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Magnetic state in URu_2Si_2 , UPd_2Al_3 , and UNi_2Al_3 probed by point contacts

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The antiferromagnetic (AFM) state has been investigated in the three heavy-fermion compounds URu_2Si_2 , UPd_2Al_3 , and UNi_2Al_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_2Si_2 points to a partially gapped Fermi surface related to the itinerant nature of the AFM state, contrary to the case of UPd_2Al_3 , where analogous features have not been found. The AFM state in UNi_2Al_3 has more similarities with URu_2Si_2 . For URu_2Si_2 , the same critical field of about 40 T along the easy axis **c** 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_2Si_2 , UPd_2Al_3 , and UNi_2Al_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 at first sight similar AFM ground state in the HF compounds mentioned reveals essential differences. While in URu_2Si_2 neutron-scattering experiments below the Néel temperature T_N = 17.5 K resolved an AFM ordered structure with a tiny ordered moment $(0.03 \pm 0.01)\ \mu_B$/U-atom along the **c** axis [1], UPd_2Al_3 below T_N = 14 K has aligned U moments equal to $(0.85 \pm 0.03)\ \mu_B$ in the basal plane [2]. Although the latter compound has the largest moment among the HF superconductors, it has the highest superconducting critical temperature T_c of about 2 K, compared to the typical value of 1.4 K in URu_2Si_2 . UNi_2Al_3 has been investigated much less than the other two compounds, probably because of specific difficulties in the preparation of good samples. This compound is isostructural and isoelectronic to UPd_2Al_3 , but has a few times smaller magnetic moment of about $(0.24 \pm 0.10)\ \mu_B$ [3] as well as lower critical (~ 1 K) and Néel (~ 5 K) temperatures.

Pronounced anomalies in the specific heat, magnetic susceptibility, resistivity, etc. for all three compounds indicate a phase transition to an AFM state. The resistivity has a well defined *N*-like structure at T_N in URu_2Si_2 , which looks like a kink for UPd_2Al_3 and is even more shallow for UNi_2Al_3 . For the interpretation of these anomalies a transition to a spin-density wave (SDW) state has been considered [4], with a partial opening of a gap of about 10 mV at the Fermi surface [4–6]. Tunneling experiments which can determine the gap in the electronic density of states (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_2Al_3 , unlike in URu_2Si_2 . For URu_2Si_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, understanding the nature of the magnetic order parameter in the AFM state of URu_2Si_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 (~ 40 T [10]) and for the tiny staggered magnetic moments (~ 14 T [11]). This has led to a speculation about some additional «hidden» magnetic order parameter in URu_2Si_2 .

To clarify some aspects of the aforementioned 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_2Si_2 [12–15], although not in applied magnetic fields. The $dV/dI(V)$ characteristics of point contacts with URu_2Si_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 one 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 devote no more attention to it. In the case of the URu_2Si_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_2Al_3 single crystal the contact axis and magnetic field were aligned along the easy basal plane direction, whereas we used UNi_2Al_3 samples of unknown orientation. The measurements were carried in magnetic fields up to 28 T at 4.2 K (but for UNi_2Al_3 up to 10 T at temperatures down to about 2 K).

The measured $dV/dI(V)$ curves of URu_2Si_2 contacts can be separated into three groups. In the first group the $dV/dI(V)$ curves mimic the behavior of bulk $\rho(T)$. The differential resistance increases with voltage and exhibits an N-type feature at about 20 mV (Fig. 1,*a*) similar to $\rho(T)$ at T_N [6]. The second type of $dV/dI(V)$ exhibits a pronounced

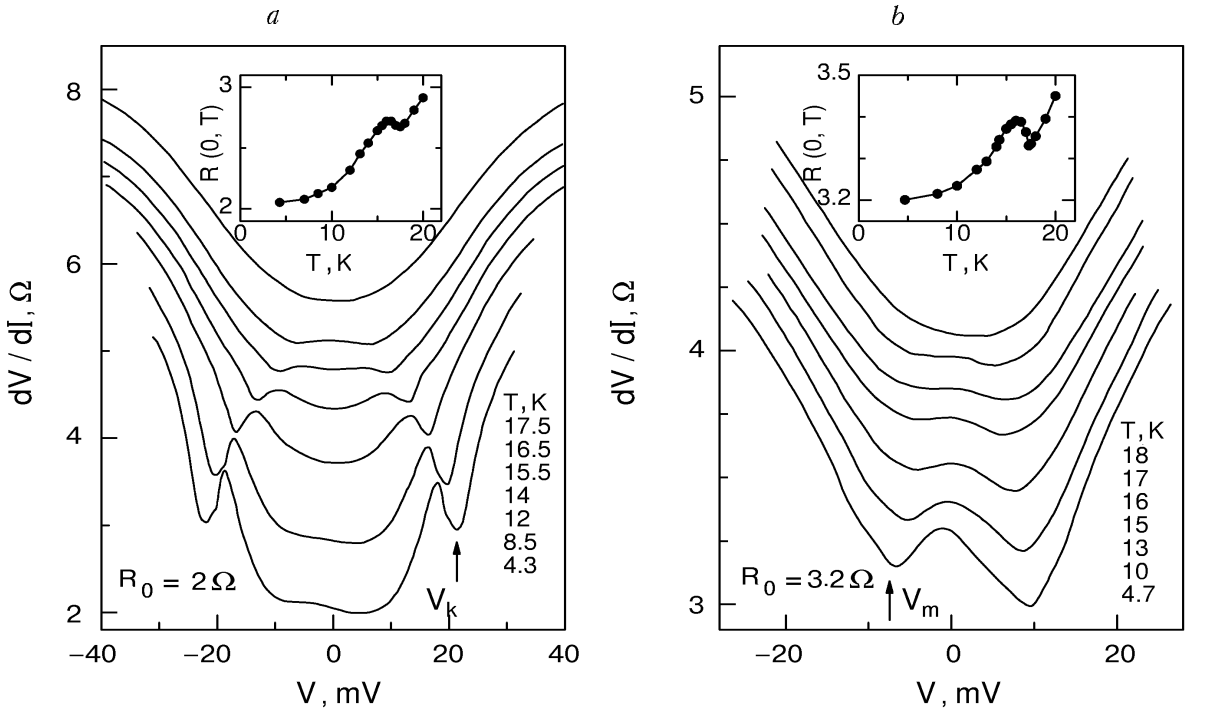

Fig. 1. Two types of behavior, (*a*) and (*b*), in the $dV/dI(V)$ curves for single crystal URu_2Si_2 homocontacts established in the basal plane at increasing temperature up to T_N . The curves are offset vertically for clarity. The insets show the temperature dependence of the zero-bias resistance, which mimics $\rho(T)$ for bulk samples.

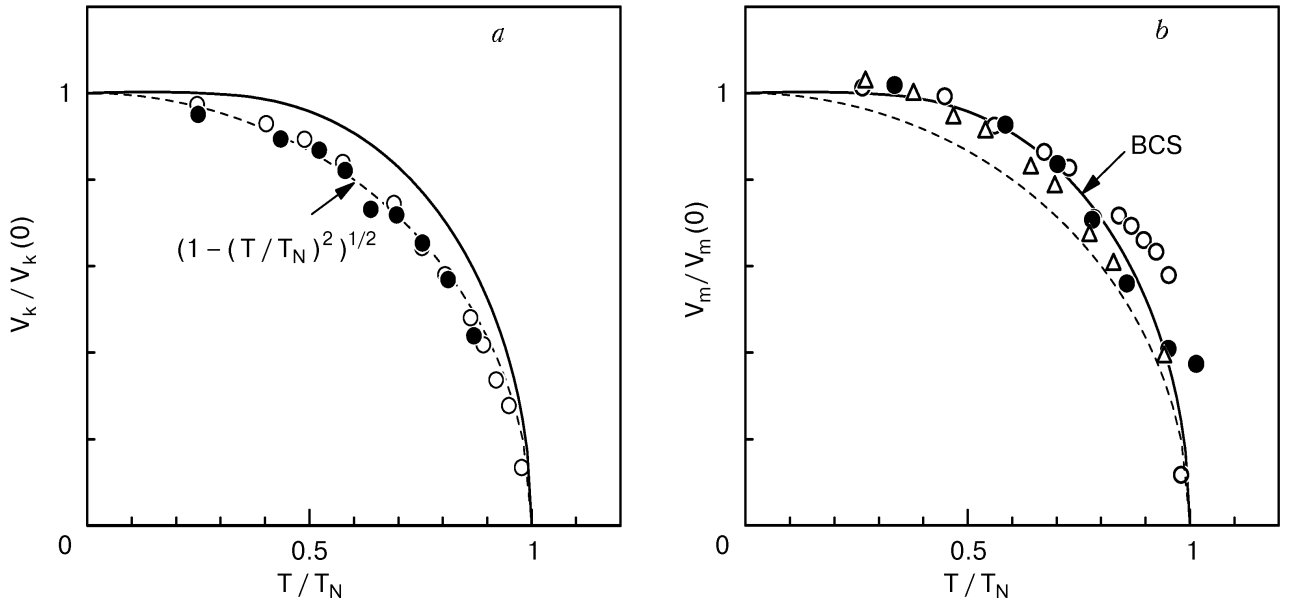

Fig. 2. Temperature dependence of the reduced voltage positions V_k (*a*) and V_m (*b*) (for definition see Fig. 1) for a few URu_2Si_2 homocontacts established in the basal plane. The solid circles for both figures correspond to the same homocontact. The solid line in both figures is the mean-field BCS dependence, while the dashed curve describes the thermal regime behavior [15].

asymmetric zero-bias maximum (ZBM) with a width of about 10 mV in $dV/dI(V)$ (Fig. 1,*b*), followed by gradually increasing signal at higher voltages. The third group 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 a $\rho(T)$ independent of the type of $dV/dI(V)$ behavior. This indicates that the material in the constriction reflects the bulk properties. The features mentioned, namely the *N*-type kink and ZBM, vanish at the Néel temperature (Fig. 1) and are therefore connected with the magnetic state.

The voltage position of the *N*-kink (marked by V_k in Fig. 1,*a*) 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. 2,*a* is expected for such a local contact heating [15]. The ZBM is more pronounced for curves with shallow or unresolved kink anomalies. Moreover, the ZBM cannot be described in the thermal model, as can be seen directly upon comparing $dV/dI(V > 0)$ with $dV/dI(V = 0, T) = R(V = 0, T)$ (see Fig. 1,*b*). These observations point to the spectral nature of the 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 to the gap value estimated in [4,6–8]. The intensity of the ZBM decreases gradually with increasing temperature [12,15] analogously to the intensity of AFM Bragg peaks describing the behavior 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_2Si_2 and their influence on the measured dV/dI are still unknown. Because the intensity of the ZBM depends on the chosen criterion for subtraction of the increasing-with-voltage background, we suggest taking the voltage position of the minima V_m (see Fig. 1,*b*) as an additional measure for the magnetic order parameter, as supported by the mean-field (BCS-like) $V_m(T)$ dependence in Fig. 2,*b*, which was also found in [13,14].

The point-contact data presented in Fig. 3,*a* for magnetic fields parallel to the easy axis **c** exhibit both types of features discussed above, i.e., ZBM and *N*-kink. The integrated intensity of the ZBM (after subtraction of a polynomal voltage-dependent background) is close to the $\sqrt{1-(B/B_c)^{3/2}}$ behavior like that for magnetic moments [11]. As shown in Fig. 3,*b*, 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 that for T_N [10] (shown in Fig. 3,*b*). The latter dependence is found also for the width of the ZBM (not shown), which is related to the SDW gap. Thus the abovementioned features in $dV/dI(V) - V_k$, the

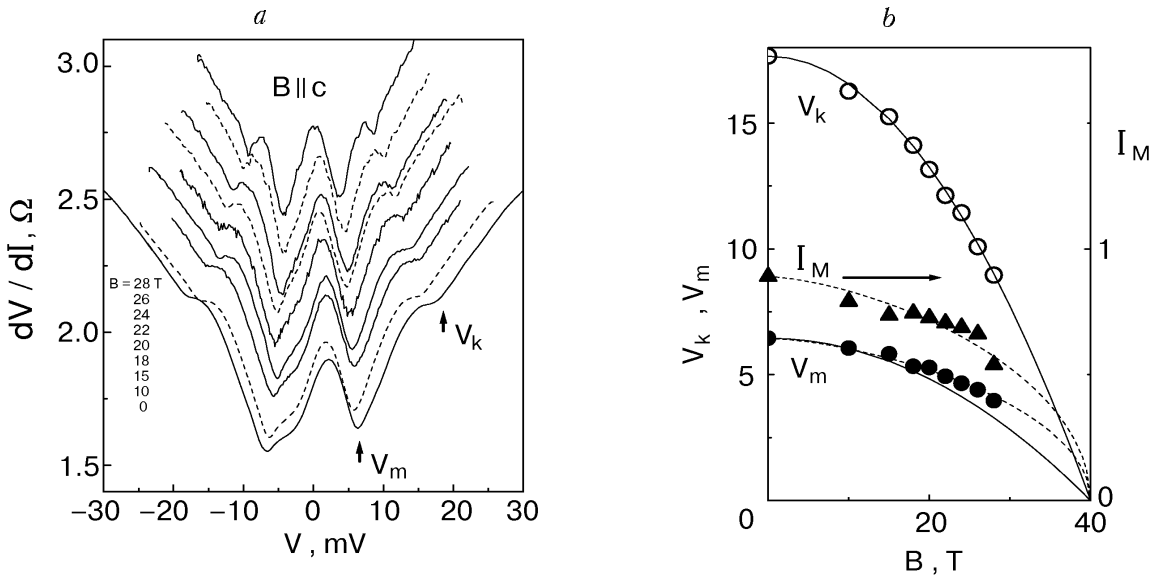

Fig. 3. $dV/dI(V)$ curves for a URu_2Si_2–Cu heterocontact in magnetic fields along the easy axis **c** at $T = 4.2$ K. The solid curves correspond to the field sweep up, while the dashed curve is for the field sweep down. The arrows indicate the kink V_k and minimum V_m positions. The curves are offset vertically for clarity (*a*). Dependence of V_k , V_m and ZBM integrated intensity I_M versus magnetic field. Note, that the position of V_k , V_m , and ZBM intensity was taken after symmetrizing of the $dV/dI(V)$ curves. The solid lines represent the dependence $(1-(B/B_c)^2)$ characteristic for $T_N(B)$ and the spin-wave gap $\Delta(B)$ [10], while the dashed line $\sqrt{1-(B/B_c)^{3/2}}$ is taken from [11] for staggered magnetic moments (*b*).

ZBM width, and the ZBM intensity or V_m measured on the same contact — are described by the magnetic field dependences characteristic for the behavior of the transition temperature T_N , magnetic gap width [10], and magnetic order parameter [11], respectively. Moreover, as can be seen in Fig. 3,*b*, independent of the type of behavior, in all cases the critical field is estimated to be about 40 T, which coincides with the B_c values measured by magnetoresistance [17]. Therefore, unlike in [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 $B_c \simeq 40$ 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 up to at least 12 T. We should emphasize that by measuring URu_2Si_2 contacts in a field perpendicular to the direction of the easy axis **c** we did not find any noticeable influence of 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_2Al_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 [18], analogously to the *N*-type feature in the case of URu_2Si_2 . However, for UPd_2Al_3 contacts we have never seen even 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_2Al_3 , unlike in URu_2Si_2 , a finding which, points to a quite different magnetic ground state as well as to a different nature of the ordered moments for the two compounds.

A magnetic field along the easy basal plane modifies the $dV/dI(V)$ curves of UPd_2Al_3 , as can be seen from Fig. 4,*a*. The maxima shift slightly ($\approx 15\%$) 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. 4,*b*,*c*). Hence the metamagnetic transition at 18 T [19] is clearly resolved in the 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_2Al_3 at a critical field of about 4 T along the basal plane. However, the influence of this low-field transition on the resistivity is at least one order of magnitude smaller than the transition at 18 T. Moreover, de Visser et al. [19] did not find transition at 4 T in their magnetoresistance data,

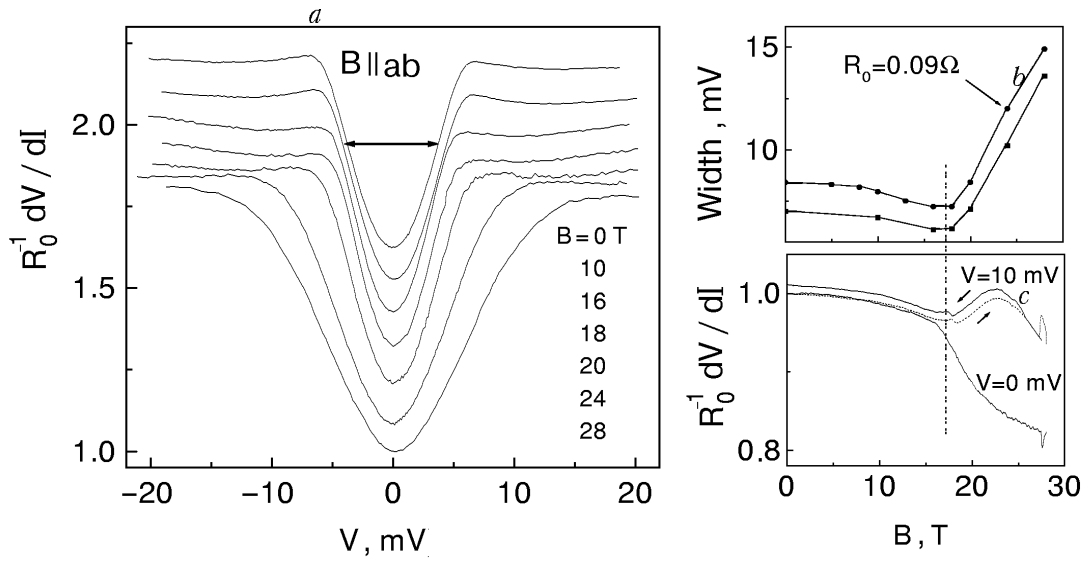

Fig. 4. $dV/dI(V)$ curves for a UPd_2Al_3–Cu heterocontact with $R_0 = 4.3$ Ω at different magnetic fields along the basal plane and $T = 4.2$ K. The horizontal line with arrows indicates the width of the minimum. The curves are offset vertically for clarity (*a*). Width of the minimum versus magnetic field for the previous contact and for another contact with $R_0 = 0.09$ Ω (*b*). Magnetoresistance of the contact with $R_0 = 4.3$ Ω at zero bias and at 10 mV. The vertical dashed line marks the position of the metamagnetic transition at 18 T (*c*).

indicating that a reorientation of the AFM domains could play a role in this phenomenon.

The $dV/dI(V)$ curves of UNi_2Al_3 usually represent 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 a few mV (often up to 10 mV), and the ZBM disappears at about 5 K (between 10–15 K for wider maxima). For a ZBM with a critical temperature of about 5 K the critical field was about 10 T. From this point of view UNi_2Al_3 behaves similarly to URu_2Si_2 , hinting at the development of a magnetic state with a partial gapping of the Fermi surface in this compound, too. It should be noted that for UNi_2Al_3 we didn't resolve any feature in $dV/dI(V)$ (Fig. 5) connected with T_N , like that in URu_2Si_2 (Fig. 1,*a*) and UPd_2Al_3 (Fig. 5). This transition at T_N is also very shallow in the $\rho(T)$ dependence of UNi_2Al_3 . Probably a better quality of the UNi_2Al_3 samples is required to register the AFM transition and to study the temperature behavior of the ZBM in $dV/dI(V)$.

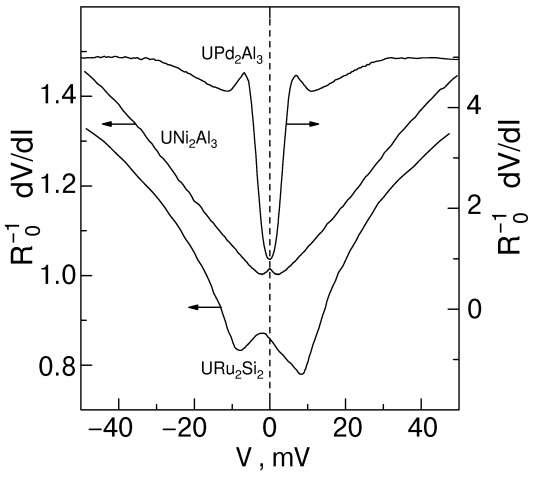

Fig. 5. Comparison of the $dV/dI(V)$ curves for homocontacts with the three HF compounds studied: UPd_2Al_3 ($R_0 = 0.61$ Ω, $T = 4.2$ K), UNi_2Al_3 ($R_0 = 1.5$ Ω, $T = 2.3$ K), and URu_2Si_2 ($R_0 = 3.2$ Ω, $T = 4.2$ K). A ZBM is only resolved for the latter two compounds. The curve for URu_2Si_2 is shifted down by 0.15.

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 $dV/dI(V)$ for the URu_2Si_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_2Al_3 , unlike for UNi_2Al_3 , where it is possible to resolve a shallow ZBM. The results for URu_2Si_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 axis **c** for all features in $dV/dI(V)$, testifying that they result from the same order parameter in the magnetic state.

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