Peculiar low-temperature properties of metallic $Nd_{2-x}Ce_xCuO_4$ caused by interactions between Nd moments and conduction electrons

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Low-energy magnetic excitations in $Nd_{2-x}Ce_xCuO_4$ with $0 \leqslant x \leqslant 0.2$ have been investigated by measurements of the specific heat. For all Ce-doped samples ($x \geqslant 0.05$), a linear term γT was found at very low temperatures ($T \lesssim 0.5$ K), with $0.3\ \mathrm{J/K^2} \lesssim \gamma \lesssim 0.6\ \mathrm{J/K^2}$ per mole Nd for $0.05 \leqslant x < 0.14$, and $3\ \mathrm{J/K^2} \lesssim \gamma \lesssim 4\ \mathrm{J/K^2}$ per mole Nd for higher Ce concentrations. In overdoped $Nd_{1.8}Ce_{0.2}CuO_4$, γ is rapidly suppressed in magnetic fields exceeding 1.5 T. The temperature dependence of the electrical resistance parallel to the CuO_2 planes corresponds to that of two-dimensional metals. The magnetoresistance of $Nd_{1.8}Ce_{0.2}CuO_4$ single crystals and c-axis-oriented thin films in magnetic fields parallel to the CuO_2 planes is monotonously negative at temperatures above 1 K. Below about 500 mK, the magnetoresistance is positive in small fields, it goes through a maximum near 2 T, and it is negative for $B > 2$ T. These features are similar to those observed in heavy-fermion systems. In contrast, in $Pr_{1.8}Ce_{0.2}CuO_4$, with nonmagnetic Pr, the magnetoresistance is negative at all temperatures of our measurements down to 20 mK. This proves unambiguously that interactions between conduction electrons and Nd moments give rise to the low-temperature maximum of the magnetoresistance. Furthermore, a comparison of the magnetoresistance with the modification of the specific heat by magnetic fields supports the interpretation of the large linear term as resulting from the interaction between Nd moments and conduction electrons. [S0163-1829(97)05844-X]

I. INTRODUCTION

A remarkable feature of cuprate superconductors is the presence of fluctuating, antiferromagnetically correlated Cu moments in CuO_2 planes in coexistence with superconductivity.[1] The carriers of supercurrents are located in the same CuO_2 planes, in contrast to the coexistence of antiferromagnetic order and superconductivity in classical superconductors in which magnetic rare-earth atoms are well separated from conduction electrons carrying the supercurrents.[2] These observations have given rise to many experimental studies of magnetic correlations and fluctuations in superconducting cuprates by neutron scattering,[1,3] by nuclear quadrupole resonance,[4] and by Raman scattering,[5] to quote a few examples.

In addition, some cuprate superconductors contain magnetic rare-earth atoms, such as the family of compounds $RBa_2Cu_3O_{7-\delta}$ (in short, R-123 compounds), where R stands for Y or any rare earth except Ce, Pr, and Tb, or compounds $R_{2-x}M_xCuO_4$ (with R=Pr, Nd, Sm, or Eu, and M=Ce or Th). In R-123 compounds, magnetic interactions between the rare-earth sublattice and Cu moments are quite weak.[6,7] In these compounds, rare-earth moments have no detrimental effects upon T_c, and the ordering temperatures of rare-earth moments are hardly affected by the oxygen content for the whole range $0 \leqslant \delta \leqslant 1$ in most cases, although Cu moments order antiferromagnetically for $\delta \gtrsim 0.6$, whereas no ordering of Cu moments occurs for higher oxygen contents.

In $R_{2-x}M_xCuO_4$ compounds, the situation seems to be quite similar at first sight. Compounds with Pr, Nd, and Sm have at optimum doping ($x \simeq 0.15$) a T_c in the range 20–25 K (for all three rare earths quoted, variations of T_c in this range have been reported, apparently depending upon preparation parameters, primarily upon the oxygen content of the samples studied), although the magnetic character of these rare earths is quite different: for Pr, the crystal-field ground state is a nonmagnetic singlet, whereas both Nd and Sm carry magnetic moments, with a relatively high ordering temperature, $T_N^{Sm} \simeq 6$ K for Sm_2CuO_4.[8]

A particularly interesting case is the system $Nd_{2-x}Ce_xCuO_4$. Similar to all other cuprates of this family, Cu moments order antiferromagnetically below $T_N^{Cu} \simeq 270$ K in undoped Nd_2CuO_4.[9] Below 10 K, an anomaly in the specific heat was observed,[10] which closely resembles a Schottky anomaly, as first noticed by Boothroyd *et al.*[11] As the first excited crystal-field level lies at an energy of about 16 meV,[11] the anomaly was ascribed to a splitting of the Nd-$4f$ ground-state doublet resulting from interactions between Nd moments and the ordered Cu moments. Adelmann *et al.*[12] showed that this interpretation is in agreement both with the specific-heat data and with the gradual ordering of Nd moments with decreasing temperature revealed by neutron diffraction.[13,14] Thus the low-temperature order of Nd moments is induced by their interaction with the ordered Cu moments.

In Ce-doped compounds $Nd_{2-x}Ce_xCuO_4$, no ordering of Cu moments occurs for $x \gtrsim 0.14$ in well-reduced samples.[15,16] However, measurements of the specific heat of superconducting samples $Nd_{2-x}Ce_xCuO_4$ and $Nd_{2-x}Th_xCuO_4$, both

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with $x=0.15$, revealed an anomaly at low temperatures[10] which is quite similar to the anomaly for undoped Nd_2CuO_4. This highly surprising observation inspired us to study the variation of the low-temperature specific heat with Ce-doping in $Nd_{2-x}Ce_xCuO_4$ with x in the range $0 \leqslant x \leqslant 0.2$.[17] The results showed a systematic shift of the anomaly towards lower temperatures with increasing x, and the appearance of a large linear term γT for $T \lesssim 300$ mK, with $\gamma \simeq 4$ J/K^2 per mole Nd for $x \gtrsim 0.15$. Furthermore, the magnetic susceptibility of overdoped $Nd_{2-x}Ce_xCuO_4$ with $x=0.2$ was found to be Curie-Weiss like for $T \gtrsim 1$ K, and to level off at a Pauli-like constant value for $T \lesssim 300$ mK.[17]

This heavy-fermion-like behavior was interpreted by Fulde and co-workers as arising from the interaction between *stable* Nd moments and *strongly correlated* conduction electrons in the CuO_2 planes.[18,19] Thus the mechanism underlying the heavy-fermion phenomena in $Nd_{2-x}Ce_xCuO_4$ is different from that in classical Kondo lattices. According to this interpretation, the formation of the heavy-fermion state is not affected by the opening of a gap with the onset of superconductivity at $T_c \simeq 20$ K in $Nd_{2-x}Ce_xCuO_4$ with $x=0.15$,[19] in agreement with the experimental results.[17]

An alternative explanation of the large linear term in the specific heat of $Nd_{2-x}Ce_xCuO_4$ is based on competing Nd-Nd and Nd-Cu interactions:[20–22] whereas the Nd-Cu interactions which are dominant in $Nd_{2-x}Ce_xCuO_4$ for $x \lesssim 0.1$ tend to align the moments of nearest Nd-neighbor pairs ferromagnetically, the interaction between these neighbors seems to be antiferromagnetic. Introduction of conduction electrons in the CuO_2 planes by Ce doping reduces the strength of Nd-Cu interactions. At a critical doping level $x_{cr} \simeq 0.1$, ''softening of Nd spin-wave modes connected with a magnetic instability of the forced Nd magnetic structure'' is proposed ''to trigger the large increase of γ for doping levels $x \gtrsim 0.1$.'' [20]

In the present work, we report on supplementary calorimetric measurements, tracing the dependence of γ upon the Ce content x, and measurements of the influence of a magnetic field upon the low-temperature specific heat, both for a polycrystalline sample and for a single crystal. Our main effort, however, was devoted to a study of the involvement of conduction electrons in the heavy-fermion-like state by measurements of the magnetoresistance of single crystals and of c-axis-oriented thin films $Nd_{2-x}Ce_xCuO_4$ with $x \simeq 0.2$, that is, in an overdoped, nonsuperconducting compound at low temperatures (20 mK$\lesssim T \lesssim$4 K). For comparison, we also measured the magnetoresistance of single crystals $Pr_{2-x}Ce_xCuO_4$ with $x \simeq 0.2$ in the same temperature range. This we considered necessary in order to distinguish effects resulting from interactions between Nd moments and conduction electrons from effects due to the tendency toward superconductivity, to fluctuating Cu moments, and to the highly two-dimensional character of these compounds. The latter effects can be expected to be similar in Nd and Pr compounds, whereas there are no interactions between rare-earth moments and conduction electrons in $Pr_{2-x}Ce_xCuO_4$ since Pr is nonmagnetic in these compounds.

Methods employed in sample preparation and experimental techniques are described in Sec. II, followed by the presentation of results in Sec. III. These results and their implications for the low-temperature state of metallic $Nd_{2-x}Ce_xCuO_4$ are discussed in Sec. IV. The paper is concluded by a short summary of our findings in Sec. V.

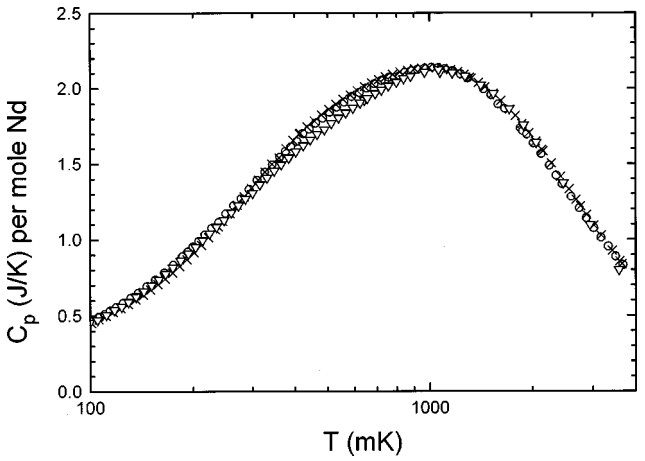

FIG. 1. Low-temperature specific heat of two polycrystalline samples of $Nd_{2-x}Ce_xCuO_4$ with $x=0.2$, (○) prepared by the standard ceramic technique, (×) prepared by the sol-gel process, and (▽) of a single crystal.

II. SAMPLES AND EXPERIMENTAL TECHNIQUES

A. Sample preparation

For the preparation of polycrystalline samples, three different methods were employed in attempts to optimize their homogeneity and properties. At first, samples were prepared by standard ceramic techniques as described in Ref. 17.

An improved homogeneity was expected for samples prepared by the sol-gel route.[23] However, the final products were extremely fine-grained and had a low density ($\simeq$60% of the theoretical density). This had no effect upon the specific heat as shown by a comparison of the specific heat of a compound prepared by standard ceramic procedures with that measured for a sol-gel sample (Fig. 1). However, a study of muon-spin relaxation in material prepared by the sol-gel technique revealed significant changes of muon relaxation rates at low temperatures within a time span of a few months.[24] Presumably, oxygen and possibly water diffuse quite rapidly along grain boundaries which are very numerous in the fine-grained material, leading to changes of its properties.

Finally, samples were prepared employing a variant of ceramic techniques which involves partial melting of the oxides above the eutectic temperature in the last sintering step, as proposed in Ref. 25. In samples prepared by this method, individual grains were substantially larger, the density was above 90% of the theoretical density, and no changes of muon relaxation rates were observed over an extended time span.[24]

Polycrystalline Ce-doped samples were reduced by annealing them for 20 h between 900 and 950 °C in Ar. For all polycrystalline samples, structural parameters and phase purity (with a sensitivity of about 2%) were determined by x-ray diffraction.

Single crystals of both $Nd_{2-x}Ce_xCuO_4$ and $Pr_{2-x}Ce_xCuO_4$ were prepared by directional solidification using CuO flux, as described in Ref. 26. The thickness of

crystals used for measurements of the electrical resistance and of the magnetoresistance was in the range 20–40 μm, the lateral dimensions parallel to the CuO_2 planes were of the order 1–2 mm. Overdoped crystals with $x \simeq 0.2$ were not subjected to a reduction treatment, since indications for the presence of superconducting regions were found in reduced crystals. These had to be avoided for measurements of the electrical resistance and of the magnetoresistance. As shown in Fig. 1, the low-temperature specific heat of single crystals is in good agreement with that of polycrystalline samples with the same level of Ce doping.

Furthermore, *c*-axis-oriented epitaxial thin films of composition $Nd_{1.77}Ce_{0.23}CuO_4$ were fabricated by pulsed-laser deposition on $LaAlO_3$ substrates in an N_2O atmosphere. A detailed description of the deposition process can be found in Refs. 27 and 28. The films were cooled from the deposition temperature (730 °C) to room temperature within 2 h in vacuum. The film thickness varied between 3000 and 6000 Å. Structural parameters and phase purity were verified by x-ray diffraction.

B. Calorimetry

The specific heat was measured with a semiadiabatic heat-pulse calorimeter[29] in a top-loading $^3He/^4He$ dilution refrigerator in the temperature range 100 mK$\lesssim T \lesssim$3 K. A RuO_2 resistance thermometer was used for the determination of the temperature. Magnetic fields up to 4 T were applied for studying the field dependence of the specific heat.

C. Electrical resistance and magnetoresistance

Measurements of electrical transport properties were performed exclusively with single crystals, and with *c*-axis-oriented thin films. Polycrystalline samples could not be expected to give conclusive results because of significant contributions of grain boundaries to the resistance, and because of the very strong anisotropy both of the resistivity[30] and of the magnetic response.[31]

The temperature dependence of the resistance in the temperature range 5 K$\leqslant T \leqslant$300 K was measured in a He-bath cryostat. At lower temperatures, electrical transport properties were determined in the dilution refrigerator at temperatures between 20 mK and about 3 K, with applied fields up to 11 T for magnetoresistance measurements. The current direction was always parallel to the CuO_2 planes, but three different orientations were chosen for the direction of the applied field: (i) parallel to the current direction, (ii) parallel to the CuO_2 planes and perpendicular to the current direction, and (iii) perpendicular to the CuO_2 planes.

In all cases, the four-contact technique was employed. Contacts for feeding current to the crystals were placed on the outermost edges of the crystals in order to contact a large number of conducting CuO_2 planes. The voltage drop was determined with two additional contacts at the surface of the crystals. The crystals were contacted either with silver epoxy or with evaporated gold pads. The contacts were substantially improved by annealing the contacted crystals for 1 h at 400 °C to allow for the diffusion of silver or gold into the surface of the crystals. For thin films, an In-Ag alloy was used for the contacts. In all cases, contact resistances were in the range 1–10 Ω.

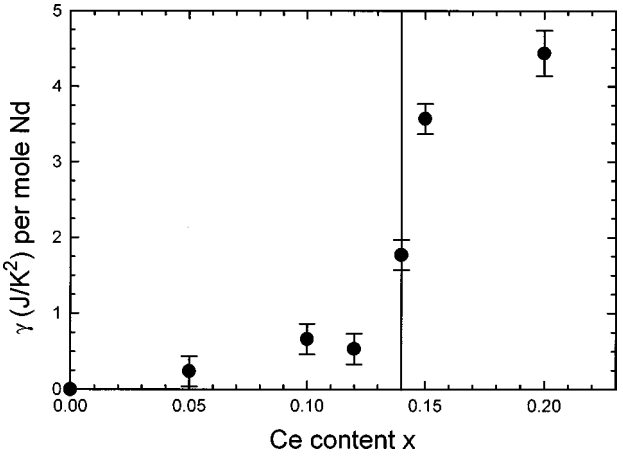

FIG. 2. Variation of γ in $Nd_{2-x}Ce_xCuO_4$ with the Ce content x.

The rather irregular shape of the crystals implied a large uncertainty in converting measured resistances to resistivities. This does not concern variations of the resistivity with temperature and with an applied magnetic field which are of primary interest in the present work.

III. RESULTS

A. Specific heat

1. Dependence of γ upon Ce content x

Nonzero values of the coefficient γ of the linear term in the low-temperature specific heat were found for all doped compounds $Nd_{2-x}Ce_xCuO_4$ with $x \geqslant 0.05$. As shown in Fig. 2, the values of γ are in the range 0.3–0.6 J/K^2 per mole Nd for $0.05 \leqslant x < 0.14$. Near $x \approx 0.14$, γ rises abruptly to values between 3 and 4 J/K^2 per mole Nd for $x \geqslant 0.15$.

For some time after the first report about the occurrence of superconductivity in $Nd_{2-x}Ce_xCuO_4$,[32] compounds with $x \lesssim 0.14$ were classified as semiconductors.[32,33] Since nonzero values of γ are not compatible with a semiconducting character of the samples, we measured the temperature dependence of the resistance of the two samples $Nd_{2-x}Ce_xCuO_4$ with the limiting Ce contents $x = 0.05$ and 0.14. In both samples the resistance increases with decreasing temperature. However, as demonstrated in Fig. 3, the temperature dependence does not correspond to thermal activation of charge carriers over a gap as expected for a semiconductor. Rather, the increase of the resistance is linear in $\ln T$ over a wide range of temperature, in agreement with data reported by Hagen *et al.* for $Nd_{2-x}Ce_xCuO_4$ crystals at low doping.[34] This temperature dependence is characteristic for weak localization in two-dimensional metals,[35] as was further confirmed by magnetoresistance measurements.[34] Hence there is no contradiction between the nonzero values of γ derived from our specific-heat measurements and the character of these compounds which are metallic after all.

2. Modification of C_p by magnetic fields

In Ref. 17, we reported that magnetic fields strongly modify the specific heat of polycrystalline $Nd_{2-x}Ce_xCuO_4$ with $x = 0.2$. In particular, the linear term is considerably reduced in applied fields, from $\gamma \simeq 4$ J/K^2 per mole Nd in

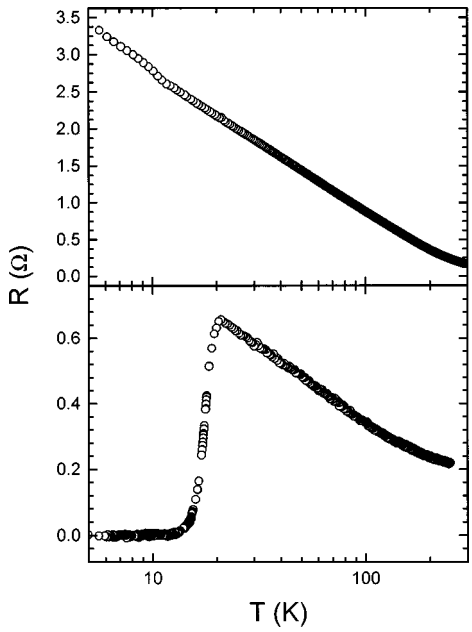

FIG. 3. Temperature dependence of the electrical resistance of $Nd_{2-x}Ce_xCuO_4$ for $x=0.05$ (top) and $x=0.14$ (bottom), plotted vs ln$T$.

zero field to $\gamma \simeq 1$ J/K^2 per mole Nd in a field of 4 T. Now we have repeated these measurements for a single crystal, again with $x=0.2$, in fields directed parallel to the CuO_2 planes. As can be seen in Fig. 4, fields up to 1.5 T hardly affect the specific heat. However, in stronger fields, the value of γ drops precipitously. It disappears completely for $B \geqslant 3.5$ T. In a field of 4 T, the C_p anomaly has the shape of a Schottky anomaly as in undoped Nd_2CuO_4.

The cause for the stronger influence of magnetic fields on the specific heat of a single crystal obviously lies in the considerable anisotropy of the Nd g factor.[31] Its value parallel to the CuO_2 planes is larger than that perpendicular to these planes by a factor of 2. Thus, a magnetic field of 4 T parallel to the CuO_2 planes is sufficient to align all Nd moments in the single crystal, whereas the same field does not align the Nd moments in all grains of the polycrystalline sample.

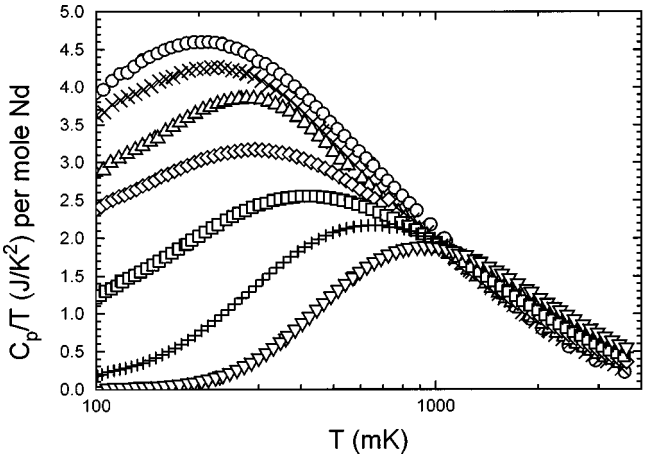

FIG. 4. C_p/T for a single crystal of composition $Nd_{1.8}Ce_{0.2}CuO_4$, (○) in zero field, and in applied fields of (×) 1.5 T, (△) 2 T, (◇) 2.5 T, (□) 3 T, (+) 3.5 T, and (▽) 4 T.

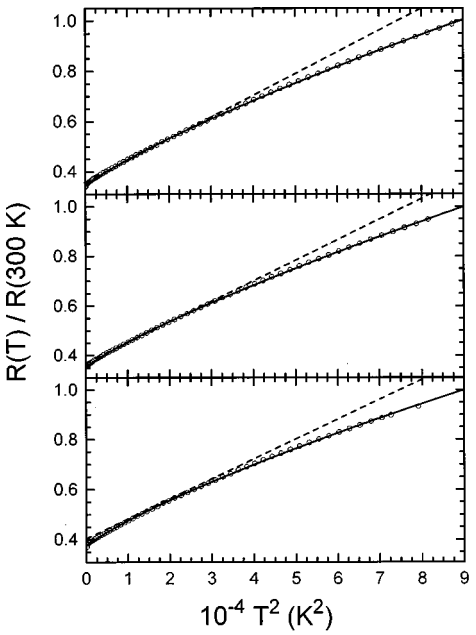

FIG. 5. Temperature dependence of the resistivity of overdoped $Nd_{1.8}Ce_{0.2}CuO_4$ single crystals parallel to the CuO_2 planes, plotted as $\rho(T)/\rho(300\ \mathrm{K})$ vs T^2 in the range from 10 to 300 K. The dashed straight lines demonstrate the approximate T^2 dependence below about 200 K. The solid lines are calculated according to Eq. (1) (Ref. 36).

B. Electrical resistivity: temperature dependence

In the temperature range between about 10 and 300 K, the temperature dependence of the resistivity of $Nd_{1.8}Ce_{0.2}CuO_4$ single crystals showed metallic behavior, varying approximately as T^2 up to about 200 K (Fig. 5, dashed lines). For the entire temperature range of our measurements, the results are very well described by the temperature dependence derived by Giuliani and Quinn[36] for electron-electron scattering in two-dimensional metals:

$$\rho(T)=\rho_0+K(T/T_F)^2\ln(T_F/T) \tag{1}$$

(Fig. 5, solid lines). This is in agreement with observations of other groups.[37,38] From our data, we derive values of T_F in the range 3400–4500 K, not far from the value $T_F=3000$ K substituted by Tsuei, Gupta, and Koren and Crusellas *et al.*,[37] substantially lower than $T_F=29\,000$ K reported by Seng *et al.*[38]

For all $Nd_{1.8}Ce_{0.2}CuO_4$ crystals and films measured, $\rho(T)$ had a minimum between 4 and 10 K (Fig. 6), with an approximately logarithmic increase toward lower temperatures (Fig. 7). For some of the crystals, a downturn of $\rho(T)$ was found below about 1 K (Fig. 7). We do not know the cause of this downturn. It may be a signature of filamentary superconductivity due to a somewhat inhomogeneous distribution of Ce in these crystals. The temperature dependence of the resistivity of our $Pr_{1.8}Ce_{0.2}CuO_4$ single crystals was qualitatively similar to that measured for $Nd_{1.8}Ce_{0.2}CuO_4$, but the resistivity minimum was at a higher temperature, between about 50 and 150 K.

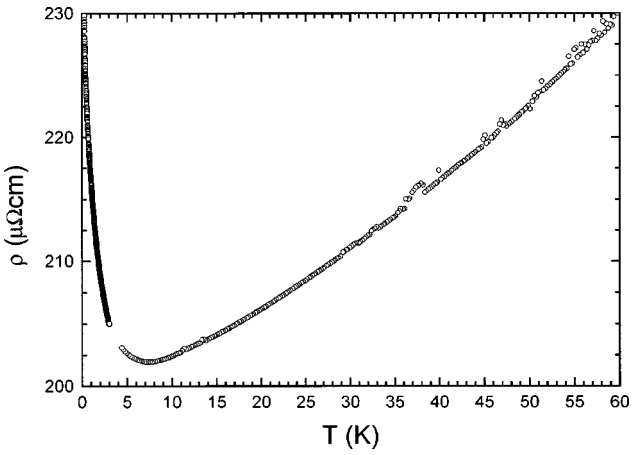

FIG. 6. Temperature dependence of the resistivity of one of the crystals in a restricted temperature range illustrating the resistivity minimum below 10 K.

C. Magnetoresistance

1. Magnetic field parallel to the CuO_2 planes

For all crystals of overdoped, nonsuperconducting $Nd_{1.8}Ce_{0.2}CuO_4$ studied in fields parallel to the CuO_2 planes, we obtained magnetoresistance curves similar to those shown in Fig. 8, top. The same is true for oriented thin films. Examples for results obtained with thin films are displayed in Fig. 9. No differences were found between the magnetoresistance measured with fields parallel or perpendicular to the current direction. Obviously, the magnetoresistance does not depend upon the angle between the directions of the current and of the magnetic field as long as both are parallel to the CuO_2 planes. This is an indication that the observed magnetoresistance is connected with magnetic scattering of the conduction electrons. In crystals with a downturn of $\rho(T)$ at very low temperatures, a strong increase of the resistance was found in small fields ($B \lesssim 0.2$ T). In larger fields, the magnetoresistance of these crystals was similar to that observed for crystals with a monotonous increase of $\rho(T)$ toward low temperature.

In all cases, we found the behavior as illustrated in Figs. 8 (top) and 9: (1) a monotonous, negative magnetoresistance at temperatures above about 1 K;[39] and (2) a positive magnetoresistance in low fields, a pronounced maximum close to a field of 2 T, and a negative magnetoresistance in higher fields at temperatures below about 500 mK.

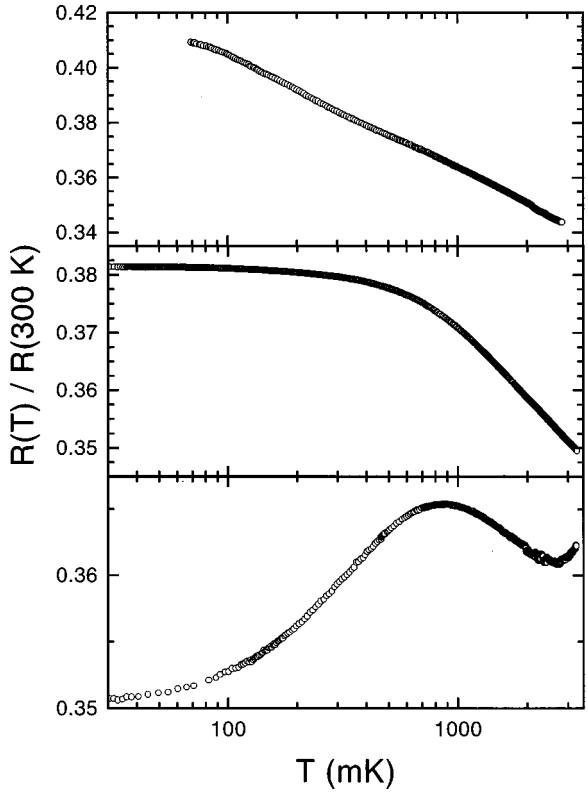

FIG. 7. Temperature dependence of the resistivity of the same crystals at very low temperatures ($T < 4$ K), plotted vs ln$T$.

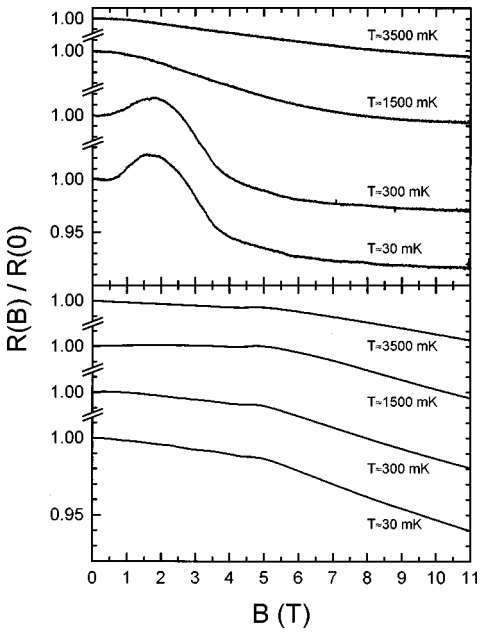

FIG. 8. Magnetoresistance of $R_{1.8}Ce_{0.2}CuO_4$ single crystals in fields parallel to the CuO_2 planes, perpendicular to the current direction, measured at the temperatures indicated in the figure. Top: R = Nd; bottom: R = Pr.

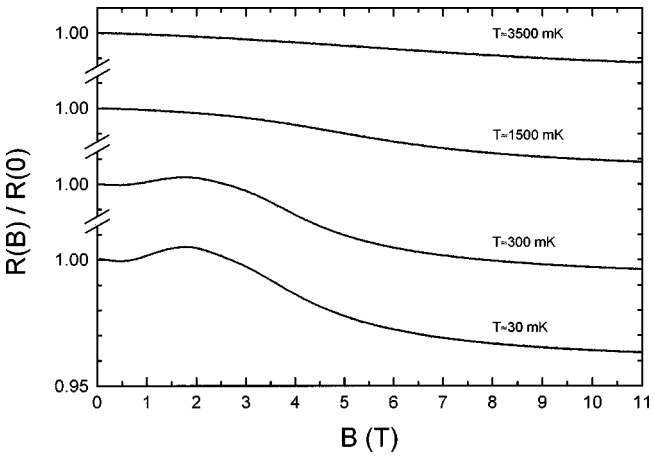

FIG. 9. Magnetoresistance of a $Nd_{1.77}Ce_{0.23}CuO_4$ thin film in fields parallel to the CuO_2 planes, perpendicular to the current direction, measured at the temperatures indicated in the figure.

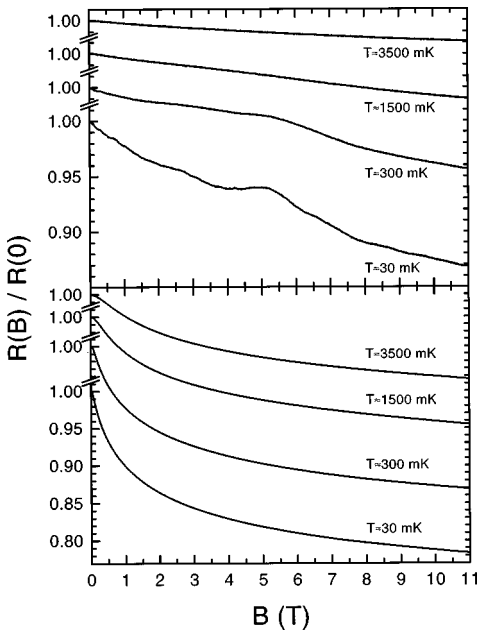

FIG. 10. Magnetoresistance of $R_{1.8}Ce_{0.2}CuO_4$ single crystals in fields perpendicular to the CuO_2 planes, measured at the temperatures indicated in the figure. Top: $R=$Nd; bottom: $R=$Pr.

In contrast, the magnetoresistance of $Pr_{1.8}Ce_{0.2}CuO_4$ was found to be monotonously negative in the whole temperature range of our measurements (Fig. 8, bottom). The most distinct feature of these magnetoresistance curves is a change of the slope dR/dB near 5 T which becomes more pronounced as the temperature is lowered. As for the Nd compound, no difference was observed between the magnetoresistance in fields parallel and perpendicular to the current direction for fields parallel to the CuO_2 planes.

As we pointed out in Sec. I, the only difference between $Nd_{2-x}Ce_xCuO_4$ and $Pr_{2-x}Ce_xCuO_4$ which is of importance for the magnetoresistance is the presence of magnetic Nd moments in the Nd compounds, and the absence of rare-earth moments in $Pr_{2-x}Ce_xCuO_4$. Thus the observed differences between the results for these compounds, in particular the maximum close to 2 T in the magnetoresistance of $Nd_{2-x}Ce_xCuO_4$ below 500 mK, obviously result from the interaction between conduction electrons and the Nd moments.

2. Magnetic field perpendicular to the CuO_2 planes

Magnetoresistance curves in fields perpendicular to the CuO_2 planes differ significantly from those in parallel fields both for $Nd_{1.8}Ce_{0.2}CuO_4$ (Fig. 10, top) and for $Pr_{1.8}Ce_{0.2}CuO_4$ (Fig. 10, bottom). In the Pr compound, the magnetoresistance in perpendicular fields has an appearance typical of the magnetoresistance of two-dimensional metal films in the case of weak localization[35] over the whole field range of our measurements. In the magnetoresistance of $Nd_{1.8}Ce_{0.2}CuO_4$, the rapid downturn of the resistance in low fields (<1 T), in particular at temperatures below 1 K, may arise from localization effects as well. Also in higher fields, the magnetoresistance of the Nd compound is negative, but with a weaker field dependence than that observed for the Pr compound. A marked shoulder near 5 T in the data obtained below 1 K most probably is the counterpart of the maximum near 2 T of the magnetoresistance in fields parallel to the CuO_2 planes, shifted to a larger field value because of the smaller g-factor of Nd perpendicular to the CuO_2 planes.

IV. DISCUSSION

Our results for the magnetoresistance of overdoped $Nd_{2-x}Ce_xCuO_4$ with $x=0.2$ have exactly the features which are characteristic of classical heavy-fermion systems:[40] negative magnetoresistance above a characteristic temperature T^* (in classical heavy-fermion systems, T^* is related to the Kondo temperature T_K), and below T^* a positive magnetoresistance in small fields with a maximum at a characteristic field B^* which is related to T^*. The positive magnetoresistance of heavy-fermion compounds for $B<B^*$ is generally ascribed to the destruction of the coherent state by the applied field, whereas the negative magnetoresistance for $B>B^*$ and for $T>T^*$ results from the scattering of conduction electrons by Kondo impurities.[40]

This negative magnetoresistance is very well described by the Bethe-ansatz solution of the $S=\frac{1}{2}$ Coqblin-Schrieffer model[41] for the magnetoresistance of independent Kondo impurities.[40] A numerical calculation of transport properties of heavy-fermion systems in Ref. 42 reproduces quite well at least the qualitative features of the observed magnetoresistance of heavy-fermion systems at low temperatures in low fields.

Although the mechanism leading to heavy-fermion-like properties of metallic $Nd_{2-x}Ce_xCuO_4$ is different from that in classical Kondo lattices,[18,19] there is a remarkable similarity of many properties of $Nd_{2-x}Ce_xCuO_4$ with those of classical heavy-fermion systems.

However, the results of our low-temperature resistance measurements are not in accordance with the T^2 dependence expected for a coherent heavy-fermion state, as is clearly seen in Fig. 7. The variation of the low-temperature behavior of $R(T)$ from one crystal to another shows that several different mechanisms contribute to the low-temperature resistivity. First, coherence cannot be perfect in the materials studied because of the Ce dopants. As demonstrated by Onuki and Komatsubara[43] for the system $Ce_{1-x}La_xCu_6$, the low-temperature resistivity is considerably affected even by small concentrations of La. Furthermore, the pronounced two-dimensional character of the cuprates under study favors weak localization which seems to dominate $R(T)$ at the lowest temperatures for the crystal whose resistance is displayed in the uppermost part of Fig. 7. Finally, filamentary superconductivity in the Ce-poor regions of some of the overdoped crystals may influence $R(T)$ (Fig. 7, lowest panel).

A comparison of the effects of magnetic fields upon the specific heat, especially upon the value of γ, (Fig. 4) with the magnetoresistance curves (Fig. 8), reveals a complete coincidence of the ranges both of the magnetic field ($\sim$1.5 to $\sim$3.5 T) and of the temperature (below $\sim$500 mK), where the value of γ is strongly suppressed and where the magnetoresistance goes through a maximum. This coincidence lends further support to the interpretation of the low-temperature properties of $Nd_{2-x}Ce_xCuO_4$ as indications for the formation of a coherent heavy-fermion state arising from

the interaction between Nd moments and conduction electrons.

The coupling between conduction electrons and Nd moments is based on the exchange interactions between Nd and Cu moments. Thus the behavior of the Cu-spin system plays an essential role for the phenomena described here. Our measurements provide no direct information about the dynamics of Cu spins. However, important clues on this have been obtained by studies of muon-spin relaxation in $Nd_{1.8}Ce_{0.2}CuO_4$ at very low temperatures (down to 70 mK) in zero field and in applied longitudinal fields.[44] First, the results of these experiments exclude the presence of static magnetic moments either in a magnetically ordered state or frozen in a spin-glass state down to 70 mK. Second, the spin-correlation time τ_c—a measure for the vanishing of the autocorrelation function $\langle \mathbf{S}_i(t)\cdot\mathbf{S}_i(0)\rangle$ for times $t>\tau_c$—has a surprisingly large value, $\tau_c \approx 10^{-9}$ s. Finally, the average hyperfine field at the muon site implies a Nd moment of about 0.2 μ_B, strongly reduced compared to $\mu_{Nd} \simeq 1.3\ \mu_B$ in Nd_2CuO_4.[14]

Since the muon-spin relaxation is primarily caused by Nd moments, the spin-correlation time τ_c quoted refers to the Nd moments. However, the coupling strength between Cu and Nd moments as measured by the splitting of the Nd ground-state doublet, corresponding to about 1 K, is larger by two orders of magnitude than $\hbar/\tau_c$. Hence the spin-correlation time of the Cu moments is expected to be comparable to that of the Nd moments. Similarly slow dynamics of Cu spins was recently derived from electron spin resonance of Gd in $La_{1.65}Gd_{0.01}Eu_{0.24}Sr_{0.1}CuO_4$. In these experiments, it was found that the spin-fluctuation frequency $1/\tau_c$ tends toward 10^{10} s^{-1} at low temperatures.[45]

The large value of τ_c can be explained by the presence of domains of antiferromagnetically correlated Cu moments. Excitations of Cu spins within domains have a spin-wave-like character with high frequencies, but low amplitude. These excitations reduce the value of the autocorrelation function, but they cannot lead to its vanishing. This can only be accomplished by moment reversals in a time shorter than τ_c. Thus τ_c is determined by the relatively slow motion of domain walls which does reverse the direction of Cu moments. This picture resembles theoretical considerations[46] on a domain-wall fluid as the dynamical counterpart to striped domains observed in Nd-doped $La_{2-x}Sr_xCuO_4$ by neutron scattering.[47] It appears possible that the situation in $Nd_{2-x}Ce_xCuO_4$ in this respect is quite similar to that found in $La_{2-x}Sr_xCuO_4$.

Direct and detailed information about magnetic correlations and excitations can be derived from neutron-scattering data. In an early neutron-scattering study of single crystals $Nd_{1.85}Ce_{0.15}CuO_4$, correlation lengths of Cu moments were determined, and the dynamics of Cu moments was studied by inelastic scattering with energy transfer $\geqslant 3$ meV.[3] These energies by far exceed the energy range of the excitations which give rise to the anomaly in the low-temperature specific heat observed in our study.

Only recently, results of high-resolution inelastic neutron-scattering studies of $Nd_{2-x}Ce_xCuO_4$ with $0\leqslant x\leqslant 0.15$ at very low energies and low temperatures have been reported.[20,22,48] In undoped polycrystalline Nd_2CuO_4, the inelastic scattering intensity was found between about 0.2 and 0.8 meV, with a clear gap below 0.2 meV.[48] Experiments with a Nd_2CuO_4 single crystal revealed several dispersive branches in the energy range from 0.2 to 0.8 meV.[20,22] The dispersion is ascribed to Nd-Nd interactions, and the multitude of branches arises from the eight magnetic Nd sublattices in the noncollinear antiferromagnetic structure.[21] The average energy of the excitations, $\simeq 0.63$ meV, is caused by the mean field acting on the Nd moments due to the Nd-Cu interactions. In Ce-doped samples, both in single crystals[22] and in polycrystalline materials,[48] these excitations are shifted to lower energies. For $x=0.15$, the scattering has essentially quasielastic character.

These results are in good agreement with those of our measurements of the specific heat.[17] However, up to now, the neutron-scattering results provide no possibility to decide whether the underlying mechanism involves interactions between Nd moments and conduction electrons,[18] or whether softening of the excitations due to competing Nd-Cu and Nd-Nd interactions is the cause.[21] Our magnetoresistance results definitely favor the first-mentioned mechanism.

V. SUMMARY

We have investigated compounds $Nd_{2-x}Ce_xCuO_4$ with $0\leqslant x\leqslant 0.2$ by measurements of the specific heat, of the temperature dependence of the electrical resistance, and of the magnetoresistance in fields up to 11 T.

For all Ce-doped compounds ($x\geqslant 0.05$), a linear term γT was found in the specific heat at low temperatures ($T\lesssim 0.3$ K). For $0.05\leqslant x<0.14$, the value of γ is in the range 0.3–0.6 J/K^2 per mole Nd. In this range of Ce concentrations, a logarithmic increase of the electrical resistance with decreasing temperature points to weak localization in a two-dimensional (2D) metal, in agreement with results reported by Hagen *et al.*[34] Near $x=0.14$, γ rises abruptly to 3–4 J/K^2 per mole Nd for $0.15\leqslant x\leqslant 0.2$. For overdoped crystals with $x=0.2$, from 10 to 300 K, the temperature dependence of the electrical resistance parallel to the CuO_2 planes is very well described by Eq. (1), derived for dominant electron-electron scattering in a 2D metal.[36] For all overdoped crystals, the resistance goes through a minimum between 4 and 10 K, with an approximately logarithmic increase toward lower temperatures.

Magnetic fields exceeding 1.5 T drastically reduce the low-temperature specific heat. For single crystals of overdoped $Nd_{1.8}Ce_{0.2}CuO_4$ in a magnetic field parallel to the CuO_2 planes, $\gamma=0$ for $B\geqslant 3.5$ T.

The magnetoresistance of $Nd_{1.8}Ce_{0.2}CuO_4$ single crystals and thin films in fields parallel to the CuO_2 planes matches exactly that reported for heavy-fermion systems:[40] It is monotonously negative for $T>1$ K; below about 500 mK, it is positive in small fields, goes through a maximum near 2 T, and is negative in higher fields. In contrast, the magnetoresistance of $Pr_{1.8}Ce_{0.2}CuO_4$ crystals is negative at all temperatures of our measurements down to 20 mK. The only difference between these compounds is the presence of magnetic rare-earth moments at Nd, whereas Pr is nonmagnetic. Thus the observed differences of the magnetoresistance, in particular the maximum near 2 T, obviously are due to the interaction between conduction electrons and Nd moments. The coincidence of the field and temperature ranges where the value

of γ is suppressed and where the magnetoresistance goes through a maximum supports the interpretation of the large linear term in the specific heat of $Nd_{2-x}Ce_xCuO_4$ in terms of the formation of a coherent heavy-fermion state arising from the interaction between the Nd moments and conduction electrons in the CuO_2 planes.[18]

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