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SINGLE CRYSTAL NEUTRON DIFFRACTION STUDIES OF THE HEAVY FERMION SUPERCONDUCTOR UPd_2Al_3

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The magnetic properties of the heavy Fermion superconductor UPd_2Al_3 are studied by elastic neutron diffraction experiments. In the vicinity of T_N the temperature dependence of the ordered moments indicates mean field behavior. Anomalies of the staggered magnetization are found at the onset of superconductivity.

HEAVY FERMION superconductors (HFS) continue to attract considerable interest due to their fascinating properties. For example, the crucial interplay between magnetism and superconductivity is far from being understood. The discovery of two new magnetic HFS, namely UM_2Al_3 (M = Pd, Ni) by Geibel *et al.* [1, 2] provides a further impact in the field. Here we shall report on the results of recent neturon-diffraction studies on UPd_2Al_3 single crystals. The samples were prepared by the Czochralski pulling method. A detailed description is given in [3]. Concurrently two neutron scattering investigations were carried out. One of these experiments has been performed on a large UPd_2Al_3 single crystal with a volume of $0.4\,cm^3$ on the triple axis spectrometer TOPAN at the JRR3M reactor at Tokai, Japan. The crystal was oriented with the (1 0 0)- and (0 0 1)-axis in the scattering plane. Collimations of $60'/30'/30'/60'$ and an incident neutron wavelength of 2.438 Å were used. A pyrolytic-graphite filter was inserted into the outgoing neutron beam to reduce higher-order contaminations. The other neutron diffraction study was carried out on the two axis spectrometer P2AX at the Saphir reactor of the PSI at Villigen, Switzerland. The highest flux was obtained by selecting a neutron wavelength of $\lambda = 1.03$ Å. the collimation was $60'/30'/60'$ and a plutonium filter was used to avoid higher-order contributions. Here a single crystal of about $0.1\,cm^3$ was oriented with the (1 1 0)- and (0 0 1)-axis in the scattering plane. The superconducting transition temperature in both samples was determined as $T_c = 1.8$ K.

Magnetic intensities were observed for the (0 0 1/2), (0 0 3/2), (0 0 5/2), (1 1 1/2) and (2 0 1/2) and magnetic Bragg peaks. These results are in agreement with antiferromagnetic order with a propagation vector $\mathbf{q} = (0, 0, 1/2)$ and the magnetic moments oriented perpendicular to the c-axis in the easy plane as reported earlier [4]. The Néel temperatures of the two samples were found to be slightly different, 14.3 K and 14.5 K for the experiments performed in Japan and Switzerland, respectively. The upper part of Fig. 1 shows the temperature dependence of the reduced ordered moment μ/μ_0 per U atom of UPd_2Al_3 for the strongest magnetic Bragg peak (0 0 1/2). No corrections for extinction were applied. This seems to be justified due to the fact that different crystals of different size were investigated using various orientations and different neutron wavelengths but resulting in the same magnetization curve. From a previous powder diffraction study the ordered magnetic moment has been determined to amount $\mu_0 = 0.85\mu_B/U$ [4]. The magnetic phase transition is close to second order. In contrast to the neutron-powder diffraction study [4], no incommensurate phase above the antiferromagnetic ordering temperature could be detected in either of the single-crystal experiments. These observations indicate a critical dependence of the magnetic properties on sample preparation procedures. Similar effects have been found in $CePd_2Al_3$ [6]. Here the polycrystalline samples revealed magnetic order below $T_N = 2.6$ K while in single crystalline material no order could be detected down to the lowest temperatures [6].

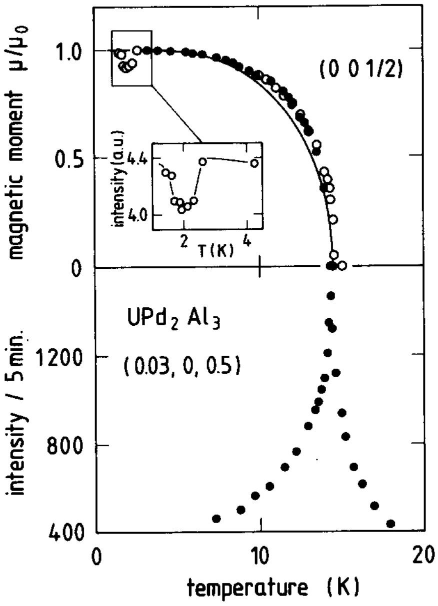

Fig. 1. Upper part: Temperature dependence of the reduced magnetic moment for the strongest magnetic Bragg peak (0 0 1/2) of UPd_2Al_3. For comparison, a $S = 1/2$ Brillouin function is plotted as well (full line). Lower part: Temperature dependence of the diffuse critical scattering measured at (0.3, 0, 1/2). Open circles and full triangles correspond to the results of the experiments performed in Switzerland and Japan, respectively. The inset shows the temperature dependence of the integrated intensity of the (0 0 1/2) peak in UPd_2Al_3 in the vicinity of the superconducting transition temperature. The line is drawn to guide the eye.

A further interesting aspect is an anomaly in the intensity and width of the magnetic Bragg peak (0 0 1/2) on crossing the superconducting transition temperature $T_c = 1.85$ K. For $T < 2.6$ K, the width decreases with decreasing temperature. Usually such behavior reflects an increase of the magnetic correlation length. Surprisingly, the amplitude and hence the integrated intensity decrease as well. The minimum is reached close to the onset of superconductivity. Below the transition temperature, the width and intensity increase again on further cooling. The anomalous temperature dependence of the integrated intensity around T_c is shown in the inset of the upper part of Fig. 1. This remarkable effect of the order of 10% for the integrated intensity is reproducible and independent of the heating or cooling procedure. The angular position of the magnetic Bragg peak remains constant within the experimental accuracy.

Finally, in the upper part of Fig. 1 we compare the T-dependence of the ordered moment with the Brillouin function for a $S = 1/2$ system. The agreement is reasonable. However, there are characteristic deviations, and the experimentally observed magnetization close to the Néel temperature is significantly enhanced compared to the mean-field predictions. This sharp drop close to T_N could correspond to the preciptious drop of a soft-mode energy at the zone boundary characteristic for a singlet–singlet system. And indeed, magnetic measurements [6] suggested a tetravalent configuration of uranium in UPd_2Al_3 and two low-lying singlet states separated by 33 K [6].

At temperatures close to T_N, diffuse critical scattering could be observed for the two strongest magnetic Bragg peaks (0 0 1/2) and (0 0 3/2). The temperature dependence of the diffuse critical scattering measured at $Q = (0.03, 0, 1/2)$ is shown in the lower part of Fig. 1. From the well defined maximum we determined the Néel temperature of $T_N = 14.32(2)$ K for the larger UPd_2Al_3 crystal.

To gain further insight into the magnetic phase transition we tried to determine critical exponents with higher accuracy. The critical exponent β contains important information about the type of magnetic ordering involved. In Fig. 2 the temperature dependence of the logarithm of the reduced magnetic moment is plotted against the logarithm of the reduced temperature. For $T^* = (T_N - T)/T_N < 0.5$, both experiments result in a critical exponent of $\beta = 0.55 \pm 0.05$, i.e. a critical exponent close to classical MF behavior. For $T^* > 0.05$ significant deviations appear.

How do these experimental observations compare with the findings in the prototypical uranium-based HFS? UPt_3 orders antiferromagnetically at $T_N = 5$ K with an extremely small ordered moment of $0.02\,\mu_B/U$ μ_B/U and reveals superconductivity for temperatures below $T_c = 0.5$ K [7]. The magnetic

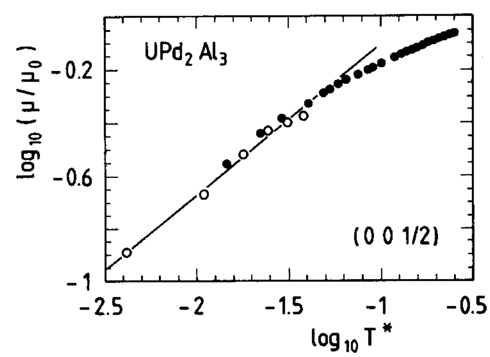

Fig. 2. Logarithm of the magnetic order parameter against the logarithm of the reduced temperature $T^* = (T_N - T)/T_N$. The straight line is a least square fit to the data. For $T^* < 0.05$ the slope corresponding to the critical exponent amounts to $\beta = 0.55$.

order parameter grows as $(T_N - T)^{0.5}$ down to the superconducting transition temperature. Below T_c magnetic order ceases to evolve [7]. This behavior was attributed [7] to the pairing of the electrons which then, can no longer contribute to the development of antiferromagnetic order. Doping the UPT_3 system with Th [8] or Pd [9], one observes that superconductivity is rapidly suppressed and magnetism with ordered moments of order 0.65 and $0.6\,\mu_B/U$ respectively, is stabilized. URu_2Si_2 is an uniaxial antiferromagnet with an ordered moment of $0.03\,\mu_B/U$ and a magnetic phase transition temperature $T_N = 17.5\,K$ [10]. Like in UPt_3, the integrated intensity of the magnetic Bragg peaks increases linearly from T_N to $0.15\,T_N$, suggesting mean-field behavior in an unusually broad temperature range. No anomalies at the superconducting transition temperature $T_c \simeq 1\,K$ were reported [11].

All the HFS are characterized by a magnetic ordering temperature T_N exceeding the superconducting transition temperature T_c considerably. Phenomenologically, this behavior distinguishes the HFS from Chevrel-type superconductors with coexisting antiferromagnetic order [12]. In these compounds the magnetic order usually appears well below T_c. However, the above comparison of the HFS reveals that there are also distinct differences in the magnetic behavior of UPd_2Al_3 compared to the "small-moment" antiferromagnets UPt_3 and URu_2Si_2. In UPd_2Al_3 superconductivity coexists with antiferromagnetic order involving a large ordered moment. The magnetic phase transitions in all these compounds are close to mean-field behavior. However, the temperature dependence of the magnetic moment in UPd_2Al_3 behaves almost like a $S = 1/2$ system. Superconductivity develops deep within the magnetically ordered phase characterized by a fully established magnetic order. In UPt_3 and URu_2Si_2 magnetic order develops continuously and is interrupted by the onset of superconductivity with quite different response in their respective magnetic order parameters.

The magnetic anomalies at the superconducting transition of UPd_2Al_3 shown in Fig. 1, demonstrate that there exists a significant interaction between superconductivity and magnetism. Further investigations at lower temperatures and in external magnetic fields are necessary to study this behavior in more detail.

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