Evidence for Multiband Superconductivity in the Heavy Fermion Compound UNi_2Al_3

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Epitaxial thin films of the heavy fermion superconductor UNi_2Al_3 with $T_c^{max} = 0.98$ K were investigated. The transition temperature T_c depends on the current direction which can be related to superconducting gaps opening at different temperatures. Also the influence of the magnetic ordering at $T_N \simeq 5$ K on $R(T)$ is strongly anisotropic, indicating different coupling between the magnetic moments and itinerant charge carriers on the multisheeted Fermi surface. The upper critical field $H_{c2}(T)$ suggests an unconventional spin-singlet superconducting state.

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Recently, clear progress concerning the superconductivity of the heavy fermion compound UPd_2Al_3 was reported. Because of combined experimental investigations based on tunneling spectroscopy on planar thin film junctions [1] and inelastic neutron scattering [2–4], an unconventional mechanism of superconductivity was identified. There is compelling evidence that the Cooper pair formation in this compound is caused by the exchange of magnetic excitations. However, the question whether these excitation are spin fluctuations [5] or magnetic excitons [4,6] is not finally answered. In this framework the investigation of the compound UNi_2Al_3, which is isostructural to UPd_2Al_3, is of great interest. Whereas UPd_2Al_3 shows a simple antiferromagnetic structure with relatively large ordered magnetic moments of $\mu_{ord} \simeq 0.85\mu_B$, the compound UNi_2Al_3 is an incommensurately ordered antiferromagnet [7] with a smaller ordered moment $\mu_{ord} \simeq 0.24\mu_B$ [8]. In contrast to UPd_2Al_3, for UNi_2Al_3 there is evidence for a spin-triplet superconducting state [9].

The preparation of the first single crystalline bulk samples of UNi_2Al_3 [10,11] was not possible until five years after the discovery of superconductivity in this compound [12]. This is due to a peritectical decomposition, which results in the formation of an UAl_2 impurity phase [13] and hence a strongly reduced sample purity compared to UPd_2Al_3. However, since thin films are not prepared from the melt, this typical problem can be avoided. We prepared superconducting single crystalline thin films of UNi_2Al_3 (typical thickness $d \simeq 200$ nm) by coevaporation of the elementary components in a molecular-beam epitaxy system. Orthorhombical $YAlO_3$ cut in the (010) or (112) direction provides an epitaxial substrate for the (100)-oriented UNi_2Al_3 with lattice misfits for the a axis of $+0.7\%$ for the former and -0.5% for the latter substrate cut. On these substrates high purity single crystalline thin films were prepared. In Fig. 1 an x-ray $\Theta/2\Theta$ scan of an UNi_2Al_3 (100) film on $YAlO_3$ (010) is shown. Only a very weak UO_x but no UAl_2 impurity phase is visible. The inset shows a scan of the crystallographic $(1KL)$ plane of UNi_2Al_3. The observed x-ray reflections prove that the film is ordered in plane, i.e., single crystalline. From the positions of 14 x-ray reflections of an UNi_2Al_3 thin film, the following lattice parameters were calculated: $a = 0.5211(5)$ nm, $b = 0.5221(8)$ nm, and $c = 0.4017(2)$ nm. These numbers are in agreement with the values obtained from polycrystalline bulk samples [12] and rule out any growth process induced stress of the films.

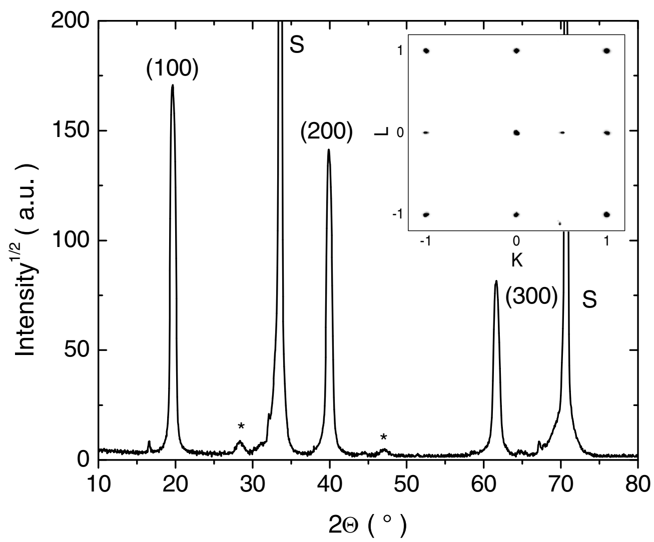

FIG. 1. X-ray $\Theta/2\Theta$ scan of an UNi_2Al_3 thin film on an $YAlO_3$(010) substrate. Please note the alignment of the reciprocal (100) axis of the hexagonal crystal perpendicular to the substrate surface. [UNi_2Al_3 reflections are labeled (H00), substrate S, impurities * (presumably UO_x)]. The inset shows a scan of the reciprocal $(1KL)$ plane of UNi_2Al_3. The x-ray intensity (linear gray scale) is plotted versus the momentum transfer in units of the reciprocal lattice vectors. From the observation of reflections at integers K and L values the in-plane order of the film is obvious. The narrow reflections close to (1 1/2 $-$1) and (1 1/2 0) are substrate reflections.

Patterning the samples by a standard photolithographic process with ion beam etching made it possible to obtain well defined geometries for the investigation of anisotropic transport properties (see inset of Fig. 2). Measurements of the temperature dependent resistivity $R(T)$ of the samples were performed with the current direction parallel to the crystallographic a axis as well as parallel to the c axis of the same thin film. A pronounced anisotropy, like that reported previously from bulk single crystals [14], is observed (Fig. 2). The residual resistivities of the best samples are $\rho_c = 25\ \mu\Omega$ cm for $I \parallel c$ and $\rho_a = 20\ \mu\Omega$ cm for $I \parallel a$. The residual resistance ratios $\mathrm{RRR} = R_{300\,\mathrm{K}}/R_{1.1\,\mathrm{K}}$ amount to $\mathrm{RRR}_{I\parallel c} = 6.6$ and $\mathrm{RRR}_{I\parallel a} = 8.3$. This is approximately half of the RRR reported for bulk single crystals [10,14], indicating a higher concentration of scattering centers in the thin films. However, these scattering centers are less effective in destroying the superconducting state since the transition temperature T_c and width ΔT_c of the films is as high and sharp as in the best bulk single crystals.

The films become superconducting at $T_c^{\max} = 0.98$ K with resistive transition widths $\Delta T_c \simeq 0.06$ K. Large critical current densities of $I_c \simeq 10^4\ \mathrm{A/cm^2}$ at $T = 0.32$ K for $I \parallel a$ and $I \parallel c$ show that the observed superconductivity is a property of the complete thin film volume. The probe current employed for measurements of T_c was adjusted to a current range where no influence of its magnitude on the resistive transition was observed (Fig. 3). All of our superconducting samples on both kinds of substrate show the superconducting transition for current direction $I \parallel c$ at a reduced temperature compared to $I \parallel a$ with $T_c(I \parallel a) - T_c(I \parallel c) \simeq 0.05$ K (Fig. 3). The same behavior is visible at a close inspection of the only published $R(T)$ data of bulk single crystals by Sato *et al.* [10]. Thus, there is strong evidence that the directional dependence of the resistive T_c is an intrinsic property of UNi_2Al_3.

A possible explanation for this effect is the existence of two bands in UNi_2Al_3 with intraband pairing interactions but without superconducting interband interaction. Such a situation can result in two superconducting gaps on the Fermi surface which open at different temperatures [15]. Up to now there is no Fermi surface calculation of UNi_2Al_3 available, but similarities to the Fermi surface of the isostructural compound UPd_2Al_3 [16,17] can be assumed based on early band structure calculations [18]. Thus, a multisheeted Fermi surface is expected with an increased anisotropy compared to UPd_2Al_3 as concluded from the stronger transport anisotropy of UNi_2Al_3. On such a Fermi surface superconducting gaps opening at different temperatures on different sheets could result in transition temperatures which depend on the current direction.

According to Ref. [15], a weak coupling between the two bands results in two gaps with a common T_c. However, in one of the bands an initially tiny energy gap is expected which opens drastically at a reduced temperature compared to T_c. In a transport experiment such a situation is presumably indistinguishable from the case of two different T_c values due to critical current effects hiding the tiny energy gap. However, the simple model of Ref. [15] presents only a first attempt to explain

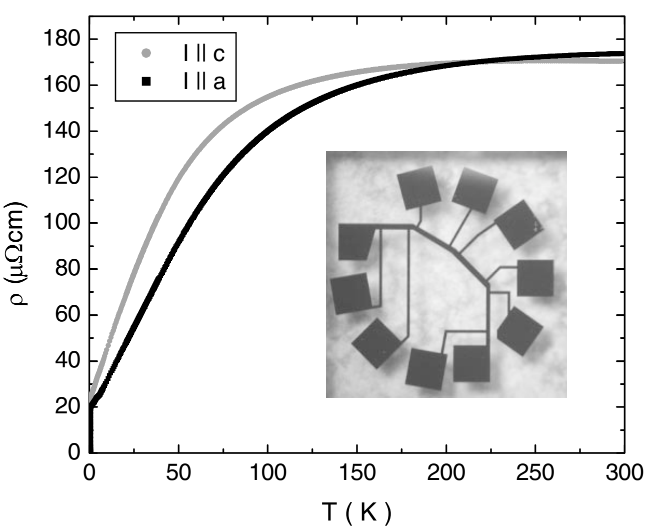

FIG. 2. Specific resistivity $\rho(T)$ of an UNi_2Al_3 thin film for different current directions (black squares: $I \parallel a$; gray dots: $I \parallel c$). Inset: Photograph of a patterned UNi_2Al_3 film ($4 \times 4\ \mathrm{mm^2}$). For $R(T)$ measurements a current is sent through the central conductor path (width $d = 100\ \mu$m), and the thin strips are voltage probes (4-point dc method).

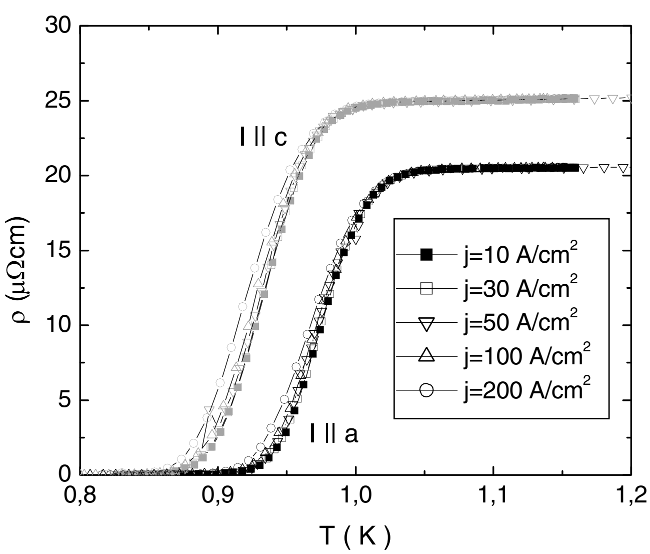

FIG. 3. Superconducting resistive transitions of an UNi_2Al_3 thin film for different current directions (black symbols: $I \parallel a$; gray symbols: $I \parallel c$). Measurements with different current densities are shown to demonstrate that the probe current does not influence the T_c values (for $j \simeq 10\ \mathrm{A/cm^2}$).

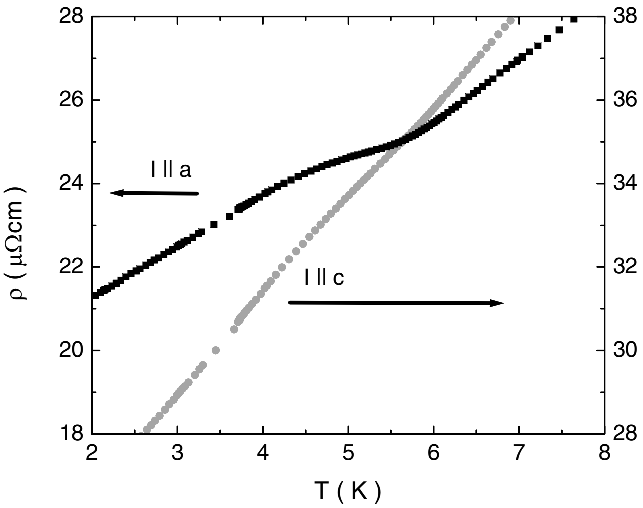

FIG. 4. Specific resistivity $\rho(T)$ of an UNi_2Al_3 thin film. Left axis: $I \parallel a$ (black squares). Right axis: $I \parallel c$ (gray dots).

the directional dependence of T_c and does not consider the coupling between superconductivity and magnetism as well as Fermi surface and order parameter anisotropies of UNi_2Al_3.

At the magnetic ordering temperature $T_N \simeq 5$ K a clear anomaly in the resistivity $R(T)$ of the films is visible for currents $I \parallel a$ but not for $I \parallel c$ (Fig. 4). The influence of the magnetic ordering on the resistivity of UNi_2Al_3 is different from the behavior of UPd_2Al_3 where a clear steepening of $R(T)$ is observed on cooling beyond T_N for $I \parallel a$ as well as for $I \parallel c$ [19]. In contrast, in the UNi_2Al_3 films we observe a flattening of $R(T)$ for $I \parallel a$ at T_N which is similar to the behavior of URu_2Si_2 [20]. This dependence can result from a change in the Fermi surface topology associated with the formation of a magnetization-density wave which opens a gap over a portion of the Fermi surface. Since $R(T)$ of UNi_2Al_3 is not affected by the magnetic ordering for currents $I \parallel c$, we conclude that only the sheet of the Fermi surface providing the a-axis transport couples to the magnetic order parameter.

The observation that the higher superconducting T_c was observed on the Fermi surface sheet which is more affected by the magnetic ordering provides evidence that the Cooper pairing in UNi_2Al_3 is mediated by magnetic excitations. However, because of the different influence of the magnetic ordering on the transport properties, it is possible that the character of these excitations is different from the one discussed for UPd_2Al_3.

To investigate the superconducting order parameter concerning a possible spin-triplet state, the upper critical field $H_{c2}(T)$ of the UNi_2Al_3 thin films was determined by measuring resistive transitions $R(T)$ in different magnetic fields using a midpoint criterion. Figure 5 shows the $R(T)$ curves obtained for probe currents $I \parallel a$ and field orientation $H \parallel c$ as an example. Please note that the width and shape of the transitions depend only weakly on the field H as opposed to the behavior reported from bulk single crystals [10]. The upper critical field was determined for field directions parallel to the real space a and c axes as well as parallel to the reciprocal a^* axis of the hexagonal compound (Fig. 6). The measured $H_{c2}(T)$ curves are almost independent of the current direction. Only a small shift to lower temperatures of the data obtained with $I \parallel c$ compared to $I \parallel a$ was observed as expected considering the reduced T_c in this direction. For $H \parallel a^*$ the film

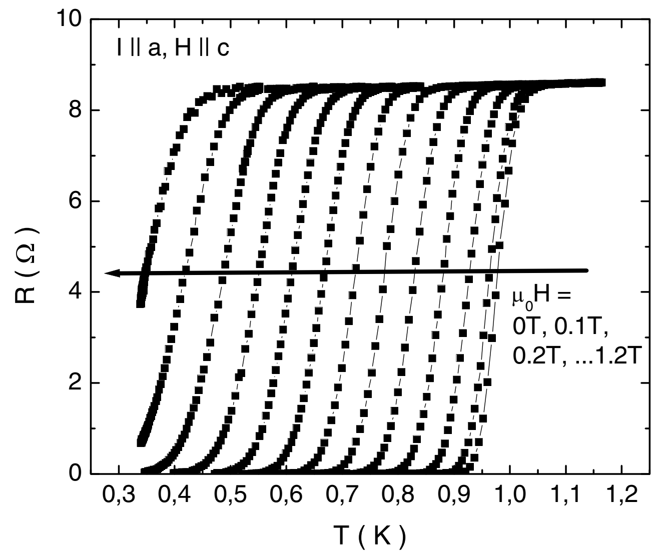

FIG. 5. Resistive superconducting transitions of an UNi_2Al_3 thin film in magnetic fields $\mu_0 H$ from 0 to 1.2 T. Probe current $I \parallel a$ and fields $H \parallel c$.

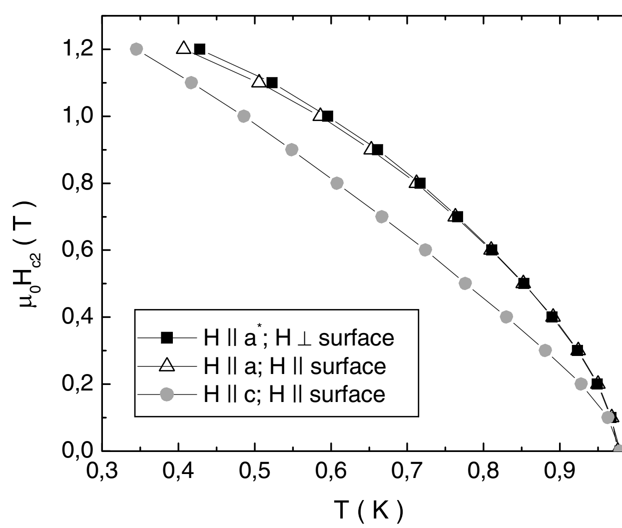

FIG. 6. Upper critical fields $\mu_0 H(T)$ of an UNi_2Al_3 thin film with different orientations relative to the field direction (see inset). Current $I \parallel a$ axis.

plane was perpendicular to the field direction, whereas for $H \parallel c$ the thin films had to be mounted with the film surface parallel to the field direction. In principle, the parallel configuration allows the appearance of increased critical fields due to finite size effects (thin film and surface superconductivity) [21]. However, these phenomena should result in a pronounced angular dependence of $H_c(\Theta)$, which was not observed. For additional investigation of these possible effects, the film was rotated by 90° on the sample platform resulting in a configuration with $H \parallel a$ parallel to the film surface. Because of the hexagonal symmetry of UNi_2Al_3, only a very weak anisotropy of the critical field is expected within the *ab* plane. Thus, the observation of very similar $H_{c2}(T)$ curves for $H \parallel a$ parallel to the film surface and for $H \parallel a^*$ perpendicular to the film surface shows that finite size effects do not influence the critical field.

For all investigated magnetic field directions a steep slope $-\mu_0 H'_{c2}(T_c) > 5T/\mathrm{K}$ was observed in contrast to the much smaller slopes reported previously from investigations of bulk polycrystals [22] and single crystals [10]. Additionally, the critical fields of the thin films are much larger and less anisotropic than reported for the only available bulk single crystals [10]. Employing the conventional Werthamer-Helfand-Hohenberg-Maki (WHHM) theory [23] of the upper critical field, the authors concluded that orbital pair breaking limits H_{c2} of UNi_2Al_3. With the same approach [24] we obtain from

$$H_{c2}^{\mathrm{orb}} = 0.693((-dH_{c2}/dT)_{T_c})T_c$$

an orbital upper critical field of $\mu_0 H_{c2}^{\mathrm{orb}} > 3.5$ T. However, from our $H_{c2}(T)$ curves we estimate $\mu_0 H_{c2}(0\ \mathrm{K}) \simeq 1.6$ T only. This discrepancy provides evidence for a non-negligible paramagnetic pair breaking contribution and thus a spin-singlet superconducting order parameter of UNi_2Al_3 as opposed to the experimental evidence for spin-triplet superconductivity of Ref. [9].

Additional conclusions concerning the order parameter symmetry cannot be drawn from the $B_{c2}(T)$ measurements. For a realistic description a concept that goes beyond the WHHM theory considering the presumably multisheeted anisotropic Fermi surface and strongly anisotropic order parameter of UNi_2Al_3 is needed.

We arrive at the conclusion that epitaxial thin films of UNi_2Al_3 allow the identification of superconducting transition temperatures T_c which clearly depend on the current direction. In conjunction with anisotropic signatures of the magnetic ordering, these measurements provide evidence for a multiband superconducting state with a magnetic Cooper-pairing mechanism. Measurements of the upper critical field indicate a spin-singlet superconducting state of UNi_2Al_3.

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