Magnetic state in URu$_2$Si$_2$, UPd$_2$Al$_3$ and UNi$_2$Al$_3$ probed by point contacts

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Abstract

The antiferromagnetic (AFM) state has been investigated in the three heavy-fermion compounds URu$_2$Si$_2$, UPd$_2$Al$_3$, and UNi$_2$Al$_3$ by measuring $dV/dI(V)$ curves of point contacts at different temperatures (1.5-20 K) and magnetic fields (0-28 T). The zero-bias maximum in $dV/dI(V)$ for URu$_2$Si$_2$ points to a partially gapped Fermi-surface related to the itinerant nature of the AFM state contrary to UPd$_2$Al$_3$ where analogous features have not been found. The AFM state in UNi$_2$Al$_3$ has more similarities with URu$_2$Si$_2$. For URu$_2$Si$_2$, the same critical field of about 40 T along the easy c axis is found for all features in $dV/dI(V)$ corresponding to the Néel temperature, the gap in the electronic density of states, and presumably the ordered moments.

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The U-based heavy-fermion (HF) systems URu$_2$Si$_2$, UPd$_2$Al$_3$, and UNi$_2$Al$_3$ exhibiting antiferromagnetic (AFM) order followed by a superconducting transition at lower temperatures attract much interest in view of the possible coupling between superconducting and magnetic order. The on the first sight similar AFM ground state in the mentioned HF compounds reveals essential differences. While neutron-scattering experiments resolved an AFM ordered structure in URu$_2$Si$_2$ with a tiny ordered moment $0.03\pm0.01\,\mu_B$/U-atom below the Néel temperature $T_N=17.5\,K$ along the c-axis [1], UPd$_2$Al$_3$ has below $T_N=14\,K$ in the basal plane aligned U-moments equal to $0.85\pm0.03\,\mu_B$ [2]. Although the latter compound has the largest moment among the HF superconductors, it has the largest superconducting critical temperature $T_c$ of about $2\,K$ compared to typically $1.4\,K$ in URu$_2$Si$_2$. UNi$_2$Al$_3$ has been investigated much less than the other two compounds probably due to specific difficulties in the preparation of good samples. This compound is isostructural and isoelectronic to UPd$_2$Al$_3$, but has a few times smaller magnetic moment of about $0.24\pm0.10\,\mu_B$ [3] as well as lower critical ($\sim1\,K$) and Néel ($\sim5\,K$) temperatures.

Pronounced anomalies in specific heat, magnetic susceptibility, resistivity etc. for all three compounds indicate the phase transition into an AFM state. The specific resistivity in URu$_2$Si$_2$ has a well defined N-like structure at $T_N$ which looks like a kink for UPd$_2$Al$_3$ and is even more shallow for UNi$_2$Al$_3$. For the interpretation of the mentioned anomalies a transition into a spin-density wave (SDW) state has been considered [4] with a partial opening of a gap at the Fermi surface [4,5,6] of about $10\,mV$. Tunneling experiments which can determine the gap in the electronic DOS and its anisotropy yield for all three compounds a gap in the range $10$ to $20\,mV$ in the basal plane [7,8]. However, far infrared absorption [9] did not resolve any gap-like features for UPd$_2$Al$_3$ unlike in URu$_2$Si$_2$. For URu$_2$Si$_2$, the most investigated compound among this class of HF systems, it is still under discussion how the large anomalies in the transport and thermodynamic properties at $T_N$ can be reconciled with the tiny ordered moments. Therefore, the understanding of the nature of the magnetic order parameter in the AFM state of URu$_2$Si$_2$ remains a challenge. Recent transport and neutron scattering measurements in a high magnetic field revealed different transition fields for the AFM order or $T_N$ ($\sim40\,T$, [10]) and for the tiny staggered magnetic moments ($\sim14\,T$, [1]). This has led to a speculation about some additional 'hidden' magnetic order parameter in URu$_2$Si$_2$.

To clarify some aspects of the mentioned magnetic ordered state, we have performed a comparative point-contact study on these U-based HF compounds in strong magnetic fields. Of the three compounds the normal state properties have been investigated previously using point-contact spectroscopy only for URu$_2$Si$_2$ [12,13,14,15], however not in applied magnetic fields. The $dV/dI(V)$ characteristics of point contacts with URu$_2$Si$_2$ show an N-type feature related to local contact heating above the Néel temperature [15] and a zero-bias maximum which has been analyzed in terms of a partial suppression of the density of states related to an itinerant AFM ground state [12,14,15]. The present study allows to follow these characteristics of the AFM ground state in a magnetic field with a complete temperature dependent study of the phenomena.
We have investigated both homocontacts between the same HF compounds and heterocontacts between a HF compound and normal metals like Cu or Ag. The main difference was only in the degree of asymmetry of the $dV/dI(V)$ curves with respect to bias-voltage polarity, which is more pronounced for heterocontacts. The origin of the asymmetry is still under discussion [15]. Because this effect has no influence on the main conclusions of the present investigations, we will put no more attention to it. For the situation of the URu$_2$Si$_2$ single crystal, the heterocontacts were established in such a way that both contact axis and magnetic field were parallel to the c-axis or perpendicular to it. For the UPd$_2$Al$_3$ single crystal the contact axis and magnetic field were aligned along the easy basal plane direction whereas we used UNi$_2$Al$_3$ samples of unknown orientation. The measurements were carried in magnetic fields up to 28 T at 4.2 K, but for UNi$_2$Al$_3$ down to about 2 K and up to 10 T.

The measured $dV/dI(V)$ curves of URu$_2$Si$_2$ contacts can be separated into three groups. In the first group the $dV/dI(V)$ curves mimic the behaviour of bulk $\rho(T)$. The differential resistance increases with voltage and exhibits a $N$-type feature at about 20 mV (Fig. 1a) similar to $\rho(T)$ at $T_N$ [6]. The second type of $dV/dI(V)$ reveals a pronounced asymmetric zero-bias maximum (ZBM) of a width of about 10 mV in $dV/dI(V)$ (Fig. 1b) followed by gradually increasing signal at higher voltages. The third one contains simultaneously both kinds of structures in $dV/dI(V)$. We note that the temperature dependence of the contact resistance (Fig. 1, insets) corresponds in all cases to $\rho(T)$ independent of the type of $dV/dI(V)$ behaviour. This indicates that the material in the constriction reflects the bulk properties. The mentioned features, namely $N$-type kink and ZBM, vanish at the Neéel temperature (Fig. 1) and are therefore connected with the magnetic state.

The voltage position of the $N$-kink (marked by $V_k$ in Fig. 1a) is determined by $T_N$ and corresponds to the transition of the contact region from the AFM to the paramagnetic state most likely due to bias-voltage heating in the constriction. The temperature dependence $\sqrt{1-(T/T_N)^2}$ of $V_k$ shown in Fig. 2a is expected for such a local contact heating [13]. The ZBM is more pronounced for curves with shallow or not resolved kink peculiarities. Moreover, the ZBM cannot be described in the thermal model what can be directly seen upon comparing $dV/dI(V > 0)$ with $dV/dI(V = 0, T) = R(V = 0, T)$ (see Fig.1b). These observations point to the spectral nature of ZBM. The latter has been related (see e. g. [15]) with the existence of a gap in the excitation spectrum of the electrons due to the formation of a SDW below $T_N$. The ZBM has a width which is comparable with the gap value estimated in [4, 5, 6, 8]. The intensity of the ZBM gradually decreases with increasing temperature [12, 15] analogously to the intensity of AFM Bragg peaks describing the behaviour of the staggered magnetic moments or the order parameter [16]. Therefore, it is tempting to connect the ZBM also with the magnetic order parameter, although the microscopic nature of the tiny staggered magnetic moments in URu$_2$Si$_2$ as well as its influence on the measured $dV/dI$ are still unknown. Because the intensity of ZBM depends on the chosen criterion for the subtraction of the increased with a voltage background, we suggest to take voltage position of the minima
Figure 1: Two types of behaviour, (a) and (b), in the $dV/dI(V)$ curves for single crystal URu$_2$Si$_2$ homocontacts established in the basal plane at increasing temperature up to $T_N$. The curves are offset vertically for clarity. The insets show the temperature dependence of the zero bias resistance, which mimics $\rho(T)$ for bulk samples.

Figure 2: Temperature dependence of the reduced voltage positions $V_k$ (a) and $V_m$ (b) (see for definition Fig.1) for a few URu$_2$Si$_2$ homocontacts established in the basal plane. The solid circles for both figures correspond to the same homocontact. The solid line in both figures is the mean-field BCS dependence while the dashed curve describes the thermal regime behaviour [15].
Figure 3: (a) dV/dI (V) curves for a URu$_2$Si$_2$-Cu heterocontact in magnetic fields along the easy c axis at T=4.2 K. The solid curves correspond to the field sweep up, while the dashed one to the field sweep down. The arrows show definition of the kink $V_k$ and minimum $V_m$ positions. The curves are offset vertically for clarity. (b) Dependence of $V_k$, $V_m$ (left scale) and ZBM integrated intensity $I_M$ (right scale) versus magnetic field. Note, position of $V_k$, $V_m$ and ZBM intensity was taken after symmetrizing of dV/dI (V) curves. The solid lines represent dependence $(1 - (B/B_c)^2)$ characteristic for $T_N(B)$ and spin-wave gap $\Delta(B)$ [10] while dashed line $\sqrt{1 - (B/B_c)^{3/2}}$ is taken from [11] for staggered magnetic moments.

$V_m$ (see Fig. 3b) as an additional measure for the magnetic order parameter as supported by the mean-field (BCS-like) $V_m(T)$ dependence from Fig. 2b as also found in [13, 14].

The point-contact data presented in Fig. 3a for magnetic fields parallel to the easy c-axis exhibit both types of features discussed above, i.e. ZBM and N-kink. The integrated intensity of the ZBM (after subtracting a polynomial voltage-dependent background) is close to the $\sqrt{1 - (B/B_c)^{3/2}}$ behaviour like that for magnetic moments [11]. As shown in Fig. 3b, the same dependence is found for $V_m$ as well. On the other hand, $V_k$ follows the magnetic field dependence $(1 - (B/B_c)^2)$ like for $T_N$ [10] (shown in Fig. 3b). The latter dependence is found also for the width of ZBM (not shown), which is related to the SDW gap. Thus, the mentioned features in dV/dI(V) - $V_k$, ZBM width and ZBM intensity or $V_m$ measured on the same contact - are described by the magnetic
field dependencies characteristic for the behaviour of transition temperature $T_N$, magnetic gap width $\Delta$ and magnetic order parameter $\langle\sigma^z\rangle$, respectively. Moreover, as can be seen in Fig. 3b, independent of the type of behaviour in all cases the critical field is estimated to be about $40\,T$ what coincides with $B_c$ values measured by magnetoresistance. Therefore, unlike in Refs. 10, 11, where for the ordered magnetic moments the critical field is estimated to be about $14\,T$ our data show the presence of one order parameter, which vanishes at $T_N=17.5\,K$ and $B_c\approx 40\,T$. This is in line with the recent observation of van Dijk et al. 16 that the ordered moments remain coupled to the energy gap in the magnetic excitation spectrum in fields at least up to $12\,T$. We should emphasize that by measuring URu$_2$Si$_2$ contacts in a field perpendicular to the easy c-axis direction we did not found any remarkable influence of a magnetic field on $dV/dI(V)$ testifying that the point contact data really reflect the bulk properties.

Let us turn to the other compounds. The $dV/dI(V)$ curve of UPd$_2$Al$_3$ contacts (see Fig. 4) show a minimum at $V=0$ with edge maxima or shoulders which are connected with the AFM transition due to the heating effect, analogously to the $N$-type feature in the case of URu$_2$Si$_2$. However, for UPd$_2$Al$_3$ contacts we have never even seen a shallow ZBM neither for homo- nor for heterocontacts after the study of more than 20 contacts both below and above the Néel temperature. Therefore no evidence of the partially gapping of the Fermi surface is observed for UPd$_2$Al$_3$ unlike in URu$_2$Si$_2$ what points to a quite different magnetic ground state as well as to the different nature of the ordered moments for both compounds.

A magnetic field along the easy basal plane modifies the $dV/dI(V)$ curves of UPd$_2$Al$_3$ as can be seen from Fig. 4a. The maxima slightly shift ($\approx 15\%$) to lower energies and broaden with increasing magnetic field up to $18\,T$, and than vanish in higher fields. The width of the $dV/dI(V)$ minimum at $V=0$ has a minimum at $18\,T$, while the contact resistance has a kink at this field both at zero bias and finite bias voltage (see Fig. 4b, c). Hence the metamagnetic transition at $18\,T$ 19 is clearly resolved in point contact measurements, while no other phase boundary was observed both at lower and higher fields up to $28\,T$. From measurements of the dc susceptibility, dc magnetization, transverse magnetoresistivity, and magnetostriction, Grauel et al. 20 have also found a phase boundary in UPd$_2$Al$_3$ at a critical field of about $4\,T$ along the base plane. However, the influence of this low-field transition on the resistivity is at least one order of magnitude smaller compared to the transition at $18\,T$. Moreover, de Visser et al. 19 did not found a 4-T transition in their magnetoresistance data indicating that a re-orientation of the AFM domains could play a role in this phenomenon.

The $dV/dI(V)$ curves of UNi$_2$Al$_3$ represent usually a smooth broad almost symmetric minimum around zero-bias. However often a shallow ZBM can be observed around $V=0$ (Fig. 5). The distance between the minima in $dV/dI(V)$ with ZBM is about a few mV (often up to $10\,mV$) and ZBM disappears at about $5\,K$ (between 10-15 K for wider maxima). For ZBM with critical temperature of about $5\,K$ critical field was about $10\,T$. From this point of view UNi$_2$Al$_3$
Figure 4: $\frac{dV}{dI}(V)$ curves for a UPd$_2$Al$_3$-Cu heterocontact with $R_0=4.3\ \Omega$ at different magnetic fields along the basal plane and $T=4.2\ \text{K}$. Horizontal line with arrows shows determination of the width of the minimum. The curves are offset vertically for clarity. Right inset: width of the minimum versus magnetic field for the previous contact and for another contact with $R_0=0.09\ \Omega$. Left inset: magnetoresistance of the contact with $R_0=4.3\ \Omega$ at zero bias and at 10 mV.
Figure 5: Comparison of \( \frac{dV}{dI} \) curves for homocontacts with three studied HF compounds: UPd\(_2\)Al\(_3\) \((R_0=0.61 \ \Omega, \ T=4.2 \ \text{K})\), UNi\(_2\)Al\(_3\) \((R_0=1.5 \ \Omega, \ T=2.3 \ \text{K})\) and URu\(_2\)Si\(_2\) \((R_0=3.2\Omega, \ T=4.2 \ \text{K})\). A ZBM is only resolved for the two latter compounds. The curve for URu\(_2\)Si\(_2\) is shifted down by 0.15.

behaves similar to URu\(_2\)Si\(_2\) what hints to the developing of a magnetic state with partially gapping of the Fermi surface in this compound too. It should be noted that we didn’t resolve any feature in \( \frac{dV}{dI}(V) \) for UNi\(_2\)Al\(_3\) (Fig. 3) connected with \( T_N \) like that in URu\(_2\)Si\(_2\) (Fig. 1a) and UPd\(_2\)Al\(_3\) (Fig. 5). This transition at \( T_N \) is also very shallow in the \( \rho(T) \) dependence of UNi\(_2\)Al\(_3\). Probably, a better quality of the UNi\(_2\)Al\(_3\) samples is required to register the AFM transition and to study the temperature behaviour of ZBM in \( \frac{dV}{dI}(V) \).

Summarizing, the point-contact measurements for the investigated U-based heavy fermion compounds yield information on the differences in the AFM ground state of these systems. The ZBM-structure in \( \frac{dV}{dI}(V) \) for the URu\(_2\)Si\(_2\) contacts points to a partially gapped Fermi surface in the magnetically ordered state, but no evidence of an analogous structure has been found in the case of UPd\(_2\)Al\(_3\) unlike for UNi\(_2\)Al\(_3\) where it is possible to resolve a shallow ZBM. The results for URu\(_2\)Si\(_2\) in the \( H-V,T \) diagram yield only one critical Néel temperature of 17 K and one critical field of about 40 T along the easy c-axis for all features in \( \frac{dV}{dI}(V) \) testifying that they result from the same order parameter in the magnetic state.
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