Z. Phys. B - Condensed Matter 83, 305-306 (1991)

Condensed
Zeitschrift für Physik B Matter

Rapid note

A new heavy-fermion superconductor: UNi_2Al_3

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Received February 18, 1991

Measurements of the resistivity, susceptibility and specific heat are reported which clearly show that the hexagonal compound UNi_2Al_3 is a heavy-fermion magnet below $T_N = 4.6$ K and a heavy-fermion superconductor below $T_c = 1$ K.

Superconductivity of strongly correlated carriers has attracted much interest during the past years. While in the perowskite-derived cuprates, superconductivity at $T_c \cong 100$ K [1] was discovered close to a metal-insulator transition, in the heavy-fermion intermetallic compounds $CeCu_2Si_2$ [2], UBe_{13} [3], UPt_3 [4] and URu_2Si_2 [5] a superconducting phase transition at $T_c \cong 1$ K takes place in the vicinity of a magnetic-nonmagnetic transition which reflects a weak delocalization of $4f$- and $5f$-electrons, respectively. A wide variety of novel phenomena, unknown to classical superconductors, have been established in these "heavy-fermion superconductors" (HFS), e.g. Cooper pairs formed by carriers with effective mass of order $100 m_{el}$, anisotropic superconducting order parameters, multiple superconducting phase diagrams and coexistence of superconductivity with long-range antiferromagnetic order in the heavy-fermion system [6]. The fact that the four HFS behave in part distinctly differently from each other, e.g. concerning the sensitivity of their superconducting properties to the quasiparticle mean free path, has so far prevented a unified microscopic description of this phenomenon [6]. However, in spite of intensive efforts, any attempts to find further examples of HFS have been unsuccessful since 1984. In this *Rapid note* we communicate the discovery of a new heavy-fermion superconductor: UNi_2Al_3.

Samples were prepared by melting together in an arc furnace under Ar atmosphere appropriate amounts of U (depleted, Merck), Ni (4N8, Johnson Matthey) and Al (5N, Heraeus): For a new series of samples, natural Uranium (4N, CENG, Grenoble) is being used. As-cast samples contain a few percent of secondary phases (mainly UAl_2) which disappear after a suitable heat treatment (1000 °C, 5 days). X-ray powder diffractometry on annealed samples shows reflexions only of the proper hexagonal $PrNi_2Al_3$ structure, with lattice parameters $a = 5.207$ Å and $c = 4.018$ Å. This yields a large U–U separation of 4.018 Å, in fact prerequisite for heavy-fermion behavior.

Measurements of the electrical resistivity, $\rho(T)$, *dc*- and *ac*-susceptibility, $\chi(T)$, and specific heat, $C(T)$, (Figs. 1-3) were performed using standard techniques.

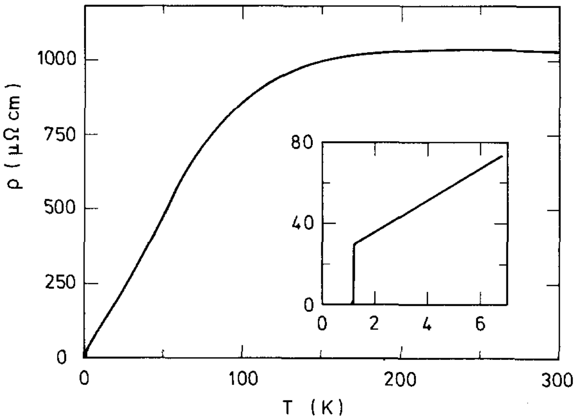

Fig. 1. Electrical resistivity, ρ, as a function of temperature for UNi_2Al_3. Owing to a large number of microcracks in the sample, data points represent an upper limit of ρ *vs* T. It is estimated that true values are lower by a factor of order five. Inset shows data below 7 K

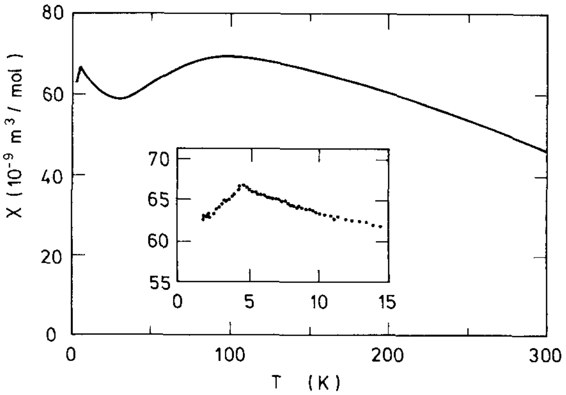

Fig. 2. Temperature dependence of the *dc*-susceptibility, χ, as measured for UNi_2Al_3 in a field of 20 mT using a SQUID magnetometer. Inset displays the antiferromagnetic transition at $T_N = 4.6$ K

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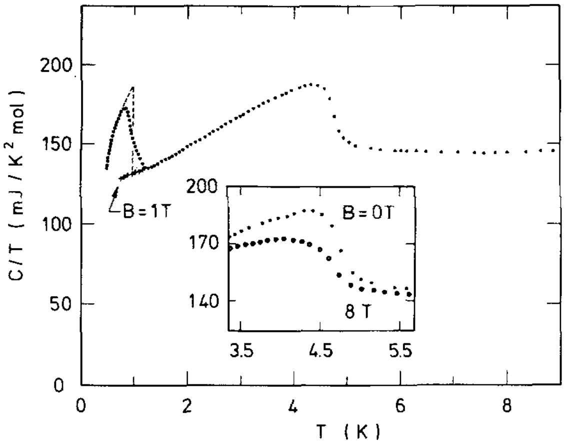

Fig. 3. Specific heat of UNi_2Al_3 in a plot C/T *vs* T between 0.5 K and 9 K. $B = 1$ T data shown represent normal state. Dashed lines are used to replace, under conservation of total entropy, broadened anomaly at T_c by idealized jump. Inset shows slight shift of antiferromagnetic transition in a field of 8 T

Three kinds of anomalies can be discerned in the zero-field data: (1) A flattening of the ρ *vs* T curve and a broad maximum in χ *vs* T near 100 K, (2) a cusp in the *dc*-susceptibility, χ *vs* T, and a somewhat broadened mean-field-type discontinuity in C *vs* T at 4.6 K, (3) a sharp drop to an unresolvably small value in the resistivity, a change of sign to a diamagnetic signal in the *ac*-susceptibility (not shown) and a broadened "jump" in the specific heat in the temperature window 0.8–1.2 K.

Although a Curie-Weiss behavior of the susceptibility is not observed below 300 K, both the absolute value and the slope of $\chi(T)$ at room temperature are typical for localized $5f$ electrons. The χ *vs* T maximum at 100 K has its correspondence in a maximum of the U-increment to the resistivity, which would show up after subtraction of the electron-phonon part to $\rho(T)$. These anomalies suggest a transition from localized to weakly delocalized $5f$-electron behavior in the spirit of the Kondo-lattice concept, with a somewhat higher characteristic temperature than e.g. for UPt_3 [6]. We note, however, that an additional low-energy scale may be reflected by a shallow maximum in the electrical resistivity near 15 K and the nearly linear $\rho(T)$ dependence between 1.2 K and 7 K (Fig. 1). The latter is often found in heavy-fermion compounds preceding the asymptotic low-T behavior, $\rho \sim T^2$, of the coherent Fermi liquid. Additional support for the existence of such a low-energy scale may derive from the rather flat C/T *vs* T dependence between 5 K and 9 K, which points to an increase of the *electronic* part, C_{el}/T *vs* T, upon lowering T.

We ascribe the anomalies in the *dc*-susceptibility (Fig. 2) and specific heat (Fig. 3) at $T_N = 4.6$ K to antiferromagnetic ordering between the U-derived $5f$-moments. The Néel temperature is only weakly depressed by an external field as high as 8 T, resembling the case of e.g. the heavy-fermion antiferromagnet $CeCu_2Ge_2$ [7]. The low value of the magnetic entropy, i.e. 0.13 Rln2, released at T_N hints at a rather small ordered U moment. The electronic specific-heat coefficient γ can be obtained in Fig. 3 by extrapolation of the $B = 1$ T data to $T = 0$ K. $\gamma = 120\,\mathrm{mJ/K^2}$ mole is of the typical order of magnitude for heavy-fermion magnets. For example, $\gamma = 250\,\mathrm{mJ/K^2}$ mole and $\gamma = 65\,\mathrm{mJ/K^2}$ mole have been determined for UCd_{11} ($T_N = 5$ K [8]) and URu_2Si_2 ($T_N = 17$ K [5]), respectively.

The third type of anomalies observed near 1 K indicates a superconducting transition. The size of the diamagnetic signal in the *ac*-susceptibility is the same as for a bulk superconductor. *dc*-magnetization measurements to determine the Meissner effect are in preparation. For the idealized specific-heat jump (dashed line in Fig. 3) we find $\Delta C/C_n(T_c) \cong 0.4$, a ratio that could recently be raised to 0.5 by using purer starting U material. This number must be considered a lower limit for $\Delta C/C_{el}(T_c)$, simply because $C_n(T)$ as measured at $B = 1$ T contains a magnetic (spinwave-derived) contribution in addition to the electronic part, $C_{el}(T)$. The scaling of ΔC with the large electronic specific heat clearly shows that superconductivity is a bulk property of UNi_2Al_3 and, most importantly, is a property of the strongly renormalized electronic system. An order-of-magnitude estimate of the effective carrier mass can be derived from the slope of the upper critical field at T_c, $B'_{c2} = -(\partial B_{c2}/\partial T)_{T_c}$, as determined via inductive transitions. Considering UNi_2Al_3 as being close to the "clean limit" (cf. caption of Fig. 1), using an isotropic one-band model, assuming that the Fermi wave number is the same as in UPt_3 and comparing our experimental result, $B'_{c2} = 1.4$ T/K, with the literature value for UPt_3, $B'_{c2} = 4 - 6$ T/K [9], we estimate from $(m^*/m_0)_{UPt_3} \cong 200$ that this ratio is about 70 in the case of UNi_2Al_3.

To summarize, the results presented above characterize UNi_2Al_3 as a new heavy-fermion compound showing both antiferromagnetic order and superconductivity. Since we have found an extremely strong T_c dependence on changes of the stoichiometry, our present efforts are devoted to optimize the annealing conditions in order to obtain homogeneous samples of stoichiometric composition. A more complete study of these samples will be described in a subsequent paper.

This work is part of the European-Japanese Joint Project on Heavy Fermions sponsored by NEDO and chaired by Prof. T. Kasuya. We are grateful for the opportunity to participate in this project. Financial support by the Deutsche Forschungsgemeinschaft under the auspices of the SFB 252 Darmstadt/Frankfurt/Mainz is also acknowledged. The work of one of us (D.K.) has been kindly supported by the Alexander-von-Humboldt Foundation.

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