Kondo behavior in superconducting $NpPd_5Al_2$

K. Gofryk,[1,*] J.-C. Griveau,[1] E. Colineau,[1] J. P. Sanchez,[1,2] J. Rebizant,[1] and R. Caciuffo[1]
[1]*European Commission, Joint Research Centre, Institute for Transuranium Elements, Postfach 2340, 76125 Karlsruhe, Germany*
[2]*CEA, Institut Nanosciences et Cryogeénie-SPSMS, 38054 Grenoble Cedex 9, France*
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The unconventional superconductor $NpPd_5Al_2$, crystallizing in the tetragonal $ZrNi_2Al_5$-type structure, was studied by means of ^{237}Np Mössbauer spectroscopy, heat capacity, electrical resistivity, magnetoresistivity, and thermoelectric power measurements. The superconductivity is clearly observed below $T_c=5$ K. The Mössbauer studies confirm the absence of ordered magnetic Np moments in both the superconducting and normal phases. The value of the isomer shift suggests that the $5f$-electron count is close to $n_{5f}=4$, in agreement with electron band structure calculations. Above T_c the electronic specific heat, the magnetic contribution to the electrical resistivity, the magnetoresistivity, and the Seebeck coefficient are governed by the interplay of Kondo and crystal-field effects. The Seebeck coefficient, negative in the whole temperature range, indicates electronlike conductivity. This perfectly agrees with the Hall effect data as well as with theoretical predictions.

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I. INTRODUCTION

Transuranium-based intermetallics show a large variety of exotic physical phenomena mainly coming from $5f$ hybridization with both on-site and neighboring ligand states. Depending on the strength of this process unusual behaviors such as long-range magnetic order, Kondo effect, heavy-fermion ground state, valence fluctuations, and/or superconductivity (SC) have been observed. Altogether, they raised a great interest to the study of physical properties of these systems especially during the last years. Recently, this interest was even strongly increased by the discovery of unconventional superconductivity in $NpPd_5Al_2$.[1] This first Np-based superconductor was found to be a paramagnetic heavy fermion with $T_c=4.9$ K and $\gamma=200$ mJ/mol K^2.[1] Moreover, the low-temperature specific heat and NMR measurements agree with the presence of line nodes in the energy gap characteristic of strong coupling d-wave superconductivity.[2,3] Electrical resistivity measurements under magnetic fields indicate the existence of a field-tuned quantum critical point (QCP) around B_{c2} and the recovery of a Fermi-liquid behavior above B_{c2}. Furthermore the application of pressure which decreases both T_c and B_{c2} suggests that $NpPd_5Al_2$ is close to a QCP at ambient pressure.[1,4] Interestingly, while $NpPd_5Al_2$ is an unconventional superconductor, $PuPd_5Al_2$ shows antiferromagnetic ordering at 5.6 K,[5] and UPd_5Al_2 and $AmPd_5Al_2$ exhibit a paramagnetic ground state down to 2 and 3 K, respectively.[6,7]

Despite the large experimental and theoretical works carried out, the nature of the mediating bosons in f-electron-based SC compounds is still unclear and several scenarios were proposed to account for their superconductivity (see Refs. 8–11). Recently, to explain the unconventional superconductivity in Ce- and Pu-based 115 compounds the cooperative two-channel Kondo model was proposed.[12,13] All these findings motivated us to undertake an extensive reexamination of the recently discovered Np-based superconductor in order to look at more details on strong electronic correlations and Kondo characteristics as a possible origin of superconductivity in $NpPd_5Al_2$. In this paper we report the results of Mössbauer spectroscopy, heat capacity, electrical resistivity, magnetoresistivity (MR), and thermoelectric power measurements performed in a wide temperature and magnetic field range (0.5–300 K; 0–14 T).

II. EXPERIMENTAL DETAILS

Polycrystalline samples of $NpPd_5Al_2$ were synthesized by arc melting the stoichiometric amounts of the elements in a Zr-gettered ultrapure argon atmosphere. The samples were examined by both single-crystal and powder x-ray diffraction methods. Moreover, the phase composition has been determined by energy-dispersive x-ray (EDX) analysis on a Philips XL40 scanning electron microscope (SEM). The powder x-ray diffraction was carried out using a Bruker D8 diffractometer (Cu $K\alpha_1$ radiation). From the inner part of the specimen, small single crystals were extracted and examined. The crystal structure refined from the single-crystal x-ray data is shown to be tetragonal with the $ZrNi_2Al_5$ structure type (S.G. $I4/mmm$) with the lattice parameters $a=4.1410$ Å and $c=14.6851$ Å. The values of the parameters obtained are very close to those derived previously.[1] The ^{237}Np Mössbauer spectra were recorded using a sinusoidal drive motion of a ^{241}Am metal source kept at 4.2 K. The temperature of the absorber containing 90 mg Np/cm^2 was varied from 4.2 to 10 K. The velocity scale of the spectrometer was calibrated with reference to an $NpAl_2$ absorber ($B_{hf}=330$ T at 4.2 K). The electrical resistivity and magnetoresistivity were measured from 2 to 300 K by a Quantum Design PPMS-14 setup. Heat-capacity experiments were performed in the temperature range of 0.5–300 K and in magnetic fields up to 14 T using the relaxation method implemented in the PPMS-14 device. The thermoelectric power was measured from 3 to 300 K in a homemade setup using pure copper as the reference material. Due to the contamination risk generated by radiotoxicity of the neptunium element, all operations of preparation and encapsulation have been achieved in special gloveboxes under inert nitrogen atmosphere and using special encapsulation systems. Due to the relatively low self-heating effect of ^{237}Np ($t_{1/2}=2.14\times10^6$ years; W

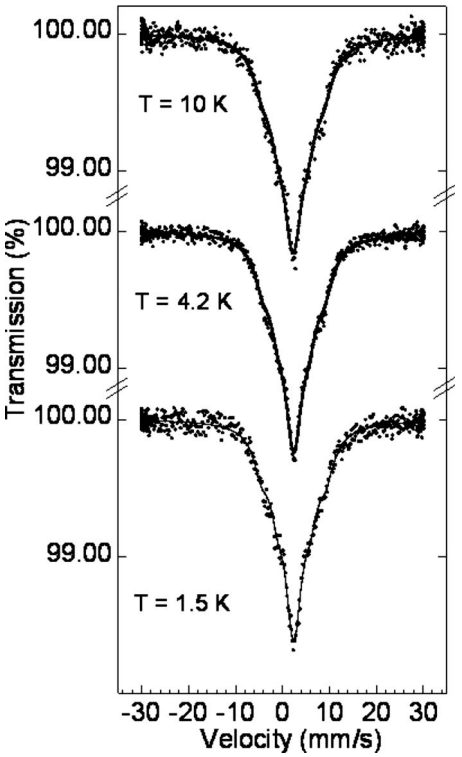

FIG. 1. ^{237}Np Mössbauer spectra of $NpPd_5Al_2$ recorded at $T=1.5$, 4.2, and 10 K. The solid lines represent the fit assuming a pure axial quadrupolar interaction (see text).

$=2.03\ \mu W/g$) the physical properties of $NpPd_5Al_2$ were observable down to 0.5 K.

III. RESULTS AND DISCUSSION

A. ^{237}Np Mössbauer spectroscopy

In order to gain more insights on the electronic and magnetic properties of $NpPd_5Al_2$ we have performed ^{237}Np Mössbauer spectroscopy measurements. Figure 1 shows Mössbauer spectra recorded at different temperatures between 1.5 and 10 K. At 10 K, i.e., in the normal state, the spectrum can be well accounted for by assuming a pure quadrupolar interaction with axial symmetry ($|eQV_{ZZ}| \approx 22.0$ mm/s; $\eta=0$) as expected for the $4/mmm$ point symmetry of the Np site. The position of the central line indicates that the isomer shift amounts to $\delta_{IS} \approx -11.1$ mm/s versus $NpAl_2$.

Since in $NpPd_5Al_2$ the Np atoms are surrounded by Pd atoms in a similar way to that of c-$NpPd_3$ (cubic $AuCu_3$-type structure, see Fig. 2), it was anticipated that their isomer shifts should be close to each other [such a similarity was observed between $NpGa_3$ (Ref. 14) and the $NpTGa_5$ series whose structure contains "$NpGa_3$" blocks[15]]. However the isomer shift of c-$NpPd_3$ [−22 mm/s vs $NpAl_2$ (Ref. 16)] differs considerably from the one observed in $NpPd_5Al_2$. Thus the Np electronic structure in the two compounds should be considerably different. This conclusion is also supported by the comparison of the quadrupole coupling constants eQV_{ZZ}. In c-$NpPd_3$ both the lattice (V_{zz}^{lat}) and $5f$(V_{zz}^{5f})

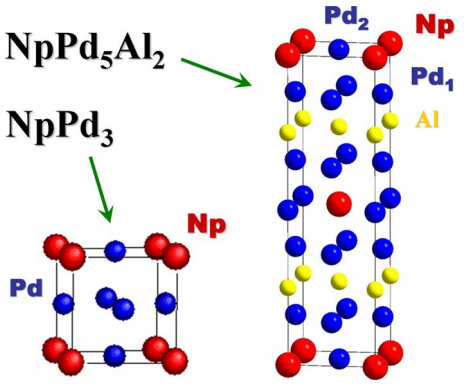

FIG. 2. (Color online) Crystal structures of $NpPd_5Al_2$ and cubic $NpPd_3$ (see text).

contributions to the electric field gradient (efg) are vanishing ($V_{zz}^{lat}=0$ due to the cubic symmetry).[16] In $NpPd_5Al_2$, the local distortion of the "$NpPd_3$" blocks, expressed by the parameter $t=1-(2cz/a)=-1.8\%$ [a and c are the lattice constants and z the positional parameter of the Pd(1) atoms[2]] is rather small. Thus one may safely conclude that the large efg observed in $NpPd_5Al_2$ is essentially due to the $5f$ electronic contribution.

The question now to be answered is what the charge state of the Np ions in $NpPd_5Al_2$ is. No clear conclusion was provided from the magnetic susceptibility data.[1,2] Indeed, the effective magnetic moment ($\mu_{eff} \sim 3\ \mu_B$) extracted from the Curie-Weiss law in a restricted temperature range[1,2] was found to be between the free ion values of the $Np^{3+}(5f^4: 2.68\ \mu_B)$ and $Np^{4+}(5f^3: 3.62\ \mu_B)$ configurations. If one considers the isomer shift value of $NpPd_5Al_2$ it appears that it is strongly shifted from the Np^{3+} ionic value (+37 mm/s vs $NpAl_2$) but close to the Np^{4+} ionic value (−9 mm/s vs $NpAl_2$).[17] It is generally rather intricate to link the isomer shift to a formal charge state in intermetallic compounds owing to the difficulty to estimate the contribution of the conduction electrons to the electronic charge density $\rho(0)$ at the Np nuclei. Nevertheless, systematic Mössbauer studies have shown that the presence of conduction electrons and hybridization effects (i.e., delocalization of the $5f$ electrons) increases $\rho(0)$, i.e., shifts δ_{IS} to the next higher charge state (e.g., $Np^{3+} \rightarrow Np^{4+}$). Thus, δ_{IS} values close to or higher than −9 mm/s can only correspond to intermetallic compounds containing trivalent neptunium.[17] This altogether with the δ_{IS} value observed in c-$NpPd_3$ (see discussion above) suggest that the f-electron count in $NpPd_5Al_2$ is close to $n_{5f}=4$ (i.e., to the Np^{3+} configuration) whereas the Np tetravalent configuration should be present in c-$NpPd_3$. This conclusion agrees well with the recent electronic band structure calculations of $NpPd_5Al_2$ which indicated the presence of a narrow hybridized $5f$ band at the Fermi level occupied by $n_{5f}=3.7$ electrons (see Ref. 18).

The spectra recorded in the superconducting state (i.e., at 4.2 and 1.5 K) are very similar to those taken in the normal state. Thus, in both phases, no magnetic hyperfine splitting is observed indicating that the Np ions do not carry any magnetic ordered moments. A careful analysis of the spectra reveals however a small increase (~0.2 mm/s) in both isomer shift and quadrupolar interaction when $NpPd_5Al_2$ enters in

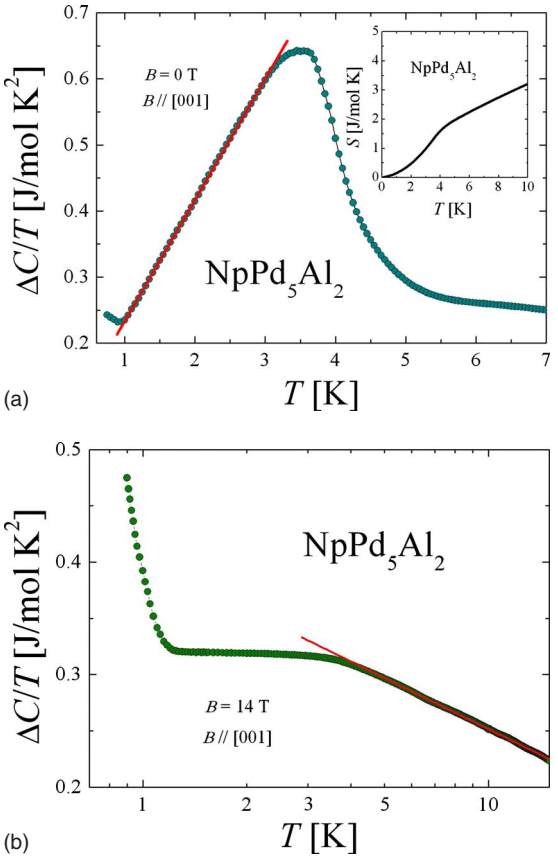

FIG. 3. (Color online) (a) Temperature dependence of the electronic specific heat of $NpPd_5Al_2$ in the form of $\Delta C/T$. The solid line shows the T^2 dependence (see text). Inset: temperature variation of the entropy of $NpPd_5Al_2$. (b) Temperature dependence of the $NpPd_5Al_2$ specific heat ($\Delta C/T$) measured in a magnetic field of 14 T. The red solid line shows a logarithmic dependence above 4 K.

the superconducting phase. This increase, albeit small, emerges from the error bars and, in the case of the isomer shift, cannot be attributed to the second-order Doppler shift which is more than 1 order of magnitude smaller.[19] Therefore both the electronic density at the Np nuclei and the efg due to the $5f$ electrons are affected by the formation of Cooper pairs. This observation could be related to the enhancement below T_c of the electronic specific heat (i.e., of the $5f$ density of states at E_F) as shown below. Indeed an increase in the $5f$ density of states at E_F enhances the shielding of s electrons and hence decreases $\rho(0)$ and the isomer shift becomes larger.[20] This speaks in favor of slightly more localized character of the $5f$ electrons in the superconducting phase.

B. Heat capacity

The temperature dependence of the "electronic" contribution of the specific heat of $NpPd_5Al_2$, $\Delta C(T)$, is shown in Fig. 3(a). This quantity was derived assuming that the phonon part of the specific heat is almost the same in the isostructural compounds $NpPd_5Al_2$ and $ThPd_5Al_2$. Then $\Delta C(T) = C_p(T)^{NpPd_5Al_2} - C(T)_{ph}^{ThPd_5Al_2}$. For this estimate we used the $C(T)_{ph}^{ThPd_5Al_2}$ data reported in Ref. 2.

The superconducting transition manifests itself as a small peak at $T_c \approx 5$ K. Below T_c the specific heat follows a T^2 dependence characteristic of strong coupling [see the solid line in Fig. 3(a)]. The presence of the T^2 behavior down to 1 K could also suggest existence of line nodes in the energy gap. It differs from the T^3 dependence that was observed by Aoki *et al.*[1] Interestingly, the recent NMR studies of this material are in accord with the presence of line nodes in the superconducting order parameter.[3] Notwithstanding, the presence of point nodes cannot be completely excluded (see Ref. 3). The previous studies have revealed that the electronic specific heat is enhanced to about 200 mJ/mol K^2.[1,2] Moreover, taking into account the entropy balance in the superconducting state the low-temperature electronic specific heat was estimated to be of about 390 mJ/mol K^2 at zero K.[1] The so-obtained electronic contribution to the specific heat of $NpPd_5Al_2$ provides the entropy [calculated as $S(T) = \int \frac{\Delta C}{T} dT$] of about 1.9 J/mol K (about 0.3 R ln 2) at the superconducting transition [see the inset in Fig. 3(a)] which indicates that the $5f$ electrons participate to the superconductivity.

In order to look at more details on the electronic contribution to the specific heat of $NpPd_5Al_2$ we have performed the heat-capacity measurements in magnetic fields as strong as 14 T applied along the [001] direction. The low-temperature $\Delta C(T)$ curve is shown in Fig. 3(b). Below 15 K, $\frac{\Delta C}{T}$ taken at 14 T increases with decreasing temperature down to about 3 K, where a "plateau" in $\frac{\Delta C}{T}$ forms with the value of about 325 mJ/mol K^2. This value is close to the one predicted in Ref. 1. Interestingly, the plateau below 3 K is followed by a logarithmic decrease in $\frac{\Delta C}{T}$ [see solid line in Fig. 3(b)], characteristic of non-Fermi-liquid (nFL) state.[21] Indeed, the nFL behavior has been observed below the upper critical field by the electrical resistivity studies under magnetic field and hydrostatic pressure (see Ref. 4). Moreover, it has been shown that with increasing magnetic field this possible nFL state in zero field is replaced by a Fermi-liquid state at fields higher than B_{c2}.[4]

At very low temperatures (below 1 K) a small increase in specific heat is observed (see Fig. 3). In a magnetic field of 14 T the increase is stronger and is shifted toward higher temperatures. This intriguing issue has recently been discussed in Ref. 2 and remains a subject of our ongoing research.

C. Transport properties

The temperature dependences of the electrical resistivity of $NpPd_5Al_2$ together with those of the isostructural nonmagnetic analog $ThPd_5Al_2$ are shown in Fig. 4(a). The $\rho(T)$ of the Th-based compound is typical for nonmagnetic simple metals and may be well described by the Bloch-Grüneisen formula as follows:

$$\rho(T) = \rho_0 + 4R\Theta_R \left(\frac{T}{\Theta_R}\right)^5 \int_0^{\Theta_R/T} \frac{x^5 dx}{(e^x - 1)(1 - e^{-x})}, \quad (1)$$

where ρ_0 is the residual resistivity and the second term describes electron-phonon scattering. Least-squares fit of this

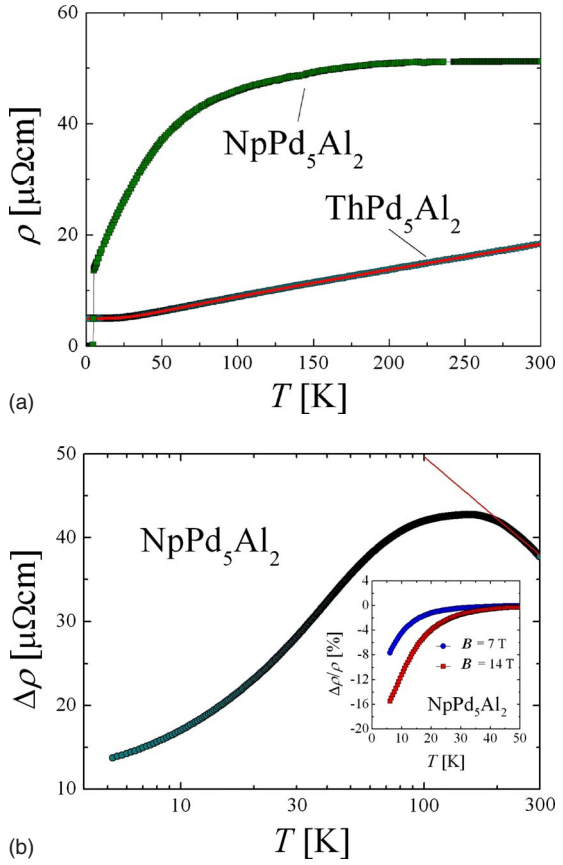

FIG. 4. (Color online) Temperature dependences of the electrical resistivity of $NpPd_5Al_2$ and $ThPd_5Al_2$. The solid line for $ThPd_5Al_2$ is a fit to Eq. (1) (see text). (b) Temperature dependence of the magnetic contribution to the electrical resistivity of $NpPd_5Al_2$. The solid line is a fit using Eq. (2). Inset: temperature variation of the magnetoresistivity taken at $B=7$ and 14 T.

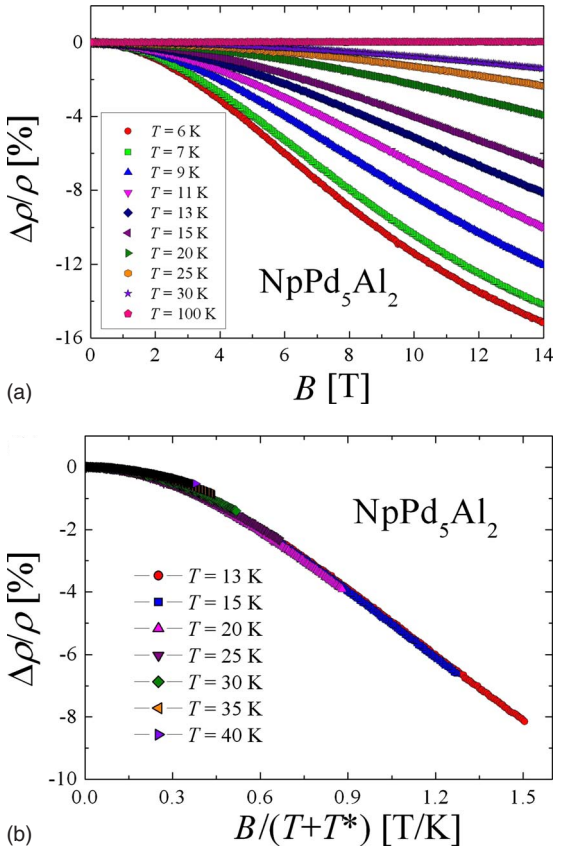

FIG. 5. (Color online) (a) Field dependences of the magnetoresistivity of $NpPd_5Al_2$ recorded at various temperatures. (b) Magnetoresistivity of $NpPd_5Al_2$ plotted as a function of $B/(T+T^*)$ (see text).

expression to the experimental data of $ThPd_5Al_2$ yields the parameters: $\rho_0=4.9$ $\mu\Omega$ cm, $R=4.6\times10^{-2}$ $\mu\Omega$ cm/K, and $\Theta_R=175$ K. The latter parameter, Θ_R, may be considered as an approximation of the Debye temperature Θ_D although Θ_R contains also some contribution related to electron-electron interactions.

While $ThPd_5Al_2$ behaves as a nonmagnetic simple metal, the temperature dependences of the resistivity of $NpPd_5Al_2$, as well as its magnitude, are similar to the one observed in dense Kondo systems. At room temperature the resistivity is 52 $\mu\Omega$ cm. With decreasing temperature the electrical resistivity decreases slightly down to about 100 K; below that it starts to decrease more rapidly down to 5 K where the superconducting transition occurs [see Fig. 4(a)]. To estimate the magnetic contribution to the resistivity, $\Delta\rho(T)$, we assume that the phonon part of the resistivity of $NpPd_5Al_2$ is similar to that of its isostructural counterpart $ThPd_5Al_2$. Thus, we write $\Delta\rho(T)^{NpPd_5Al_2}=\rho(T)^{NpPd_5Al_2}-\rho(T)_{ph}^{ThPd_5Al_2}$. The results are shown in a semilogarithmic scale in Fig. 4(b). The general shape of the $\Delta\rho(T)$ curve with a broad maximum around 150 K reminds the behavior of Ce-, U-, or Pu-based dense Kondo systems with crystal-field interactions (Refs. 22–24). According to the theory developed by Cornut and Coqblin,[25] the position of the maximum in $\Delta\rho(T)$ gives an estimation for the total crystal-field splitting. The value of 150 K is very similar to that obtained from the thermoelectric power data (see below). At high temperature the magnetic contribution to the electrical resistivity of $NpPd_5Al_2$ follows a negative logarithmic dependence characteristic of Kondo systems. Above 200 K the $\Delta\rho(T)$ curve may be well described by the expression,

$$\rho(T) = (\rho_0 + \rho_0^{\infty}) - c_K \ln T, \tag{2}$$

that counts for scattering of conduction electrons by defects, imperfections in the crystal lattice, disordered spins (first term), and Kondo-type impurities (second term), respectively. A least-squares fit of expression (2) to the experimental data yields the parameters: $(\rho_0+\rho_0^{\infty})=99$ $\mu\Omega$ cm and $c_K=11$ $\mu\Omega$ cm.

The temperature dependence of the transverse $(\vec{B}\perp\vec{i})$ magnetoresistivity, defined as $\frac{\Delta\rho}{\rho}=\frac{\rho(B)-\rho(0)}{\rho(0)}$ and measured in applied magnetic fields of 7 and 14 T, is shown in the inset of Fig. 4(b). With decreasing temperature the MR, measured in magnetic field of 14 T (7 T), decreases and reaches a value of about −16% (−8%) at 5 K. Figure 5(a) shows the magnetoresistivity isotherms taken in transverse fields up to 14 T. In agreement with the inset of Fig. 4(b), MR exhibits small and positive values at high temperatures. Below 50 K the magnetoresistivity curves have a characteristic Kondo-type be-

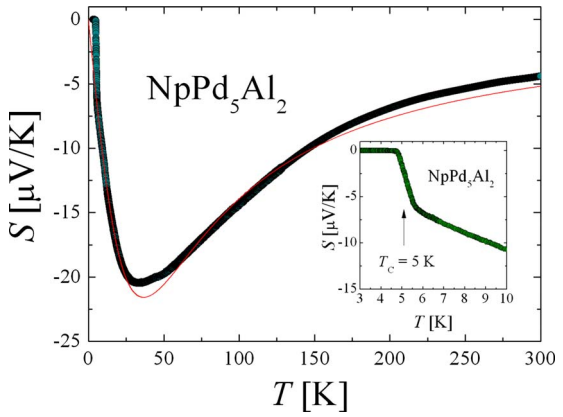

FIG. 6. (Color online) Temperature dependence of the thermoelectric power of $NpPd_5Al_2$. The solid line is a fit of Eq. (6) to the experimental data. Inset: Seebeck coefficient in the vicinity of the superconducting transition.

havior coming from the suppression of incoherent Kondo scattering in applied magnetic field. In the Kondo systems within the Bethe ansatz[26] calculations of the Coqblin-Schrieffer model, it has been shown that MR(B) curves may be scaled using a Schlottmann description,[26,27]

$$\frac{\Delta\rho}{\rho}(B) = f\left[\frac{B}{T+T^*}\right]. \tag{3}$$

It reflects the fact that for the single impurity the physics of the Kondo impurity is dominated by only one energy scale. In the case of $NpPd_5Al_2$ the best overlapping of the MR isotherms was obtained for a characteristic temperature T^* of −3.5 K which is a measure of the Kondo energy scale [see Fig. 5(b)]. However, its negative sign suggests the presence of some ferromagnetic interactions in the superconducting $NpPd_5Al_2$. It is worth to note that similar behavior has been previously reported for the unconventional superconductor UBe_{13} (Ref. 28) and for some Ce- and Yb-based Kondo lattices such as $CeNiGe_3$ (Ref. 22) or YbPtSn.[29]

The temperature dependence of the Seebeck coefficient of $NpPd_5Al_2$ is displayed in Fig. 6. The overall shape and magnitude of the thermoelectric power are similar to the one observed in dense Kondo systems. At room temperature the thermopower is negative and amounts to about −4 μV/K. With decreasing temperature the Seebeck coefficient decreases and shows a distinct minimum of about −20 μV/K near 40 K. Similarly to the electrical resistivity, the superconducting transition manifests itself as a sharp step on the $S(T)$ curve (inset of Fig. 6), and below that the thermopower becomes zero as expected for a superconducting state.

Generally, the diffusion part of the total thermoelectric power is related to the density of states near the Fermi level, to the details of the Fermi surface, and to the different scattering mechanisms. It may be expressed as[30]

$$S(T) = \frac{k_B^2\pi^2 T}{3|e|E_F}\left[\frac{\partial \ln n(E)}{\partial \ln E} + \frac{\partial \ln \nu^2(E)}{\partial \ln E} + \frac{\partial \ln \tau(E)}{\partial \ln E}\right], \tag{4}$$

where $n(E)$, $\nu(E)$, and $\tau(E)$ are the density of states, the velocity of carriers, and the relaxation time of carriers, respectively. E_F is the Fermi energy, k_B is the Boltzmann constant, and $|e|$ is the elementary charge. Thus, the thermoelectric power is very sensitive to details of the electronic band structure and thermopower measurements may be used as a probe of the electronic structure in the vicinity of E_F. Especially, it is the case in compounds that have very narrow bands near the Fermi energy. In order to describe the temperature dependence of the thermoelectric power in such materials a simple phenomenological model has been developed.[31] Taking into account a single-band model [i.e., $\nu^2(E) \sim E$ and $\tau(E) \sim E^{3/2}$], together with the assumption that the so-called Kondo peak is well approximated by a Lorentzian shape as follows:

$$n(E) \propto \frac{\Gamma}{\varepsilon_F^2 + \Gamma^2}. \tag{5}$$

Equation (4) may be expressed by[31,32]

$$S(T) = \frac{aT}{T^2 + b^2} \tag{6}$$

in which the parameters a and b are defined by

$$a = \frac{2\varepsilon_F}{|e|}, \tag{7}$$

$$b^2 = \frac{3(\varepsilon_F^2 + \Gamma^2)}{\pi^2 k_B^2}, \tag{8}$$

where Γ denotes the width of the peak and ε_F its position with respect to the Fermi energy. The least-squares fit of Eq. (6) to the experimental data of $NpPd_5Al_2$ yielded $\varepsilon_F = 0.8$ meV and $\Gamma = 5.7$ meV which are similar to those derived for 4f-and 5f-electron-based compounds with strong correlations (see Refs. 24 and 32–34).

Very recently Behnia *et al.*[35] showed that for a wide range of systems (including heavy fermions, organic, and various oxide families) a remarkable correlation between the thermopower and the electronic specific heat could be established. The dimensionless coefficient relating these two phenomena may be expressed as[35]

$$q = \frac{S}{T}\frac{N_A|e|}{\gamma}, \tag{9}$$

where $N_A|e|$ is the so-called Faraday number ($N_A|e| = 9.6 \times 10^5$ C/mol). It was argued that in the case of strongly correlated electron systems the absolute value of this ratio should remain close to unity.[35] For $NpPd_5Al_2$ the appropriate analysis gives $q \approx 0.66$ which is similar to the one obtained for $CeRu_2Si_2$ or $YbCu_2Si_2$ [see Ref. 35]. The small difference between the obtained and expected value may come from the fact that the temperature range used for such an analysis is too high due to the presence of the superconducting state. Similar problems have been already observed in U-based heavy-fermion superconductors UPt_3 or UBe_{13}.[36,37] In the case of $NpPd_5Al_2$ the low-temperature measurement of the thermoelectric power in the presence of a strong magnetic field, required to destroy superconductivity, would be crucial.

The models discussed are quite simple and neglect some important interactions present in crystalline solids (such as phonon-drag or crystal-field effects) and their influence on the electronic structure. The Coqblin-Schrieffer model[38,39] which describes the Kondo effect in the presence of crystal-field interactions leads to strong temperature variations in the Seebeck coefficient. In particular, a characteristic broad extremum in $S(T)$ is predicted to occur at $T_{\mathrm{ext}} \simeq \frac{\Delta}{3}$, where Δ is the total crystal-field splitting. In the case of $NpPd_5Al_2$ the minimum in $S(T)$ is located around 50 K that implies $\Delta \approx 150$ K consistent with the value obtained from the electrical resistivity data. Recently, Zlatić *et al.*[40–43] extended the Coqblin-Schrieffer model by taking into account not only crystal-field interactions but also hybridization effects between the conduction and f electrons. This approach was successfully applied to describe the temperature dependences of the thermoelectric power of $CeCu_2Si_2$ (Ref. 41) and $CeRu_2Ge_2$,[44,45] and especially their $S(T)$ behavior under pressure. Unfortunately, to our best knowledge there is no model of thermoelectric power for the Np- and Pu-based intermetallics. Clearly, a rigorous theoretical investigation of this issue is strongly needed.

IV. SUMMARY AND CONCLUSIONS

The unconventional superconductor $NpPd_5Al_2$ was synthesized and studied in wide temperature and magnetic field ranges. The compound crystallizes in the tetragonal unit cell of $ZrNi_2Al_5$-type structure with the lattice parameters $a = 4.1410$ Å and $c = 14.6851$ Å. Superconductivity is clearly observed at 5 K. The Mössbauer spectroscopy experiments confirm the absence of ordered neptunium moment in both normal and superconducting phases. The isomer shift value indicates that the Np ions are nearly trivalent as found by electronic band structure calculations. The specific-heat data confirms the presence of a large density of states at the Fermi level. The low-temperature electronic specific heat is highly enhanced and amounts to 325 mJ/mol K^2. It agrees well with the one estimated from previous studies.[1] Moreover, this study confirms the presence of non-Fermi-liquid behavior up to B_{c2} as already suggested from electrical resistivity measurements (see Ref. 4). The magnetic contribution to the resistivity shows behavior characteristic of a Kondo effect in the presence of a crystal-field interaction. Magnetoresistivity data may be well described in term of the Schlottmann formalism with the characteristic temperature $T^* = -3.5$ K. Moreover, the magnitude and overall temperature dependence of the thermoelectric power are typical of dense Kondo systems. The negative sign of the Seebeck coefficient in the whole temperature range indicates that the charge and heat transport is likely to be dominated by electrons. This agrees perfectly with Hall effect experiments[2] as well as with the recent band structure calculations.[18] From the electrical resistivity and thermoelectric power data the crystal-field splitting of the Np multiplet was estimated to be about 150 K, but inelastic neutron-scattering experiments are strongly needed to confirm this point.

Altogether, the large negative paramagnetic Curie temperature (see Refs. 1 and 2), the negative logarithmic slope of the magnetic contribution to the resistivity, the strongly enhanced low-temperature electronic specific heat, and the magnitude and characteristic temperature dependence of the Seebeck coefficient are all indicative that $NpPd_5Al_2$ belongs to a family of dense Kondo systems.

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*Present address: Los Alamos National Laboratory, MPA-10 Division, P. O. Box 1663, MS K764, Los Alamos, NM 87545.

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