Suppression of asymmetric differential resistance in the non-Fermi-liquid system $YbCu_{5-x}Al_x$ ($x=1.3-1.75$) in high magnetic fields

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The non-Fermi-liquid system $YbCu_{5-x}Al_x$ ($x=1.3-1.75$) has been investigated in heterocontact as well as homocontact arrangement in magnetic fields up to 22.5 T. The observed differential resistance $[dV/dI(V)]$ characteristics reveal asymmetry in heterocontact arrangement and do not agree with the model of thermal contact heating, at least close to zero-bias voltage. In the case of a heterocontact arrangement we have observed a maximum at only one voltage polarity at about 1.3 mV (for $x=1.5$) and this asymmetry is suppressed in an applied magnetic field. We show that such behavior is connected with the non-Fermi-liquid state of the studied system.

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I. INTRODUCTION

Point-contact spectroscopy (PCS) is a very efficient tool for the study of electronic scattering processes in metallic conductors.[1] In the case of ballistic transport of the charge carriers across the contact, the applied contact voltage defines directly the energy scale of the scattering processes investigated, e.g., the electron-phonon interaction. This is valid for simple metals and compounds but there are some polemics about applicability of PCS in more complicated systems, such as heavy-femion systems, which are systems with strongly correlated electrons. Most of the papers relating to PCS and tunneling spectroscopy are connected with superconducting systems, where the critical points are covered by superconductivity. Here, we present the results of the application of PCS to the nonsuperconducting system $YbCu_{5-x}Al_x$ ($x=1.3-1.75$) showing non-Fermi-liquid (NFL) behavior in the vicinity of the quantum critical point (QCP).

$YbCu_{5-x}Al_x$ is a very interesting compound with a valency change from $\nu \approx 2.2 (x=0)$ to $\nu \approx 3 (x=2)$, which causes a magnetic instability near a critical concentration $x_{cr}=1.5$ in the crossover from the almost nonmagnetic $4f^{14}$ state in $YbCu_5$ to the magnetic $4f^{13}$ state in $YbCu_3Al_2$.[2] The intermetallic system $YbCu_{5-x}Al_x$ with concentration in the vicinity of x_{cr} exhibits a typical NFL behavior similar to a negative logarithmic term in the temperature-dependent specific heat or deviations from the quadratic temperature dependence of the electrical resistivity.[2] Such a NFL behavior is often found in strongly correlated electron systems where the magnetic ordering temperature tends toward zero leading to a QCP. In our system the QCP is near x_{cr}.[2] There exist scenarios for the occurrence of NFL behavior,[3,4] but the microscopic basis of the NFL ground state is not yet completely understood. Theoretical work by Shaginyan and Popov[5] predicted the asymmetric shape of dynamic conductance for strongly correlated electron systems in the vicinity of a QCP in the normal and/or superconducting state. This asymmetric part is expected to have a linear contribution (part). Moreover, the absence of the classical quasiparticles in spectra is characteristic for the NFL systems.[5]

Our previous PC experiments on $YbCu_{5-x}Al_x$ ($x=1.3-1.6$) have been performed at temperatures down to 1.5 K and in magnetic fields up to 6 T in the heterocontact configuration using a Cu or Pt counterelectrode.[6,7] The differential resistance $dV/dI(V)$ as a function of the applied voltage V revealed an asymmetric behavior. In the case of $x_{cr}=1.5$ we have observed a maximum at 1.3 mV in only one voltage polarity. This different type of asymmetry is connected with the NFL behavior at the QCP.[6,7] The application of a magnetic field strongly changed the shape of the differential resistance, showing a recovering of the Fermi-liquid (FL) behavior characteristic for Kondo compounds.[8,9] The behavior of PC dependencies of other concentrations differs from the one at the critical concentration.

Since previous measurements were done in the magnetic fields up to 6 T only, which is not enough to fully suppress the NFL behavior and restore the FL behavior, we applied high magnetic fields up to 22.5 T. In this overview we present systematic point-contact measurements in the heterocontact and homocontact arrangements at low temperatures in magnetic fields up to 22.5 T, studying several $YbCu_{5-x}Al_x$ compounds in the vicinity of the QCP ($x_{cr}=1.5$). Measurements of heterocontacts were done down to 100 mK. We used the same samples as characterized and studied in previous work by Bauer *et al.*[2,10]

II. EXPERIMENTAL DETAILS

Samples have been prepared from stoichiometric amounts of elements using high-frequency melting. The ingots were remelted several times and subsequently annealed for 10 days at 750 °C. The detailed fabrication of samples was described by Bauer *et al.*[10]

Measurements were carried out by the "needle-anvil" technique[11,12] using a differential screw mechanism for the tip-sample adjustment. Before each series of measurements

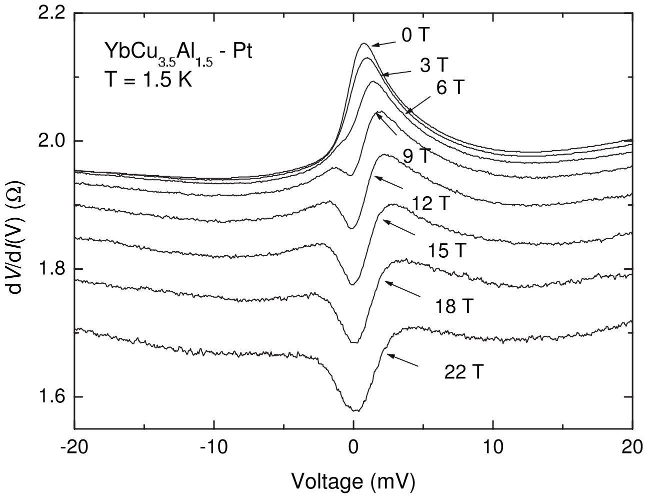

FIG. 1. Characteristic magnetic-field behavior of $dV/dI(V)$ for heterocontact $YbCu_{3.5}Al_{1.5}$-Pt at 1.5 K.

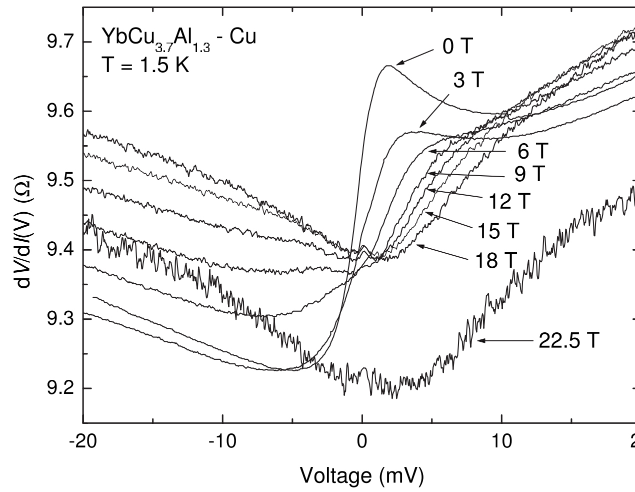

FIG. 2. Characteristic magnetic-field behavior of $dV/dI(V)$ for heterocontact $YbCu_{3.7}Al_{1.3}$-Cu at 1.5 K.

the polycrystalline sample was mechanically polished or freshly broken in order to obtain a clean surface. In the case of heterocontacts, the counterelectrode was made from a copper or a platinum wire of 100 μm diameter with a sharpened tip. In the homocontact arrangement, both parts of the contact were made from broken pieces of the polycrystalline sample under study. We recorded the differential resistance $dV/dI(V)$ of the current-voltage characteristic using standard phase-sensitive detection.[11,12]

III. RESULTS AND DISCUSSION

In order to investigate the influence of a magnetic field, which is known to destroy the NFL state in the $YbCu_{5-x}Al_x$ system, we have applied magnetic fields up to 22.5 T. The $dV/dI(V)$ curves of a $YbCu_{3.5}Al_{1.5}$-Pt heterocontact at 1.5 K are shown in Fig. 1 in the applied magnetic fields. With increasing magnetic field the characteristic maximum at 1.3 mV (only present in one polarity of the applied voltage) shifts to higher voltages and evolves subsequently into a split two-peak structure. In general, the point-contact resistance is decreasing with increasing magnetic field. For high magnetic fields (above 12 T) the voltage position of both peaks is symmetric with respect to zero voltage but their intensities are different. This kind of slightly asymmetric behavior ($B > 12$ T) is characteristic for the point-contact $dV/dI(V)$ dependencies of heavy-fermion compounds in magnetic field.[1,8] From measurements of the electrical resistivity, magnetic susceptibility, and specific heat on bulk samples of $YbCu_{3.5}Al_{1.5}$, it is evident that approximately 12 T is enough to recover FL behavior and that the compounds with concentrations $x < x_{cr}$ exhibit Kondo-type behavior.[2] Therefore, we conclude that in our PC spectra 12 T is enough to recover the FL behavior of the Kondo-type. This observation gives an argument for the NFL origin of our observed asymmetry in magnetic fields below about 12 T.

Further confirmation of this conclusion can be seen in the point-contact characteristics of heterocontacts for $x \neq x_{cr}$ shown in Figs. 2 and 3. In comparison with Fig. 1 one can see for $x=1.3$ and 1.75 data that the asymmetric maximum is already suppressed by weaker magnetic fields and the remains of a split two-peak structure are suppressed in the highest magnetic fields. The main difference for the $x \neq x_{cr}$ data lies in the high-field response, which tends to correspond to metalliclike $dV/dI(V)$ dependence while for $x = x_{cr}$ the maxima in $dV/dI(V)$ remain similar to PCs of the Kondo systems.[8,9]

In order to shed light on the origin of the observed asymmetric maximum in the heterocontacts, we performed measurements of $dV/dI(V)$ in the homocontact arrangement too. In Fig. 4 the characteristic behavior of homocontacts is presented (up to 9 T). Because of differences in the magnetic forces between the contacting parts in the case of the homocontact arrangement, the contacts were less stable in an applied magnetic field compared with heterocontacts. The main result is that the maximum in $dV/dI(V)$ occurs for the homocontacts at zero applied voltage. With the applied magnetic field the splitting of the maximum (symmetrically positioned around zero voltage) occurs like in the case of heterocon-

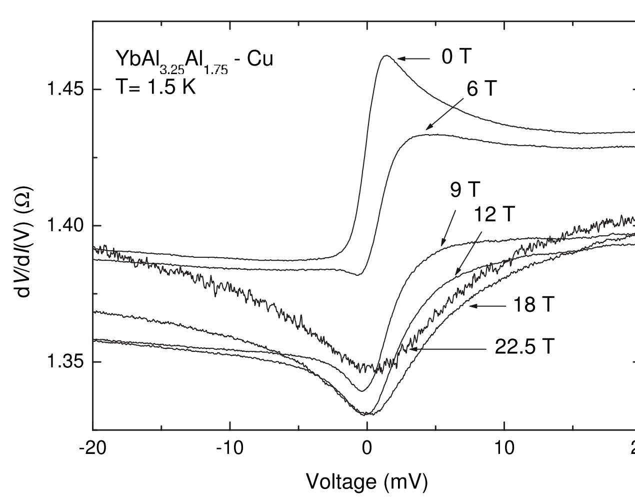

FIG. 3. Characteristic magnetic-field behavior of $dV/dI(V)$ for heterocontact $YbCu_{3.25}Al_{1.75}$-Cu at 1.5 K.

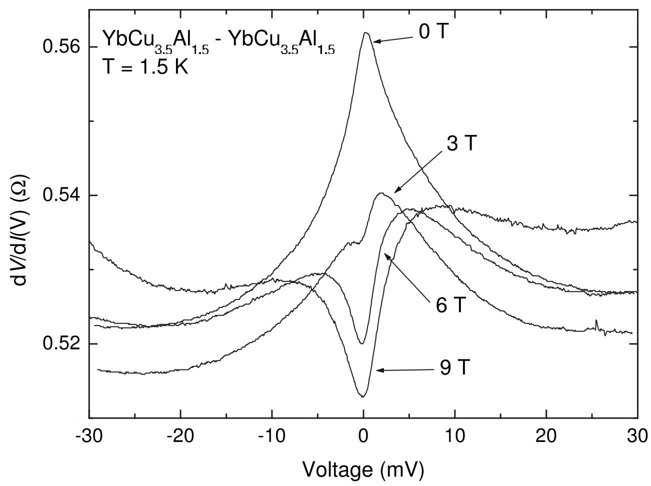

FIG. 4. Characteristic magnetic-field behavior of $dV/dI(V)$ for homocontact $YbCu_{3.5}Al_{1.5}$-$YbCu_{3.5}Al_{1.5}$ at 1.5 K.

tacts. In the case of homocontacts for $x \neq x_{cr}$ we also observed a maximum in $dV/dI(V)$ at zero-bias voltage splitting up in the applied magnetic field.

The comparison of $dV/dI(V)$ curves for homocontact $YbCu_{3.5}Al_{1.5}$-$YbCu_{3.5}Al_{1.5}$ and for heterocontact $YbCu_{3.5}Al_{1.5}$-Pt is shown in Fig. 5. The observed asymmetry in the differential resistance does not depend on the used needle (Cu or Pt). The asymmetry observed in the $dV/dI(V)$ curves of the heterocontact is directly related to the configuration of a contact between metals with different material properties. From the similarities between the point-contact spectra of the heterocontacts and homocontacts, we conclude that the $dV/dI(V)$ maximum is connected to the properties of the $YbCu_{5-x}Al_x$ electrode of the contacts. The asymmetric voltage position of the maximum is only observed in the NFL phase of the samples.

In order to explain the observed behavior in the point-contact spectra we have to consider the regime of current flow across the constriction, which can be ballistic, diffusive, or thermal, depending on the mean-free-path lengths, elastic l_{el} and inelastic l_{in}, with respect to the contact diameter d.

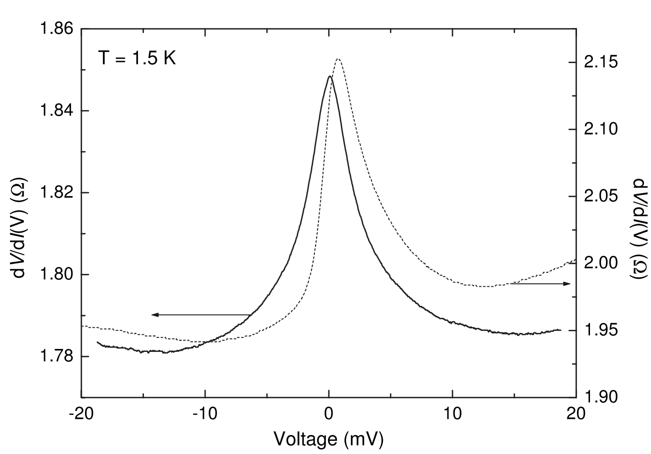

FIG. 5. Characteristic $dV/dI(V)$ for heterocontact $YbCu_{3.5}Al_{1.5}$-Pt (dashed line) and homocontact $YbCu_{3.5}Al_{1.5}$-$YbCu_{3.5}Al_{1.5}$ (solid line) arrangements at 1.5 K.

Covering the whole range of the mean-free-path values with respect to the PC diameter, a simple formula was derived by Wexler[13] for the PC resistance,

$$R_{PC} = \frac{16\rho l_{el}}{3\pi d} + \beta\frac{\rho(T)}{d}, \qquad (1)$$

where $\rho l_{el} = p_F/ne^2$ with $\rho(T)$ as the resistivity, p_F as the Fermi momentum, n the as density of electrons, and e as the electron charge. The coefficient $\beta \simeq 1$ for $l_{el} \ll d$.[14] The Wexler formula represents simply an interpolation between the ballistic Sharvin ($l_{el} \gg d$) resistance (first term) and the diffusive Maxwell ($l_{el} \ll d$) resistance (second term). In our case the $\rho(T)$ of $YbCu_{3.5}Al_{1.5}$ is about 100 $\mu\Omega$ cm (at T=4.2 K), and typical values of R_{PC} are from 0.5 to about 30 Ω. Then from Eq. (1), we can estimate the diameter of the PCs between 50 and 2000 nm taking the typical value of $\rho l_{el} = 10^{-11}$ Ω cm^2 for metals.[14] We estimate the typical elastic mean-free path, l_{el}, for our compounds in the nanometer range. As a result, one can hardly expect ballistic transport of the conduction electrons across the PC for the x_{cr} compound, but there exists still the possibility of a long inelastic diffusion length at low applied voltage as compared to the contact diameter. For an inelastic diffusion length $(l_{in}l_{el})^{1/2} > d \geq l_{el}$ the point-contact data can still contain spectroscopic information where the applied voltage defines the energy scale.

In order to exclude the thermal regime with $l_{el}, l_{in} < d$ where energy-resolved spectroscopy is no longer possible, we performed a critical analysis of our data. In the case of thermal regime, $dV/dI(V)$ resembles $\rho(T)$ and a simple relation exists between the contact resistance as a function of an applied voltage and the resistivity as a function of temperature,[15]

$$I(V) = Vd\int_1^0 \frac{dx}{\rho(T\sqrt{1-x^2})}, \qquad (2)$$

with $T = eV/3.36k_B$ for the maximum temperature at the contact center as a function of the applied voltage.

For the heated contact between two different metals, the thermoelectric voltage caused by the difference of the Seebeck coefficients of the contacting metals results in an asymmetry of the differential resistance curves versus bias voltage. To quantify the asymmetry of the differential resistance curves, we separate the $dV/dI(V)$ curves into the symmetric part $dV/dI(V)^s = [dV/dI(V<0) + dV/dI(V>0)]/2$ and the asymmetric part $dV/dI(V)^{as} = [dV/dI(V>0) - dV/dI(V<0)]/2$. For our experimental configuration, the negative potential is connected to the $YbCu_{5-x}Al_x$ sample. As shown by Itskovich and Shekhter,[16] the asymmetric differential resistance behaves like

$$\frac{1}{R_0}\left[\frac{dV}{dI}(V)\right]^{as} \propto S_1(T_{PC}) - S_2(T_{PC}), \qquad (3)$$

where $S_{1,2}$ are the Seebeck coefficients of two metals. The maximum temperature T_{PC} in the contact is determined by

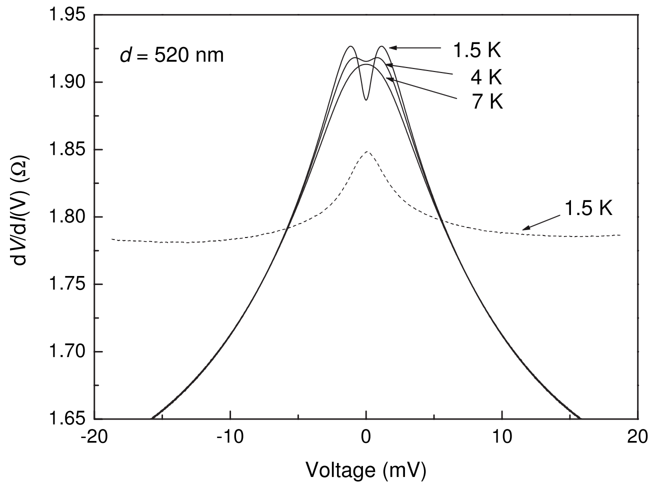

FIG. 6. Comparison of calculated $dV/dI(V)$ curves at different temperatures (solid curves) and experimental data of homocontact $YbCu_{3.5}Al_{1.5}$-$YbCu_{3.5}Al_{1.5}$ (dashed curve) at 1.5 K.

$$T_{\mathrm{PC}}^2 = T_{\mathrm{bath}}^2 + \frac{V^2}{4L}, \tag{4}$$

where T_{bath} is the surrounding bath temperature which reduces Eq. (4) to the previously cited expression $T_{\mathrm{PC}} = eV/3.36k_B$ for the free-electron Lorenz number L. If the metal 1 has much higher resistivity and thermopower in comparison to the metal 2, we have $dV/dI(V)^s \propto \rho_1(T)$ and $dV/dI(V)^{as} \propto S_1(T)$.

Figure 6 shows a comparison of the experimental $dV/dI(V)$ data for a homocontact of $YbCu_{3.5}Al_{1.5}$ at 1.5 K (dashed curve) with the calculated characteristics for different temperatures (solid curves) using the bulk resistivity data[2] (see also the inset of Fig. 7) in Eq. (2). The diameter d of the PC is obtained from the expression of the Maxwell contact resistance. From the theoretical analysis, a minimum is expected in $dV/dI(V)$ at zero-bias voltage for temperatures below 4 K, corresponding to a decrease in resistivity in this temperature range. However, a zero-bias maximum is observed in experiment all the time. In addition, the relative change in the measured $dV/dI(V)$ is smaller than calculated using Eq. (2). This latter discrepancy between experiment and theory is often observed in the analysis of point-contact data of the heavy-fermion compounds and interpreted in terms of an additional contact resistance resulting from different causes.[17,18]

Figure 7 shows the temperature evolution of (a) symmetric and (b) asymmetric parts of heterocontact $YbCu_{3.5}Al_{1.5}$-Cu at zero applied magnetic field. The inset shows the bulk resistivity $\rho(T)$ of this compound. Also for the case of a heterocontact, the behavior of symmetric part $dV/dI(V)^s$ does not show the corresponding decrease in the bulk $\rho(T)$ (see inset) at the lowest temperatures as would be expected for the thermal heating model at the lowest bias voltages.

In order to analyze if experimental broadening could explain the absence of a minimum in $dV/dI(V)$ at zero bias as expected from the decreasing resistivity at the lowest tem-

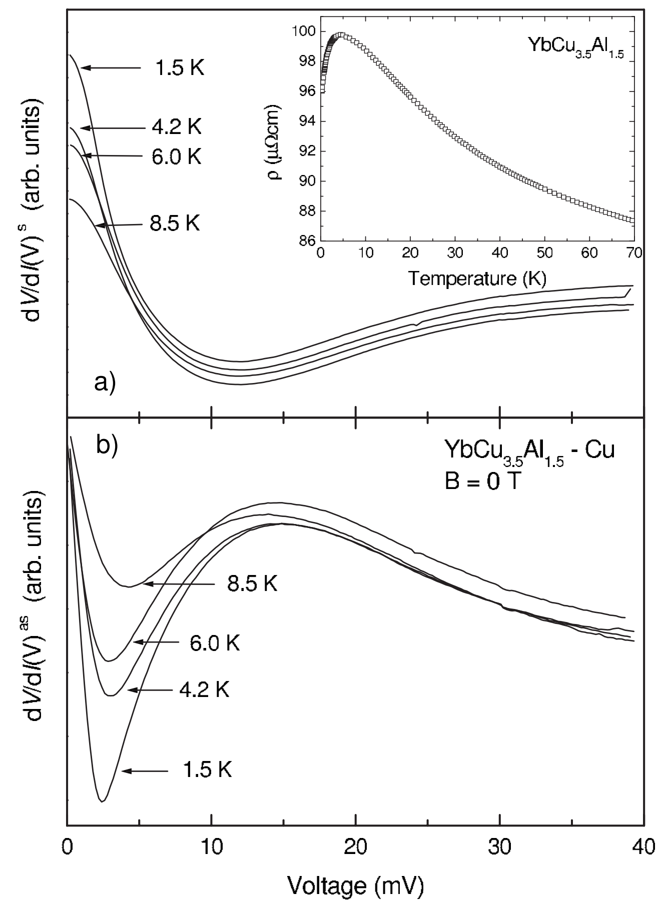

FIG. 7. Characteristic temperature behavior of (a) symmetric and (b) asymmetric parts of $dV/dI(V)$ for heterocontact $YbCu_{3.5}Al_{1.5}$-Cu at $B=0$ T. The inset shows the bulk resistivity $\rho(T)$ of $YbCu_{3.5}Al_{1.5}$ (Ref. 2).

peratures, we will consider the experimental broadening in more detail. The experimental resolution is given by $\sqrt{(3.53k_BT/e)^2+(1.73\sqrt{2}V_1)^2}$, where the first term results from thermal broadening at a bath temperature T and the second term from modulation broadening with amplitude V_1 for the recording of first derivative $dV/dI(V)$ of the current-voltage characteristic.[14] For a typical modulation amplitude $V_1=0.3$ mV, the thermal broadening dominates at temperatures above about 5 K. In order to improve thermal resolution and to verify the regime of current transport through the contact, we have performed PCS at temperatures near 100 mK using a dilution refrigerator. Moreover, at these low temperatures we approach the QCP ($T=0$ K) as close as possible for our measurement facility. In the case of $T=100$ mK, the typically applied modulation voltage of about 0.3 mV determines the experimental resolution to 0.7 mV.

The comparison of $dV/dI(V)$ at 6 K and at about 100 mK is presented in Fig. 8. At lower temperatures the thermal broadening is reduced but the position of the maximum remains the same. Once more, at 100 mK the decrease in $\rho(T)$ at the lowest temperatures below 4 K (bulk sample) is never observed in $dV/dI(V)^s$ at the lowest bias.

For the case of the thermal regime, the asymmetric parts of the $dV/dI(V)$ (see Fig. 7) would be connected to the ther-

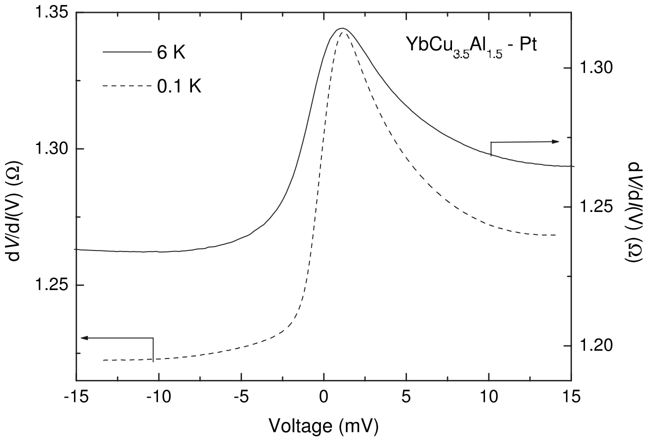

FIG. 8. Characteristic $dV/dI(V)$ of heterocontact $YbCu_{3.5}Al_{1.5}$-Pt at 6 K (solid curve) and at 0.1 K (dashed curve).

mopower of $YbCu_{5-x}Al_x$ and we could estimate their temperature and magnetic-field behavior. Taking into account the work of Mitsuda *et al.*[19] and Zlatić *et al.*[20] for the thermopower of the related $YbCu_{5-x}Ag_x$ system and other Yb-based systems, and the small magnitude of the Cu thermopower at low temperatures, one could expect a negative bulk thermopower for $YbCu_{5-x}Al_x$ with a minimum at low temperatures. The voltage position of the minimum (at about 2 mV) would correspond to a temperature of about 7 K in a temperature scale for thermopower, taking the free-electron Lorenz number. However, this value is questionable for NFL compounds. Moreover, the voltage position of minimum shifts to higher voltages with increasing temperature, which is in contradiction to the thermal regime. The shift to lower voltages is expected with its disappearance when temperature overcomes the minimum in thermopower. Further, the minimum in thermopower of Yb compounds is usually at about 100 K.[20] Therefore, this is another argument to exclude the thermal regime.

Figure 9 shows the magnetic-field influence on the asymmetric part of $dV/dI(V)$ for the same heterocontact $YbCu_{3.5}Al_{1.5}$-Pt as in Fig. 1. One can see that increasing magnetic field suppresses the asymmetry. We suppose that the application of magnetic field destroys the NFL behavior and restores the symmetric shape of $dV/dI(V)$. Taking into account the explanation of the origin of an asymmetry as connected with the density of states,[5] we should expect the linear behavior in the vicinity of zero-bias voltage. Our $dV/dI(V)^{as}$ dependencies show linear behavior below about 1 mV close to x_{cr}. Then we could suppose that the low-voltage behavior of an asymmetry is governed by the mechanism suggested by Shaginyan and Popov.[5]

Presented analysis and high magnetic-field measurements confirm our previously published data.[6,7] We have demonstrated that our spectra are not in the thermal regime, and thus, they are bringing the information about scattering of conduction electrons. The maximum in differential resistance close to 1 mV reflects the increasing magnitude of scattering of conduction electrons. On the other hand the absence of clear maxima in $d^2V/dI^2(V)$ in homocontact as well as in heterocontact arrangement is in agreement with a concept of

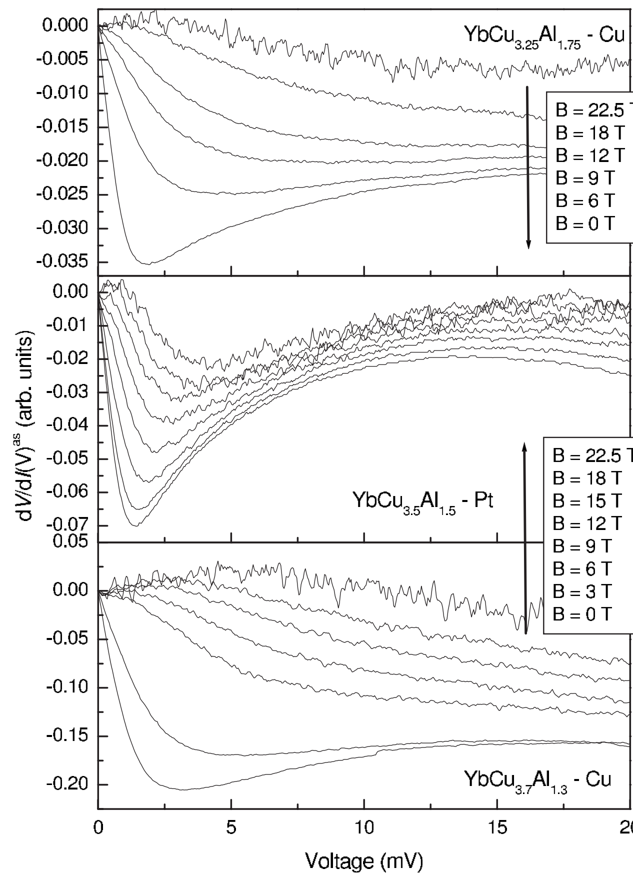

FIG. 9. Characteristic magnetic-field behavior of $dV/dI(V)^{as}$ for heterocontacts with different $x=1.3$, 1.5, and 1.75 at 1.5 K.

absence of classical quasiparticles in NFL systems.[4,5] With increasing applied magnetic field we observed features of $dV/dI(V)$ which are characteristic for Kondo systems.[8,9] One can see that spectra in the NFL regime clearly differ from that of the FL regime. A different type of $dV/dI(V)$ asymmetry has been observed; therefore a different theoretical explanation is required. We could conclude that the observed type of asymmetry in the $dV/dI(V)$ dependencies of heterocontacts is connected with the NFL ground state of $YbCu_{5-x}Al_x$ system. The magnetic-field influence on the asymmetry confirms the suppression of the NFL state and the recovery of the FL state at sufficiently high magnetic fields.

IV. CONCLUSIONS

The PCS measurements of the NFL compounds $YbCu_{5-x}Al_x$ in the vicinity of the QCP (near $x_{cr}=1.5$) have revealed an asymmetric shape of $dV/dI(V)$ of the current-voltage characteristics for the heterocontact arrangement. The application of a magnetic field suppresses the asymmetry, similar to the NFL properties. Therefore the asymmetry has its origin in the NFL state of the sample. The analysis of the point-contact differential resistance curves excludes the thermal regime.

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