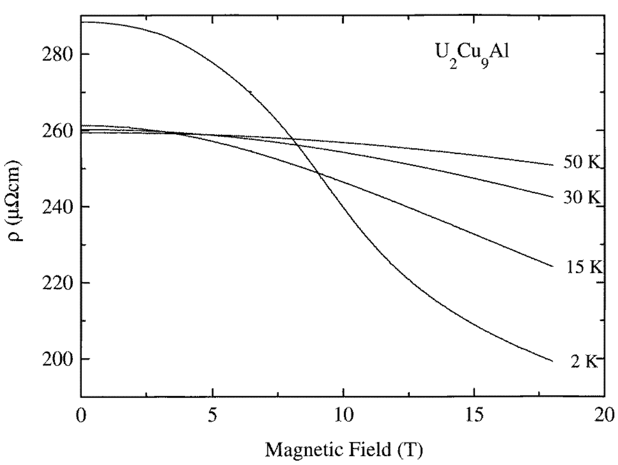

Figure 7. The field dependences of the electrical resistivity at different temperatures.

$\rho_{0.26\,K}/\rho_{290\,K}$ ratio amounts to 1.31. The values of both $\rho_{290\,K}$ and $\rho_{0.26\,K}/\rho_{290\,K}$ are unusually high for a metallic system (although rather common for U-based compounds), which suggests that the conduction electrons are strongly hybridized with 5f states. At the same time, a large portion of the scattering of the conduction electrons ceases upon application of a magnetic field. The temperature dependence of the electrical resistivity at 18 T exhibits at low temperatures the T^2-dependence expected for FL systems. The best fit to $\rho(T) = \rho_0 + aT^2$ between 2 and 6 K yields $\rho_0 = 194.3 \pm 0.1\ \mu\Omega$ cm and $a = 0.174 \pm 0.001\ \mu\Omega$ cm K^{-2}. As can also be seen from figure 6, while the lower anomaly is not present, the anomaly at higher temperature is still visible (as a small bump at around 30 K) with its position hardly affected by the magnetic field.

Shown in figure 7 are the field dependences of the electrical resistivity at various temperatures. The electrical resistivity decreases slowly at all temperatures with increasing field. For the measurement at 2 K, the reduction amounts to 31% of its zero-field value, suggesting that a large part of the resistivity can be attributed to AF scattering.

4. Discussion and conclusions

The electronic structure of actinide systems is characterized by partially occupied 5f-electron states which have a large spatial extent. Therefore, the 5f electrons in actinides are usually strongly delocalized due to their participation in bonding, and there is considerable hybridization of the 5f states with the valence states of neighbouring atoms (5f–ligand hybridization) and conduction electrons. Consequently, one observes reduced magnitudes of magnetic moments. Highly correlated 5f-electron states are expected to form a band pinned at the Fermi surface, which is reflected in enhanced γ-values of the low-temperature specific heat and highly anomalous magnetic and transport properties.

U_2Cu_9Al exhibits an enhanced low-temperature specific-heat coefficient (namely γ = 210 mJ $mol_U^{-1}K^{-2}$) which classifies this compound as a moderately enhanced heavy-fermion system and can be viewed as a consequence of the presence of correlated 5f-electron states at E_F, possibly combined with competing magnetic interactions. At the same time it is quite clear that 5f states are strongly hybridized with other electronic states in the solid, presumably with conduction states and Cu 3d and Al 2p ligand states. Consequently, the U moments involved in the magnetic ordering are small. On the other hand, we cannot discard the possibility that the enhancement originates partly from the randomness in the occupation of the 6(h) sites by Cu and Al atoms. Although the U sublattice is intact (no randomness is found at the 2(a) sites, where U atoms are situated) and even the nearest sites, 4(f), are occupied only by Cu atoms, Gschneidner *et al* [20] have argued that the enhancements of the low-temperature specific heat can be due to randomness in the crystal lattice at non-magnetic sites, which promotes some randomness in the exchange interactions and may eventually lead to the formation of a spin-glass state. The spin-glass behaviour arising from non-magnetic atomic disorder may cause a large enhancement of the low-temperature specific heat, and these materials should not be categorized as 'real' heavy-fermion compounds. However, in U_2Cu_9Al the Cu_2Al planes, where the randomness is found, are separated from the U planes by Cu layers with no structural disorder, and randomness is not likely to be found in exchange interactions. We also did not find any evidence for spin-glass behaviour in U_2Cu_9Al.

Special conditions in the vicinity of E_F may lead under certain circumstances [1] to a NFL behaviour. While the low-temperature electrical resistivity in zero field resembles the behaviour of $Y_{0.8}U_{0.2}Pd_3$ [19] and can be interpreted in the NFL framework with a critical temperature of 17 K, the low-temperature specific heat exhibits the temperature dependence expected for FL materials. We think that U_2Cu_9Al does not belong to a class of materials showing NFL behaviour and exhibiting a low-temperature specific heat proportional to the temperature (3d antiferromagnets) [18], and we therefore conclude that, unlike the case for Al-rich UCu_xAl_{5-x} compounds [14, 17], no unambiguous indication for NFL behaviour has been found for U_2Cu_9Al on the basis of specific-heat measurements.

The electrical resistivity in a field of 18 T varies as T^2. Therefore the unusual temperature dependence at zero field is due to AF order or fluctuations or overdamped Kondo screening in U_2Cu_9Al which is at least partially removed by application of a magnetic field. The first possibility is supported by the fact that the low-temperature specific-heat coefficient γ increases with magnetic field (by 5% of its zero-field value in 18 T) suggesting that a modification of the Fermi surface takes place with application of a magnetic field. On the other hand, the relatively small change in γ (+5%) compared with the large change in the electrical resistivity (−31%) implies not only the partial disappearance of the Fermi surface due to AF superzone gapping, but also the reduction of the Fermi velocity around the gapping plays an important role in U_2Cu_9Al. Similar behaviour has been observed for, e.g., UNiGa [21].

To be able to make a comparison of U_2Cu_9Al with other heavy-fermion systems, we calculate the Wilson ratio [22] $R_W = 1.30$ and the Kadowaki–Woods ratio [23] $a_\rho/\gamma^2 = 0.38 \times 10^{-5}$ $\mu\Omega$ cm mJ^{-2} mol^2 K^2. In our calculations we put $\chi_0(B = 5$ T, $T \rightarrow 0) = 1.56 \times 10^{-7}$ m^3 mol^{-1} and $\gamma(B = 0$ T$) = 210$ mJ mol^{-1} K^{-2} for R_W and $\gamma(B = 18$ T$) = 214$ mJ mol^{-1} K^{-2} and $a_\rho(B = 18$ T$) = 0.174$ $\mu\Omega$ cm K^{-2}. The former coefficient, R_W, is expected to be of the order of unity for a free-electron gas, and it can be enhanced by a factor of two due to electron–electron correlations if the enhancements of χ_0 and γ have the same origin. The latter coefficient should be close to a universal value of 1.0×10^{-5} $\mu\Omega$ cm mJ^{-2} mol^2 K^2 if the same quasiparticles participate in both the electrical conduction and the low-temperature specific-heat properties. From these results one can conclude that if such an approach can be used for U_2Cu_9Al then (i) the electron states that are responsible for magnetic and specific-heat properties are correlated and (ii) at 18 T the electrical conduction and the low-temperature specific-heat properties are not due to the same type of quasiparticle, which implies the existence of two electron subsystems. This however does not rule out strong hybridization of conduction electrons with other states in zero field.

In conclusion, the compound U_2Cu_9Al crystallizes in the hexagonal structure (space group $P6_3/mmc$). It orders magnetically below 31 K with an additional magnetic phase transition at 9.5 K. Magnetization measurements show that the lower phase transition in U_2Cu_9Al is AF in origin. The high-temperature part of the magnetic susceptibility is described well by a Curie–Weiss law with an effective moment of 3.15 μ_B/U. However, the low-temperature behaviour cannot be described in a localized picture. Magnetization, susceptibility and specific-heat measurements suggest that the magnetism in U_2Cu_9Al is governed to a certain extent by delocalized uranium 5f-electron states (with possible fluctuations) confined in a band which is hybridized with conduction electrons. The high-field magnetization at low temperatures reveals a weak step in the magnetization, which is tentatively attributed to the destruction of ground-state AF order of small U moments. The resistivity behaviour furthermore suggests that the strong hybridization between the conduction and 5f-electron subsystems is at least partially destroyed by magnetic fields.

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