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Low-temperature specific heat of $Nd_{2-x}Ce_xCuO_{4-y}$ single crystals ($x=0$ and 0.15) in applied magnetic fields

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Results are presented of low-temperature (0.3–6 K) specific-heat measurements on bulk single crystals of $Nd_{2-x}Ce_xCuO_{4-y}$ ($x=0$ and 0.15) in applied magnetic fields up to 8 T. The magnetic-field effect shows a highly anisotropic behavior: for both compounds only minor effects on the specific-heat curve have been found for fields applied parallel to the crystallographic *c* axis, whereas if the field is applied in the *ab* plane, a substantial broadening and shift of the zero-field maximum to higher temperatures is observed. A satisfactory quantitative description in terms of crystal-field and exchange interactions is given for the experimental data of the $x=0$ crystal. It is shown that application of the same description to the $x=0.15$ case is inadequate. [S0163-1829(98)08509-9]

I. INTRODUCTION

Since their discovery,[1] the electron-doped $Nd_{2-x}Ce_xCuO_{4-y}$ compounds (with $x\leqslant 0.22$; y is the oxygen deficiency) have been the subject of intensive investigation. As is now known,[2–7] these compounds exhibit complex magnetic properties. We mention here that the parent compound (with $x=0$; in this paper also called the ''undoped'' compound) is an insulator. The so-called ''doped'' compounds are superconducting for values of x between 0.14 and 0.18 and $y\approx 0.02$. Figure 1 shows the tetragonal crystal structure for the $Nd_{2-x}Ce_xCuO_{4-y}$ compounds[1] in combination with the noncollinear spin structure proposed[3–6] for both the Cu and Nd sublattices in the undoped compound. In this compound the Cu sublattice orders antiferromagnetically at $T_N\approx 250$ K in the noncollinear spin structure mentioned. On further decreasing the temperature the Cu moments undergo two spin-reorientation transitions: one at $T_{r1}=75$ K from the noncollinear structure into a collinear one (as defined in the caption of Fig. 1), and another at $T_{r2}=30$ K back to the original noncollinear configuration. Considering in Fig. 1 the ferromagnetically ordered Nd_2Cu chains in both the noncollinear and collinear structure, we note that, presumably due to a ferromagnetic Nd-Cu interaction, in both structures each Nd moment has the same orientation as the Cu neighbor along the *c* direction. Furthermore, in the superconducting doped compounds the long-range antiferromagnetic order of the Cu moments is destroyed.[7]

The crystalline electric field (CEF) in the $Nd_{2-x}Ce_xCuO_{4-y}$ compounds has been studied extensively by inelastic neutron scattering (INS) (Refs. 8–14) and Raman scattering.[15–18] While consistency in the energy-level schemes has been achieved, there is a large divergence in the CEF-parameter sets proposed among the different groups of authors to interpret these energy-level schemes.

Measurements of the specific heat, particularly in magnetic fields, can provide valuable additional information on the magnetic-ordering phenomena as well as on the magnetic interactions in these compounds. These measurements provide another possibility to verify the CEF parameters.

To our knowledge, low-temperature specific-heat data of the $Nd_{2-x}Ce_xCuO_{4-y}$ compounds have been reported on a single-crystalline sample with $x=0$ in the temperature range above 1.6 K at zero magnetic field,[19] and on polycrystalline samples with $x=0-0.2$ down to the millikelvin temperature range in applied magnetic fields up to 6 T.[20–24] The quantitative description of the specific-heat data, however, cannot be considered as completed. In this paper, we report on specific-heat measurements of $Nd_{2-x}Ce_xCuO_{4-y}$ single crystals with $x=0$ and 0.15, in the temperature range between 0.3 and 6 K in magnetic fields up to 8 T, applied both parallel and perpendicular to the crystallographic *c* axis. We present a description of these experimental data in terms of

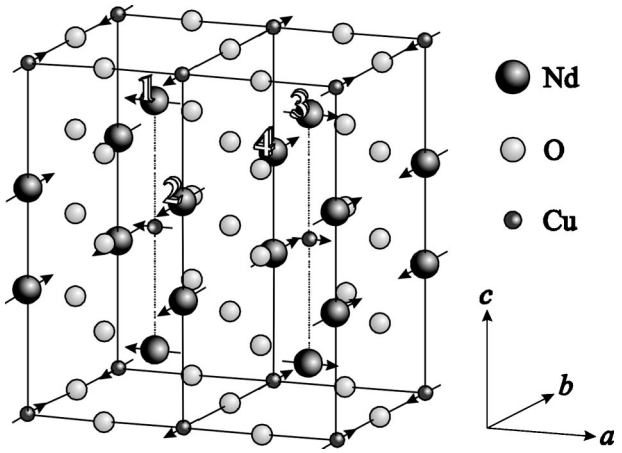

FIG. 1. Tetragonal crystal structure (two chemical unit cells) for $Nd_{2-x}Ce_xCuO_{4-y}$ and noncollinear spin structure of Nd_2CuO_{4-y}. The Nd subsystem is divided into 4 Nd sublattices, denoted 1, 2, 3, and 4; Nd ions that in the figure have the same moment, belong to the same sublattice. (In a similar way there are 4 Cu sublattices.) In the so-called collinear spin structure the moments in every second CuO_2 layer rotate over 90°, keeping their antiferromagnetic arrangement; also in this structure the orientation of the Nd ions is such that the moments in each Nd_2Cu chain along the *c* axis are ferromagnetically ordered.

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crystal field and exchange interactions.

The organization of the paper is as follows. In Sec. II, experimental details are given and in Sec. III, the results of the specific-heat measurements. The model for our calculations is described in Sec. IV. Results of these calculations are shown in Sec. V and discussed in Sec. VI. The paper ends with some concluding remarks (Sec. VII).

II. EXPERIMENTAL DETAILS

Single crystals of $Nd_{2-x}Ce_xCuO_{4-y}$ (with $x=0$ and 0.15) have been grown by the traveling solvent floating zone (TSFZ) method in an oxygen-poor ambient atmosphere using a NEC SC-N35HD light-image furnace. For the compound with $x=0.15$, a multiscan growth method[25,26] was applied. The as-grown crystalline materials (that each consist of only a few large single crystals) were characterized by neutron-diffraction, x-ray Laue- and powder-diffraction, electron-probe micro analysis (EPMA), and by ac-susceptibility and resistivity measurements. As confirmed by neutron-diffraction experiments,[27] single-crystalline grains with a volume in the order of 400 mm^3 and 150 mm^3 for $x=0$ and 0.15, respectively, have been obtained. X-ray powder diffraction has revealed that for both compounds the as-grown crystalline materials are of the same phase as the polycrystalline starting materials. The lattice parameters have been determined by x-ray powder diffraction. For $x=0$, we found $a=3.9394$ Å, $c=12.1557$ Å for the as-grown crystalline material, slightly smaller than $a=3.9419$ Å, $c=12.1618$ Å for the sintered polycrystalline starting material. For $x=0.15$, the corresponding values are $a=3.9416$ Å, $c=12.0552$ Å, and $a=3.9419$ Å, $c=12.0669$ Å, respectively. These values are in good agreement with the published data (see, e.g., Ref. 1). Ac-susceptibility measurements on as-grown crystalline $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ samples showed a broad superconducting transition with an onset temperature at about 16 K. This transition temperature is confirmed by resistivity measurements where a metallic behavior of the resistivity in the temperature range above the superconducting transition has been found. These results imply that the as-grown crystalline material is oxygen-deficient (see, e.g., Ref. 1). After an additional oxygen-reduction treatment at 900 °C for 12 h in flowing nitrogen, the crystalline sample shows a superconducting transition with an onset temperature at 20 K, the same temperature as observed also for the starting material after a similar oxygen-reduction annealing. No attempt was undertaken to determine the actual oxygen content in any of our samples. EPMA results have revealed that the composition of the as-grown crystalline sample is in close agreement with that of the starting polycrystalline material. Other details of the crystal growth and characterization will be published elsewhere.[26]

For the specific-heat measurements, single crystals with a mass of 276 mg for $x=0$ and 250 mg for $x=0.15$, were cut from the last part of the as-grown boules. The zero-field specific heat was measured by an adiabatic method from 1.8 to about 140 K. The measurements in magnetic fields were carried out for the low-temperature region between 0.3 and 6 K by the same adiabatic method in fields from 0 to 8 T. With the magnetic-field direction in the *ab* plane, actually along the [110] direction as evidenced by a neutron-diffraction experiment,[28] the specific heat has been measured at field strengths up to 8 T. In case the field direction is parallel to the *c* axis, we have carried out the measurement only for field strengths of 2 and 4 T. In our attempt to carry out the measurement on the undoped crystal ($x=0$) at 6 T with this orientation of the applied field, the sapphire plate used as the sample holder was broken due to the torque exercised by the magnetic field on the sample.

In order to obtain an impression of the accuracy of our experimental method, the results of specific-heat measurements on a 1 g 4N copper sample were compared with literature values. (Nowhere in the temperature range 1.2–20 K the specific heat of this copper sample exceeds the specific heat of the $Nd_{2-x}Ce_xCuO_{4-y}$ samples used in our experiment. In the range 1.2–4 K, where the specific heat of our samples goes through the maximum to be analyzed in the present paper, the specific heat of this copper sample is less than 5% of that of our samples. When measuring our samples the empty-holder contribution to the specific heat is less than 1%.) The accuracy of the measurement on the copper sample turned out to be better than 1% in the temperature range 1.2–20 K, and becomes worse above 20 K due to radiation losses.

III. EXPERIMENTAL RESULTS

A. Zero-field specific heat

In Fig. 2, we show the zero-field specific heat of the $Nd_{2-x}Ce_xCuO_{4-y}$ single-crystalline samples in the temperature range up to 30 K. In close agreement with the results published on polycrystalline samples,[20–24] the zero-field data for the Nd_2CuO_{4-y} single crystal show a maximum of 3.44 J/(K mol-Nd) near 1.6 K. For the sample with $x=0.15$, the maximum is suppressed considerably to 2.53 J/(K mol-Nd) and shifted to about 1.1 K. Furthermore, the curve is dramatically broadened. For low temperatures, below 4 K, say, the specific heat is interpreted mainly as a Schottky curve connected with the splitting by the molecular field of the ground-state doublet of the Nd^{3+} ion. In the temperature range above 4 K, there are certainly other contributions (from the electronic system and lattice) to the measured specific heat—also for our undoped crystal.

Our interpretation of the specific-heat results is confirmed by recent high-resolution neutron-scattering experiments[27] on single-crystalline samples prepared from the same batches. In these experiments, in the undoped compound an excitation at about 0.39 meV has been found that is ascribed to the splitting of the ground-state doublet due to the molecular field; for the doped compound, the observed excitation is shifted down to around 0.2 meV and substantially broadened.

We found that the specific-heat data for our single-crystalline sample with $x=0$ in the temperature range up to 8 K are in close agreement with those reported on polycrystalline samples in Refs. 20 and 23. In the temperature range above 10 K, our specific-heat data are considerably higher than those reported therein.

The maximum specific heat of our crystal with $x=0.15$, however, is obviously higher than that reported in Refs. 20 and 23, although the positions of the maximum of the specific-heat curves agree quite well. We obtained the same

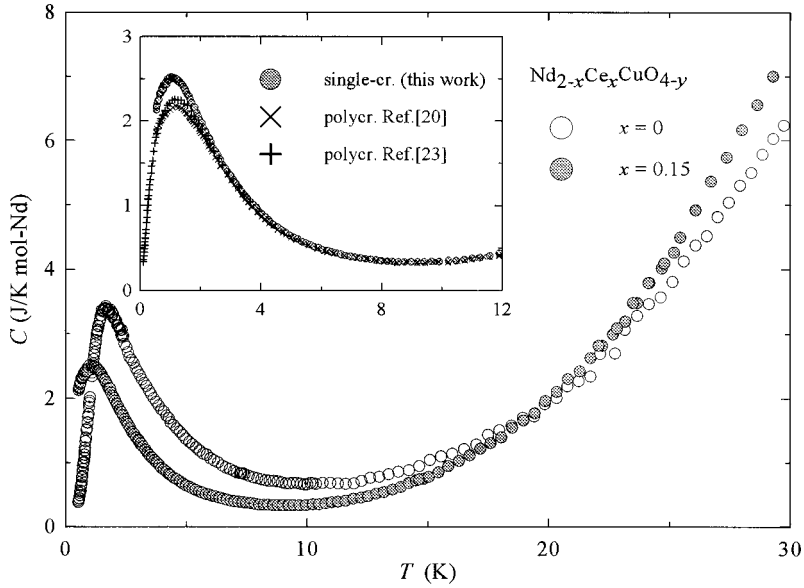

FIG. 2. Experimental specific heat of the $Nd_{2-x}Ce_xCuO_{4-y}$ single crystals (per mole Nd ions) in zero applied magnetic field in the temperature range between 0 and 30 K (y indicates the oxygen deficiency). The inset shows the difference between the specific-heat data for the $x=0.15$ compound, measured on our single-crystalline sample, and on polycrystalline samples of Ghamaty *et al.* (Ref. 20) and Brugger *et al.* (Ref. 23).

specific-heat curve on another single-crystalline sample of about 400 mg cut from another part of the same batch. For the temperature range from 2 up to 15 K, however, our data measured on both samples of the doped crystal agree well with those reported on polycrystalline samples in Refs. 20 and 23 (see inset of Fig. 2).

B. Specific heat in applied magnetic field

Figures 3(a), 3(b), 4(a), and 4(b) present the results of our low-temperature specific-heat measurements in magnetic fields for the two $Nd_{2-x}Ce_xCuO_{4-y}$ single crystals. The specific-heat data exhibit a highly anisotropic behavior with respect to the magnetic-field effect.

For the undoped sample, in case the field is applied parallel to the c axis [Fig. 3(a)], the $C(T)$ curves obtained at 0, 2, and 4 T show only minor differences: the curve becomes slightly broader with increasing field. With increasing field applied perpendicular to the c axis [Fig. 3(b)], however, the $C(T)$ curve shifts towards higher temperature and becomes substantially broadened. The maximum of the curve is gradually suppressed when the applied field increases from 2 to 6 T. It is raised again at 8 T to reach almost the zero-field

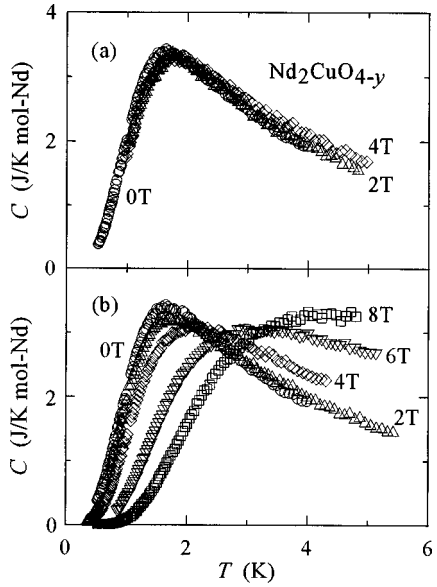

FIG. 3. Experimental specific heat of single-crystalline Nd_2CuO_{4-y} (per mole Nd ions) in magnetic fields applied parallel (a) and perpendicular (b) to the crystallographic c axis.

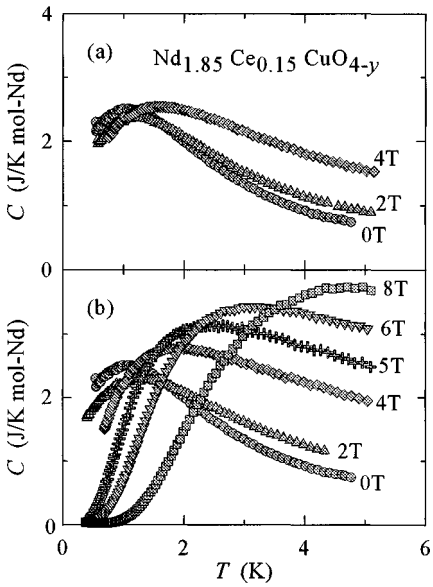

FIG. 4. Experimental specific heat of single-crystalline $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ (per mole Nd ions) in magnetic fields applied parallel (a) and perpendicular (b) to the crystallographic c axis.

level. The field effect on the specific heat observed here is unambiguously of a nonlinear type.

For the doped crystal, in case the field is applied parallel to the c axis [Fig. 4(a)], the effects of the applied magnetic field exhibit obviously another behavior than for the undoped crystal. At 2 T the maximum of the zero-field specific-heat curve is slightly suppressed, shifted to about 1.2 K and the curve is substantially broadened. At 4 T the maximum is raised again to the level observed at zero field, shifted to about 1.5 K and the curve is much broader. With the field direction perpendicular to the c axis [Fig. 4(b)], similar effects are observed, but the suppression and leveling up of the maximum as well as the broadening of the curve with the increasing field are more pronounced. At 4 T, the maximum specific heat is already increased by more than 10% compared to that measured at 0 T. These results show again an anisotropic and nonlinear behavior of the field effect on the specific heat.

For the doped compound we cannot exclude that the observed field effect in the data measured at 2 and 4 T along the c axis, is due to small misorientations in the sample. Neutron-diffraction experiments[29] have shown that the last 30 mm of the as-grown crystal boule with $x=0.15$ actually consists of three large grains developing along the growth [110] direction with their c axis tilted by some degrees with respect to each other. The single-crystalline sample used in the specific-heat experiments was cut from one of these grains, but the inclusion of small parts of the other grains cannot be excluded.

IV. MODEL CALCULATIONS

In the present paper we concentrate on the magnetic part C of the specific heat, calculated according to

$$C=-NT\partial^2F/\partial T^2, \tag{4.1}$$

where N is the number of Nd ions in the sample, and $F(T)=-k_BT\ln Z$ the single-ion free energy, with $Z(T)=\sum_{i=1}^{10}e^{-E_i(T)/k_BT}$ the partition function corresponding to a single Nd ion. The calculation along this line is justified by our model that is defined by the following characteristics (assumptions, approximations, etc.).

(1). We base our considerations on the noncollinear spin structure shown in Fig. 1. This structure has been proposed[3–6] for Nd_2CuO_{4-y} in the absence of an applied field. The 4 distinct directions of the Cu moments divide the Cu subsystem into 4 interpenetrating simple tetragonal Cu sublattices within each of which the Cu moments have the same orientation. The primitive unit cell of each sublattice contains one Cu ion and is rotated by 45° with respect to the chemical unit cell, having a factor of $\sqrt{2}$ larger basal axes. In a similar way, the Nd subsystem is divided into 4 sublattices, each with the same primitive unit cell as defined for the Cu sublattices but containing two Nd ions rather than one. We assume that this division of the Nd subsystem into 4 sublattices according to the orientation of the Nd moments remains the same for the two directions of the applied field for which our model is worked out below.

(2). Just like in the family of R_2CuO_4 compounds, the Cu moments in Nd_2CuO_4 order antiferromagnetically in the ab plane at a high Néel temperature[2–7] (in the order of 250 K). From this, we conclude that the exchange interaction between certain pairs of Cu ions is so strong that we can make the approximation that in all applied fields of interest in the present paper, the Cu moments keep their strict antiferromagnetic order in the CuO_2 planes. Moreover, we assume that the Cu moments remain parallel to these planes. This implies that the Cu moments stay strictly perpendicular to all applied fields along the c axis. As a further approximation we assume that the same holds for fields applied in the ab plane that are not too small. In this case a rotation is necessary, in half of the CuO_2 planes, of the moments over 90°, with the antiferromagnetic order preserved. (Neglecting a possible in-plane anisotropy of the Cu subsystem, such a rotation does not seem to be too difficult in view of the antiferromagnetic ordering of the Cu moments in the CuO_2 planes that results in a zero net interaction between neighboring planes.) This approximation implies that for the calculation of the (magnetic part of the) specific heat it suffices to consider the system of the Nd ions. Apart from the applied field and the crystalline electric field, each ion in this system is subjected to an additional constant (molecular) field originating from the exchange interaction with the system of the Cu moments.

Considering the ferromagnetic order in the Nd_2Cu chains along the c axis and the zero net interaction with one of the neighboring CuO_2 planes, the larger part of this molecular field is probably due to a (ferromagnetic) interaction of the Nd ion considered, with the neighboring Cu ion on the same chain.

(3). Apart from the 2-ion exchange of the Nd subsystem with the Cu subsystem (leading to the constant molecular field considered in characteristic 2), we also assume a 2-ion exchange within the Nd subsystem. We use a mean-field approximation to rewrite the Hamiltonian for the system of N exchange-coupled Nd ions, as a sum of N single-ion Hamiltonians of the form:

$$H=H_{\mathrm{CEF}}+g_{\mathrm{Nd}}\mu_B\boldsymbol{J}\cdot(\boldsymbol{B}_{\mathrm{appl}}+\boldsymbol{B}_{\mathrm{mol}}^{\text{Nd-Cu}}+\boldsymbol{B}_{\mathrm{mol}}^{\text{Nd-Nd}})+E_{\mathrm{corr}}. \tag{4.2}$$

This Hamiltonian operates in the 10-dimensional Hilbert space corresponding to the energetically lowest lying $J=\frac{9}{2}$ multiplet of the free Nd^{3+} ion. In Eq. (4.2) $g_{\mathrm{Nd}}=\frac{8}{11}$ is the Landé g factor of the Nd ion (so, the magnetic-moment operator of the Nd ion is given by $\boldsymbol{m}_{\mathrm{Nd}}=-g_{\mathrm{Nd}}\mu_B\boldsymbol{J}$), and

$$H_{\mathrm{CEF}}=B_2^0O_2^0+B_4^0O_4^0+B_6^0O_6^0+B_4^4O_4^4+B_6^4O_6^4 \tag{4.3}$$

is the Hamiltonian corresponding to the crystalline electric field, in terms of the Stevens operators O_n^m and the CEF parameters B_n^m. Furthermore, $\boldsymbol{B}_{\mathrm{mol}}^{\text{Nd-Cu}}$ and $\boldsymbol{B}_{\mathrm{mol}}^{\text{Nd-Nd}}$ in Eq. (4.2) are the molecular fields at the Nd ion considered, due to the Nd-Cu and Nd-Nd two-ion interactions, respectively. By characteristic 2 we take, in accordance with other authors,[5,22] the molecular field $\boldsymbol{B}_{\mathrm{mol}}^{\text{Nd-Cu}}$ as constant, in particular as independent of the temperature. The molecular field $\boldsymbol{B}_{\mathrm{mol}}^{\text{Nd-Nd}}$ will be considered under characteristic 4 below. Finally, $E_{\mathrm{corr}}=\frac{1}{2}\boldsymbol{B}_{\mathrm{mol}}^{\text{Nd-Nd}}\cdot\boldsymbol{m}_{\mathrm{Nd}}$ is a corrective additive ''constant'' that appears as a c number in the mean-field approximation at the reduction of the original Hamiltonian for the system of N exchange-coupled Nd ions, into a sum of N single-ion Hamiltonians.

(4). The magnetic moment of any Nd ion is given by $\boldsymbol{m}_{\mathrm{Nd}}=-g_{\mathrm{Nd}}\mu_B\langle \boldsymbol{J}\rangle$, where $\langle \boldsymbol{J}\rangle$ is the statistically averaged expectation value. In Eq. (4.2), the molecular field $\boldsymbol{B}_{\mathrm{mol}}^{\mathrm{Nd\text{-}Nd}}$ is a function of the different magnetic moments $\boldsymbol{m}_{\mathrm{Nd}}$ that occur in the Nd subsystem. By characteristic 1, the number of different Nd moments is restricted to 4 (one for each Nd sublattice). In this way we obtain a system of 4 coupled Hamiltonians of the form (4.2), each representing the typical Nd ion of one of the 4 Nd sublattices. In each of the 4 Hamiltonians, the vectors for $\boldsymbol{B}_{\mathrm{mol}}^{\mathrm{Nd\text{-}Cu}}$ and $\boldsymbol{B}_{\mathrm{mol}}^{\mathrm{Nd\text{-}Nd}}$ are different. For $\boldsymbol{B}_{\mathrm{mol}}^{\mathrm{Nd\text{-}Cu}}$, the 4 vectors occurring in the different Hamiltonians are related by simple symmetry transformations of the problem. This follows from characteristic 2 concerning the orientation of the ions in the Cu subsystem; for example, for $\boldsymbol{B}_{\mathrm{appl}}$ along the c axis, successive rotations over 90° about the c axis of $\boldsymbol{B}_{\mathrm{mol}}^{\mathrm{Nd\text{-}Cu}}$ as it occurs in one of the Hamiltonians, yield the values of $\boldsymbol{B}_{\mathrm{mol}}^{\mathrm{Nd\text{-}Cu}}$ in the other (three) Hamiltonians. There is no fundamental reason why similar relations should hold for the molecular fields $\boldsymbol{B}_{\mathrm{mol}}^{\mathrm{Nd\text{-}Nd}}$ that occur in the different Hamiltonians. The vector $\boldsymbol{B}_{\mathrm{mol}}^{\mathrm{Nd\text{-}Nd}}$ in each Hamiltonian is a function of the value of the magnetic moments $\boldsymbol{m}_{\mathrm{Nd}}$ in the Nd sublattice connected with the Hamiltonian, and of the $\boldsymbol{m}_{\mathrm{Nd}}$ vectors of the other three sublattices. It seems quite possible that there exist self-consistent diagonalizations of the system of the 4 coupled Hamiltonians in which the 4 $\boldsymbol{m}_{\mathrm{Nd}}$ vectors (and therefore the molecular fields $\boldsymbol{B}_{\mathrm{mol}}^{\mathrm{Nd\text{-}Nd}}$) are not related by simple symmetry transformations. The fourth characteristic of our model is now, that we shall restrict ourselves to those self-consistent diagonalizations in which the connection by symmetry between the $\boldsymbol{m}_{\mathrm{Nd}}$ occurring in the four Hamiltonians does exist. This restriction enables us to perform the self-consistent diagonalization of the system of 4 Hamiltonians by means of a self-consistent diagonalization of only a single Hamiltonian, the one connected with sublattice 1, say. If we consider this Hamiltonian, the molecular field $\boldsymbol{B}_{\mathrm{mol}}^{\mathrm{Nd\text{-}Nd}}$ can be written as a function of the moment $\boldsymbol{m}_{\mathrm{Nd}}$ of the typical Nd ion of the same sublattice. The self-consistent diagonalization is reached after an iterative process: the calculated magnetic moment $-g_{\mathrm{Nd}}\mu_B\langle \boldsymbol{J}\rangle$ has then obtained the same value as the input parameter $\boldsymbol{m}_{\mathrm{Nd}}$. How the restriction is incorporated in the model in detail, will be given presently (see the models with 4 and 2 Nd-moment directions below). We note that it is the restriction in characteristic 4 on the self-consistent diagonalizations considered, which justifies that the number N of Nd ions occurs as a coefficient in Eq. (4.1).

All our calculations have been based on the model defined by characteristics 1–4. According to characteristic 1, there are 4 distinct Nd sublattices, denoted 1, 2, 3, and 4 in Fig. 1. The model works out in two different ways, depending on the orientation of the applied field.

The model with 4 Nd-moment directions: $B_{\mathrm{appl}}\parallel c$ axis ($=z$ axis). In this case (this includes the case of zero applied field), we consider the four Nd sublattices mentioned in characteristic 1. Concentrating on sublattice 1 (Fig. 1), and taking $\boldsymbol{B}_{\mathrm{mol}}^{\mathrm{Nd\text{-}Cu}}$ at the position of the ions of this sublattice in the y direction, we see that neither $\boldsymbol{B}_{\mathrm{appl}}$ nor $\boldsymbol{B}_{\mathrm{mol}}^{\mathrm{Nd\text{-}Cu}}$ has an x component. In combination with the requirements on symmetry in characteristic 4, this implies that the Nd moments in sublattice 1 are given by $\boldsymbol{m}_{\mathrm{R}_1}=(0, m_{\mathrm{R}_1 y}, m_{\mathrm{R}_1 z})$. The Nd moments in the other Nd sublattices are then given by $\boldsymbol{m}_{\mathrm{R}_2}=(-m_{\mathrm{R}_1 y},0,m_{\mathrm{R}_1 z})$, $\boldsymbol{m}_{\mathrm{R}_3}=(0,-m_{\mathrm{R}_1 y},m_{\mathrm{R}_1 z})$, and $\boldsymbol{m}_{\mathrm{R}_4}=(m_{\mathrm{R}_1 y},0,m_{\mathrm{R}_1 z})$. Denoting the exchange parameters between the sublattices 1 and i by $n_{\mathrm{RR}}^{(i)}$ (e.g., $n_{\mathrm{RR}}^{(1)}$ describes the interaction within one Nd sublattice), the molecular field $\boldsymbol{B}_{\mathrm{mol}}^{\mathrm{Nd\text{-}Nd}}$ is now calculated by simple addition:

$$\boldsymbol{B}_{\mathrm{mol}}^{\mathrm{Nd\text{-}Nd}}=\{0,[n_{\mathrm{RR}}^{(1)}-n_{\mathrm{RR}}^{(3)}]m_{\mathrm{R}_1 y},[n_{\mathrm{RR}}^{(1)}+2n_{\mathrm{RR}}^{(2)}+n_{\mathrm{RR}}^{(3)}]m_{\mathrm{R}_1 z}\}, \tag{4.4a}$$

from which follows for E_{corr} in Eq. (4.2):

$$E_{\mathrm{corr}}=\tfrac{1}{2}\boldsymbol{B}_{\mathrm{mol}}^{\mathrm{Nd\text{-}Nd}}\cdot\boldsymbol{m}_{\mathrm{R}_1}=\tfrac{1}{2}[n_{\mathrm{RR}}^{(1)}-n_{\mathrm{RR}}^{(3)}]m_{\mathrm{R}_1 y}^2 + \tfrac{1}{2}[n_{\mathrm{RR}}^{(1)}+2n_{\mathrm{RR}}^{(2)}+n_{\mathrm{RR}}^{(3)}]m_{\mathrm{R}_1 z}^2. \tag{4.5a}$$

In deriving Eq. (4.4a) we considered that $n_{\mathrm{RR}}^{(2)}$ and $n_{\mathrm{RR}}^{(4)}$ are equal on the basis of the crystal structure (Fig. 1).

The model with 2 Nd-moment directions: $\boldsymbol{B}_{\mathrm{appl}}\perp c$ axis. In case $\boldsymbol{B}_{\mathrm{appl}}$ is perpendicular to the c axis and large enough (this excludes the case of zero applied field), we take $\boldsymbol{B}_{\mathrm{mol}}^{\mathrm{Nd\text{-}Cu}}$ perpendicular to the applied field (according to characteristic 2) and, as in the previous case, perpendicular to the c axis ($=z$ axis). Ignoring for the moment the in-plane anisotropy of the Nd subsystem (we will give a brief remark on this in Sec. VI), we take for simplicity $\boldsymbol{B}_{\mathrm{appl}}$ in the x direction, hence the direction of $\boldsymbol{B}_{\mathrm{mol}}^{\mathrm{Nd\text{-}Cu}}$ is parallel to the y direction. This implies, in combination with the symmetry requirements in characteristic 4, that there are only two Cu-moment directions and two Nd-moment directions: for the sake of definiteness we let the moments in the Nd sublattices 1 and 2 have the same direction, and the same holds for the moments in sublattices 3 and 4. In accordance with this model, we assume for the calculation of $\boldsymbol{B}_{\mathrm{mol}}^{\mathrm{Nd\text{-}Nd}}$ that the Nd moments in the 4 Nd sublattices orient themselves according to the scheme: $\boldsymbol{m}_{\mathrm{R}_1}=\boldsymbol{m}_{\mathrm{R}_2}=(m_{\mathrm{R}_1 x},m_{\mathrm{R}_1 y},0)$ and $\boldsymbol{m}_{\mathrm{R}_3}=\boldsymbol{m}_{\mathrm{R}_4}=(m_{\mathrm{R}_1 x},-m_{\mathrm{R}_1 y},0)$. Using the same exchange parameters as in the previous model, the molecular field $\boldsymbol{B}_{\mathrm{mol}}^{\mathrm{Nd\text{-}Nd}}$ is calculated here also by a simple addition:

$$\boldsymbol{B}_{\mathrm{mol}}^{\mathrm{Nd\text{-}Nd}}=\{[n_{\mathrm{RR}}^{(1)}+2n_{\mathrm{RR}}^{(2)}+n_{\mathrm{RR}}^{(3)}]m_{\mathrm{R}_1 x},[n_{\mathrm{RR}}^{(1)}-n_{\mathrm{RR}}^{(3)}]m_{\mathrm{R}_1 y},0\}, \tag{4.4b}$$

from which follows for E_{corr} in Eq. (4.2):

$$E_{\mathrm{corr}}=\tfrac{1}{2}\boldsymbol{B}_{\mathrm{mol}}^{\mathrm{Nd\text{-}Nd}}\cdot\boldsymbol{m}_{\mathrm{R}_1}=\tfrac{1}{2}[n_{\mathrm{RR}}^{(1)}-n_{\mathrm{RR}}^{(3)}]m_{\mathrm{R}_1 y}^2 + \tfrac{1}{2}[n_{\mathrm{RR}}^{(1)}+2n_{\mathrm{RR}}^{(2)}+n_{\mathrm{RR}}^{(3)}]m_{\mathrm{R}_1 x}^2. \tag{4.5b}$$

The magnetic specific heat C is calculated according to Eq. (4.1). At each temperature, the 10 energy levels E_i of a single Nd ion, and hence the free energy, are easily calculated by a self-consistent diagonalization of the Hamiltonian (4.2) above. We did take into account in our calculations all 10 energy levels corresponding to $J=\frac{9}{2}$. However, we consider that the splitting (in the order of 5 K) of the two levels originating from the Kramers doublet with the lowest energy is small compared to the distance (in the order of 175 K) of the two lowest doublets of a Nd ion (see Sec. VI). Therefore, at the low temperatures considered in the present paper, only

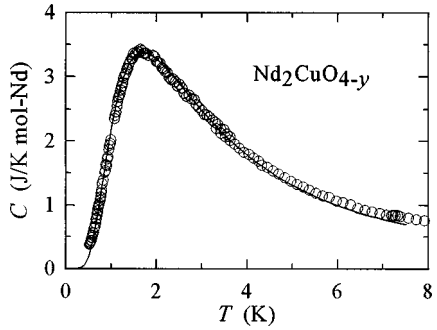

FIG. 5. Experimental (open circles) and calculated (solid line) specific heat of single-crystalline Nd_2CuO_{4-y} in zero applied magnetic field.

the positions of the two levels in the lowest doublet influence the results of our specific-heat calculations.

It should be noted that neither the model with 4 Nd-moment directions as described above, nor the model with 2 Nd-moment directions can handle applied fields which are in other directions than parallel or perpendicular to the c axis. This is because for such other field directions the symmetry considerations we used in the development of the two models are not applicable.

As can be seen from Eqs. (4.4a), (4.5a), (4.4b), and (4.5b), there are only 2 linear combinations of the exchange parameters $n_{\mathrm{RR}}^{(i)}$ that account for the Nd-Nd exchange in our specific-heat calculations, viz.

$$n_{\mathrm{RR}}^{\mathrm{dif}} := n_{\mathrm{RR}}^{(1)} - n_{\mathrm{RR}}^{(3)}, \tag{4.6a}$$

$$n_{\mathrm{RR}}^{\mathrm{sum}} := n_{\mathrm{RR}}^{(1)} + 2n_{\mathrm{RR}}^{(2)} + n_{\mathrm{RR}}^{(3)}. \tag{4.6b}$$

V. FITTING RESULTS

A. The undoped crystal ($x=0$)

We performed the following fitting procedure to obtain a combination of parameters that gives an adequate description of the experimental data. Considering the wide spread in the CEF-parameter sets proposed so far,[9–13,17] the first step was to choose an adequate set of CEF parameters. For this choice we required that the set should reproduce reasonably well the anisotropic behavior of the field effect as observed in our specific-heat measurements, i.e., (almost) no shift of the (maximum of the) specific-heat curve for fields $\boldsymbol{B}_{\mathrm{appl}}$ applied along the c axis, and a substantial shift for fields applied in the ab plane. We carried out the specific-heat calculations using all CEF-parameter sets published so far,[9–13,17] while ignoring the Nd-Nd exchange interaction (so $\boldsymbol{B}_{\mathrm{mol}}^{\mathrm{Nd\text{-}Nd}}=0$, and no iterations). Using a nonzero value for $\boldsymbol{B}_{\mathrm{mol}}^{\mathrm{Nd\text{-}Cu}}$ ($B_{\mathrm{mol}}^{\mathrm{Nd\text{-}Cu}}=2.1$ T and oriented in the ab plane; otherwise (almost) no splitting of the ground-state doublet was achieved for $\boldsymbol{B}_{\mathrm{appl}} \parallel c$ axis), we found[26] that only the sets published by Boothroyd *et al.*,[10b] Muzichka *et al.*,[12] and Jandl *et al.*,[17] reproduce the anisotropic behavior mentioned. Considering the similarity of these three CEF-parameter sets, we made an arbitrary choice and used the set of Jandl *et al.* in our further analysis below: $B_2^0=1.513$ K, $B_4^0=0.1185$ K, $B_6^0=-0.7345\times10^{-3}$ K, $B_4^4=-0.722$ K, and $B_6^4=-56.6\times10^{-3}$ K. (Note that the CEF parameters of Ref. 17 are given here in Stevens-operator notation; see Ref. 30 for the conversion relations.)

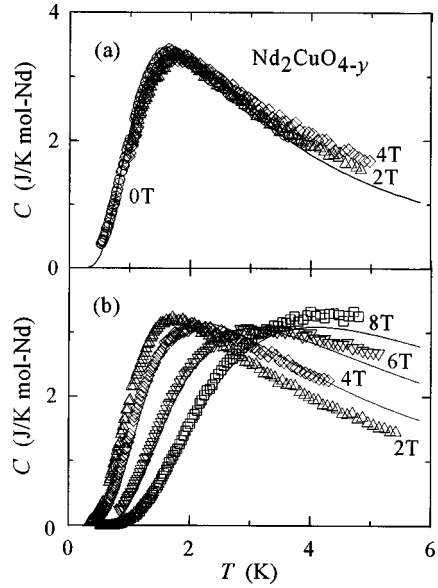

FIG. 6. Experimental (isolated points) and calculated (solid lines) specific heat of single-crystalline Nd_2CuO_{4-y} in different magnetic fields applied parallel (a) and perpendicular (b) to the crystallographic c axis. In Fig. 6(a) the calculated curves for applied fields of 0, 2, and 4 T coincide.

It is self-evident (in view of the splitting of the ground-state doublet compared to the distance of the next doublet) that in our calculations the maximum in the specific-heat curve is constant for all applied-field strengths as long as we ignore the Nd-Nd interaction. This maximum is about 7.5% higher than the one measured at zero applied field. Furthermore, in these calculations the shift of the position of the maximum under the influence of an applied magnetic field in the ab plane, is much larger than the one experimentally found (for the different fields applied along the c axis the calculated curves essentially coincide).

As mentioned in Sec. IV, in our approach there are only 2 parameters that account for the Nd-Nd exchange, viz., $n_{\mathrm{RR}}^{\mathrm{dif}}$ and $n_{\mathrm{RR}}^{\mathrm{sum}}$, given by Eqs. (4.6a) and (4.6b), respectively. For zero $\boldsymbol{B}_{\mathrm{appl}}$, the model with 4 Nd-moment directions applies and, since in this case $m_{\mathrm{R}_1 z}=0$, we derive from Eqs. (4.2), (4.4a), and (4.5a) that, apart from the CEF parameters chosen already above, the only parameters to be adjusted are (the strength of) $\boldsymbol{B}_{\mathrm{mol}}^{\mathrm{Nd\text{-}Cu}}$ (that we take in the y direction) and $n_{\mathrm{RR}}^{\mathrm{dif}}$. The solid line in Fig. 5 shows the best fit. This fit has been calculated using $B_{\mathrm{mol}}^{\mathrm{Nd\text{-}Cu}}=2.70\pm0.05$ T and $n_{\mathrm{RR}}^{\mathrm{dif}}=-0.37\pm0.02$ (Nd-ion T)/μ_B.

Using the same values for these parameters in the calculation of the specific heat for nonzero $\boldsymbol{B}_{\mathrm{appl}} \perp c$ axis, we only have to adjust the remaining parameter $n_{\mathrm{RR}}^{\mathrm{sum}}$ to fit the experimental curves. In this case, as explained in Sec. IV, the model with 2 Nd-moment directions applies. Figure 6(b) shows the best fit found with $n_{\mathrm{RR}}^{\mathrm{sum}}=-2.75\pm0.05$ (Nd-ion T)/μ_B for the experimental data measured at $B_{\mathrm{appl}}=4$, 6, and 8 T. We note that using the same values for $B_{\mathrm{mol}}^{\mathrm{Cu-Nd}}$ (2.70 T), $n_{\mathrm{RR}}^{\mathrm{dif}}$ [-0.37 (Nd-ion T)/μ_B], and $n_{\mathrm{RR}}^{\mathrm{sum}}$ [-2.75 (Nd-ion

T)/μ_B], in the calculations just described for $\boldsymbol{B}_{\rm appl} \parallel x$ axis, the iteration in diagonalizing self-consistently the Hamiltonian (4.2) did not converge with $B_{\rm appl}=2$ T. The iteration did not converge either in the temperature range below 1.5 K for $B_{\rm appl}=4$ T when using $n_{\rm RR}^{\rm sum}=-2.80$ (Nd-ion T)/μ_B, i.e., only a slight change in $n_{\rm RR}^{\rm sum}$. Apparently, a stable solution with 2 Nd-moment directions does not exist for fields of such small strength applied in the x direction. Finally, we note that the calculations with the same parameters as mentioned above for fields of 2 and 4 T applied along the c axis, have resulted in curves that essentially coincide with the one obtained for $B_{\rm appl}=0$ T [Fig. 6(a)].

B. The doped crystal ($x=0.15$)

In the undoped compound we have essentially only one type of Nd ions: each single-ion Hamiltonian has the same form and the same values of the parameters, apart from the molecular-field vector parameters that are related by simple symmetry transformations if the Nd ions belong to different sublattices (cf. Sec. IV, characteristic 4). In contrast to this, the Nd ions in $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ are expected to have various environments with, for example, a different number and different positions of Ce neighbors. This implies that there are many inequivalent types of Nd sites in the doped compound. In any fitting procedure, all these different types of Nd ions have to be incorporated with the correct weight and the typical CEF-parameter set. Moreover, one has to enter into the fundamental question whether the sets of CEF-parameter values published in the literature[9,10,18,31] concerning $Nd_{1.85}Ce_{0.15}CuO_{4-y}$, are able to describe the crystalline electric field at all, considering that the sets of adjustable parameters are not consistent with the point symmetry of most Nd ions in $Nd_{1.85}Ce_{0.15}CuO_{4-y}$. After brushing aside this objection, here it is much less obvious which choice is to be made out of the CEF-parameter sets proposed, than in case of the undoped compound. This is also because the observed anisotropy of the magnetic-field effect on the specific heat in the doped compound shows quite another behavior than in the undoped compound.

As regards the experimental findings by now, we mention that on another single-crystalline sample cut from the same batch as ours, Loewenhaupt *et al.*[27] have observed in INS experiments excitations of the Nd ground-state doublet that suggest a distribution of the molecular field on the Nd sites. Furthermore, we recall Jandl *et al.*,[18] who proposed the existence of three inequivalent Nd sites to explain the spectra observed in their Raman-scattering experiments on single-crystalline $Nd_{1.85}Ce_{0.15}CuO_{4-y}$.

As mentioned in Sec. II, our specific-heat measurements have been performed on an as-grown sample that shows a broad superconducting transition at about 16 K. With respect to the magnetic configuration of the Nd ions, we referred to Ref. 7 already in Sec. I to mention that the long-range order of the Cu moments is destroyed in the superconducting compound with $x=0.15$. However, we always expect short-range order from which it seems reasonable to assume that the magnetic configuration looks like the one in the undoped compound. On the other hand, we refer to the neutron-diffraction experiments of Matsuda *et al.*,[32] where long-range antiferromagnetic order of the Cu moments has been observed below $T_N=160$ K in (nonsuperconducting) as-grown single crystals of $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ that were prepared by the TSFZ-method, in a similar way as ours. The authors reported that, even after an appropriate oxygen-reduction annealing procedure, inclusions of 10% to 30% of a coexisting antiferromagnetic phase were found to persist in the superconducting samples.

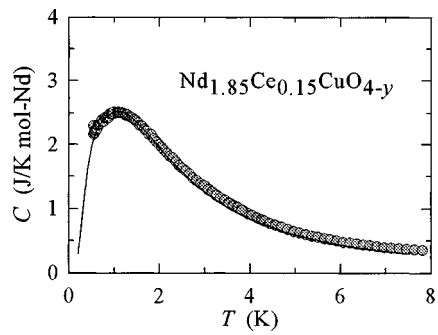

FIG. 7. Experimental (hatched circles) and calculated (solid line) specific heat of single-crystalline $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ in zero applied magnetic field.

Above we pointed out that any fitting procedure should treat all inequivalent types of Nd ions that occur in $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ with the appropriate weights, treating among other quantities the CEF parameters of each Nd-ion type as adjustable. Consequently, an enormous number of fitting parameters has to be taken into account. Under these circumstances, the values of the fitting parameters used for reproducing the experimental data cannot be claimed to have any reasonable physical meaning. As can be expected from these considerations, we failed to fit the zero-field data as long as we based the calculations on the same model—with only one single type of Nd ions—as applied for the undoped compound, using any of the CEF-parameter sets proposed for the doped compound in the literature.[9,10,18,31]

We have also attempted to describe the experimental data by a phenomenological treatment based on a Gaussian distribution of the molecular field without taking into account the Nd-Nd interaction. By adjusting the parameters for the Gaussian molecular-field distribution, we could simultaneously reproduce—albeit not very satisfactorily—the data measured in zero field as well as at 8 T applied in the ab plane, but failed to fit the data obtained at other fields.

In order to give an idea of the consequences of ignoring the objections raised above against such an approach in the case of the doped compound, we present some results obtained using parameter values published in the literature. The fitting procedure applied was based on the three inequivalent types of Nd ions, I, II, and III, with the corresponding weights $w=0.37$, 0.46, and 0.17, respectively, and the corresponding CEF-parameter sets proposed by Jandl *et al.*[18] Proceeding along the same lines as for the undoped compounds, we derived from the zero-field specific-heat data values of the fitting parameters $B_{\rm mol}^{\rm Nd\text{-}Cu}$ and $n_{\rm RR}^{\rm dif}$ for each of the three inequivalent types of Nd ions. Figure 7 shows the best fit we found. Using the same values for these parameters, we adjusted for each type of Nd ions the corresponding value of the fitting parameter $n_{\rm RR}^{\rm sum}$ in order to fit the experimental data obtained at the different nonzero fields applied perpendicular

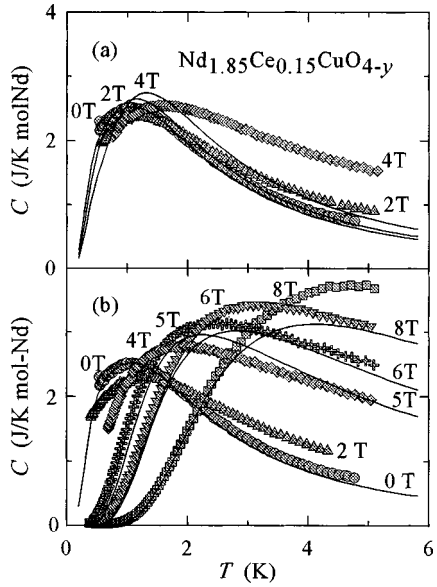

FIG. 8. Experimental (isolated points) and calculated (solid lines) specific heat of single-crystalline $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ in different magnetic fields applied parallel (a) and perpendicular (b) to the crystallographic c axis.

to the c axis. The best fitting results of the calculations are shown in Figs. 8(a) and 8(b) for fields applied parallel and perpendicular to the c axis, respectively. These results have been obtained with the parameters given in Table I. The calculated curves for 2 and 4 T in Fig. 8(b) are missing because the computer program failed to function for these fields (very slow convergence or no convergence at all on iteration).

VI. DISCUSSION

As suggested by the noncollinear spin structure described in Fig. 1, for zero applied field the molecular field, $\boldsymbol{B}_{\mathrm{mol}}^{\mathrm{Nd\text{-}Cu}}$, is pointing in a $\langle 100\rangle$ direction. In agreement with this suggestion, we took in our calculations the direction of $\boldsymbol{B}_{\mathrm{mol}}^{\mathrm{Nd\text{-}Cu}}$ parallel to the y direction for any $\boldsymbol{B}_{\mathrm{appl}} \parallel c$ axis (including $B_{\mathrm{appl}}=0$). In the actual experiment we applied the field, if oriented in the ab plane, along a $\langle 110\rangle$ direction. In contrast to this, the computer program was written for $\boldsymbol{B}_{\mathrm{appl}}$ in the x direction in the model with 2 Nd-moment directions, i.e., we performed all calculations where $\boldsymbol{B}_{\mathrm{appl}} \perp c$ axis, with $\boldsymbol{B}_{\mathrm{appl}}$ in a $\langle 100\rangle$ direction. The results of our calculations would presumably not have been different if in our calculations $\boldsymbol{B}_{\mathrm{appl}}$ would have been along a $\langle 110\rangle$ direction (and $\boldsymbol{B}_{\mathrm{mol}}^{\mathrm{Nd\text{-}Cu}}$ perpendicular to it in the ab plane). Indeed, when we ignore the Nd-Nd interaction, the calculations yield essentially the same results for any orientation of the applied field in the ab plane with the fixed configuration $\boldsymbol{B}_{\mathrm{appl}} \perp \boldsymbol{B}_{\mathrm{mol}}^{\mathrm{Nd\text{-}Cu}}$.[26]

Following other authors, see, e.g., Refs. 22 and 23, we interpret our experimental specific-heat curves as Schottky curves connected with the two energy levels of the ground-state doublet of the Nd ions. Accordingly, the experimental results on both crystals indicate that the magnetic field, if applied in the ab plane, has the tendency to enhance the splitting of the ground-state doublet. This results in scaling up the specific-heat curve towards higher temperature. As can be noticed from our calculations, the Nd-Nd interaction counteracts this effect of the applied field. Furthermore, the specific-heat maximum is suppressed and the curve is broadened by the Nd-Nd interaction. Below we shall explain why the Nd-Nd interaction has these effects.

A. The undoped crystal ($x=0$)

Expressing our experimental results on the Nd_2CuO_{4-y} crystal in terms of the splitting ΔE of the ground-state doublet, the magnetic field has an anisotropic effect on this splitting: the magnetic field, if oriented along the c axis, has only a subtle effect on ΔE and a substantial one if oriented in the ab plane. This is confirmed by our calculations in Sec. V A. Using the CEF-parameter set of Jandl *et al.*,[17] we found: $\Delta E/B_{\mathrm{eff}}^{\mathrm{Nd}}=0.026$ K/T and 1.841 K/T for the effective magnetic field $\boldsymbol{B}_{\mathrm{eff}}^{\mathrm{Nd}}$ at the Nd site parallel and perpendicular to the c axis, respectively. (These two values turn out to be about constants at all strengths of $\boldsymbol{B}_{\mathrm{eff}}^{\mathrm{Nd}}$ in the range of interest in the present paper.) We note, however, that without any effective field our calculation with this CEF-parameter set gives an energy-level scheme with the five doublets: 0, 185, 352, 405, and 1121 K, for the Nd^{3+} ion in the $^4I_{9/2}$ lowest multiplet. This deviates considerably from the one found in INS measurements:[33] 0, 174, 242, 313, and 1102 K. One should realize that the parameter set of Jandl *et al.* was derived to be used in many-multiplet calculations, whereas we used the set in a lowest-multiplet calculation. Our approach is justified by considering that for low temperatures only the lowest doublet (of the lowest multiplet) is relevant in our specific-heat calculations.

Our fitting procedure in Sec. V A yielded $B_{\mathrm{mol}}^{\mathrm{Nd\text{-}Cu}}=2.70$ T (corresponding to $\Delta E=5.0$ K), which value can be used to

TABLE I. Fitting parameters (CEF, exchange and weight) for the three types of Nd ions in $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ (sites I, II, and III, Ref. 18; see Sec. V B for details) used in the calculations corresponding to Fig. 8. (The CEF parameters of Ref. 18 are presented here in Stevens-operator notation; see Ref. 30 for the conversion calculations.) For comparison the fitting parameters used in and resulting from this work for Nd_2CuO_{4-y} (see Sec. V A) are also shown.

	B_2^0 (K)	B_4^0 (mK)	B_6^0 (mK)	B_4^4 (K)	B_6^4 (mK)	$B_{\mathrm{mol}}^{\mathrm{Nd\text{-}Cu}}$ (T)	$n_{\mathrm{RR}}^{\mathrm{dif}}$ [(Nd-ion T)/μ_B]	$n_{\mathrm{RR}}^{\mathrm{sum}}$ [(Nd-ion T)/μ_B]	w
Site I	−2.57	125.13	−1.44	−0.733	−53.2	0.80	−0.05	−2.00	0.37
Site II	0.81	124.08	−0.86	−0.728	−56.4	2.35	−0.24	−2.70	0.46
Site III	1.94	130.07	−0.45	−0.827	0.00	0.75	−0.05	−1.60	0.17
Nd_2CuO_{4-y}	1.51	118.50	−0.73	−0.722	−56.6	2.70	−0.37	−2.75	1.00

describe all our specific-heat curves. In our calculations we assumed $\boldsymbol{B}_{\mathrm{mol}}^{\mathrm{Nd\text{-}Cu}}$ to be independent of temperature. Since the effective field $\boldsymbol{B}_{\mathrm{eff}}^{\mathrm{Nd}}$, at any Nd position is the vector sum of $\boldsymbol{B}_{\mathrm{appl}}$, $\boldsymbol{B}_{\mathrm{mol}}^{\mathrm{Nd\text{-}Cu}}$ and $\boldsymbol{B}_{\mathrm{mol}}^{\mathrm{Nd\text{-}Nd}}$, the splitting of the ground-state doublet would also be independent of the temperature if there was no Nd-Nd interaction. A simple Schottky curve with the standard, field-independent value of 3.652 J/(K mol-Nd) for the maximum would be the result. Now we consider what effects the Nd-Nd interaction has. In our calculations we found that the Nd moment decreases on increasing the temperature. Because of this and in view of the signs of the parameters $n_{\mathrm{RR}}^{\mathrm{dif}}$ and $n_{\mathrm{RR}}^{\mathrm{sum}}$, which have both been found to be negative, we derive from Eq. (4.4b) that the strength of the effective field at the Nd ion, and hence the splitting ΔE (we remind that $\Delta E/B_{\mathrm{eff}}^{\mathrm{Nd}}$ is about a constant), increases with increasing temperature. This results in a lowering of the maximum and broadening of the specific-heat curve. To put it quantitatively: our calculations at 0.2 K and in zero applied field yield the value $1.39\mu_B$ for the Nd moment, which corresponds to a value of 0.51 T for $B_{\mathrm{mol}}^{\mathrm{Nd\text{-}Nd}}$ and a decrease of $B_{\mathrm{eff}}^{\mathrm{Nd}}$ by this value. The corresponding values at 1.5, 4.2, and 7.5 K are $1.23\mu_B$ (corresponding to 0.46 T), $0.69\mu_B$ (corresponding to 0.26 T), and $0.44\mu_B$ (corresponding to 0.16 T), respectively. In our calculations, the splitting of the ground-state doublet at the four temperatures mentioned are 4.03, 4.14, 4.50, and 4.68 K, respectively. The value 4.14 K at 1.5 K for ΔE is in good agreement with the value 0.35 meV ($=4.06$ K), derived from INS experiments.[9] The splitting $\Delta E=4.03$ at 0.2 K, however, deviates somewhat from $\Delta E=0.39$ meV ($=4.5$ K) at 0.5 K, observed in an INS experiment by Loewenhaupt *et al.*[27] on a single-crystalline sample of the same batch as ours. The value 1.39 μ_B at 0.2 K is in good agreement with the value (1.3 ± 0.1) μ_B at 0.4 K, reported in neutron-diffraction experiments.[2] The values for the moments at higher temperatures, however, are significantly higher than those found experimentally by other authors: 0.44 μ_B at 4.1 K (Ref. 6) and 0.28 μ_B at 8 K.[2,34] This deviation is probably connected with the incorrect energy-level scheme in our calculations mentioned above.

The calculated results of the specific heat of the Nd_2CuO_{4-y} single crystal in different applied fields are shown in Figs. 6(a) and 6(b). First concentrating on Fig. 6(a), $\boldsymbol{B}_{\mathrm{appl}} \parallel c$ axis, the difference between the calculated curves and the experimental ones in the temperature range above 4 K can be attributed to lattice and electronic contributions. According to Brugger *et al.*[23] and Chattopadhyay and Siemensmeyer,[35] the difference between calculated and experimental curves in the temperature range below about 0.5 K can be accounted for by the nuclear specific heat of the ^{143}Nd and ^{145}Nd isotopes.

Next we turn our attention to Fig. 6(b), $\boldsymbol{B}_{\mathrm{appl}}$ in the ab plane. Also in this case, we attribute the difference between the calculated curves and the experimental ones in the temperature range above 4 K to lattice and electronic contributions. As noted in the previous section, a calculated 2 T curve is missing in this figure, because the computer program failed to produce a self-consistent solution for this field.

The computer program searches for such solutions that allow only two possibilities for the Nd moment, the two allowed vectors being equal and having symmetrical orientations with respect to the applied field. Apparently, at such low fields, stable configurations with only 2 values of the Nd moments are not compatible with the crystalline electric field and exchange interactions involved. However, using the same computer program, we found solutions for fields above 4 T. Furthermore, we also found for zero field (using the computer program handling any value of the applied field along the c axis) a solution with 4 Nd-moment directions. These considerations lead to the conclusion that our calculations in some sense reproduce what was found experimentally by Cherny *et al.*,[5] namely, a first-order phase transition at about 4.4 T in magnetization measurements at a temperature of 0.7 K, in fields applied along the a axis. At this field the low-field configuration with 4 Nd-moment directions abruptly changes into the high-field configuration with 2 Nd directions.

An approach that can be considered as closely connected to ours, in the sense that the fitting parameters can be translated into the parameters used in the present paper, has been given by Adelmann *et al.*[22] In order to explain theoretically their zero-field experimental specific-heat data of polycrystalline Nd_2CuO_{4-y}, they have treated each Nd ion as a two-level system, rather than the ten-level system of our analysis. The very low temperatures involved justify the approach of Adelmann *et al.* that leads—for zero applied field only, see below—to a very elegant description by isolating just the essentials of the assumptions we made in the underlying paper. These authors proposed a splitting of the Nd^{3+} ground-state doublet according to

$$\Delta E=\Delta_0+2B^{\mathrm{Nd}}m^{\mathrm{Nd}}=\Delta_0+2B^{\mathrm{Nd}}m_0^{\mathrm{Nd}}\tanh\left(\frac{1}{2}\frac{\Delta E}{k_BT}\right). \tag{6.1}$$

(In our opinion the coefficient $\frac{1}{2}$ in the argument of tanh( ) is erroneously missing in Ref. 22.) Equation (6.1) consists of a temperature-independent contribution Δ_0 due to the Nd-Cu interaction and a temperature-dependent contribution proportional to m^{Nd}, the moment of the Nd ion. If we consider the system of a single Nd ion as a doublet, the latter contribution is proportional to $\tanh(\Delta E/2k_BT)$ with a coefficient $2B^{\mathrm{Nd}}m_0^{\mathrm{Nd}}$ due to the Nd-Nd interaction. For each fixed combination of parameters Δ_0 and $2B^{\mathrm{Nd}}m_0^{\mathrm{Nd}}$, Eq. (6.1) yields a self-consistent solution for ΔE as a function of the temperature T, which enables the determination of the corresponding specific heat. Adelmann *et al.* found that the values $\Delta_0=4.5$ K and $2B^{\mathrm{Nd}}m_0^{\mathrm{Nd}}=-0.8$ K give the best agreement with their measured zero-field specific-heat data. It is easy to derive the analogue of Eq. (6.1) in terms of the analysis described in Sec. IV. For the (almost temperature-independent) expectation values of the Nd moment in the two states of the ground-state doublet, our calculations yielded $m_{\mathrm{low}}^{\mathrm{Nd}}=1.39\mu_B$ and $m_{\mathrm{high}}^{\mathrm{Nd}}=-1.35\mu_B$. The splitting of the ground-state doublet being given by $\Delta E=(m_{\mathrm{low}}^{\mathrm{Nd}}-m_{\mathrm{high}}^{\mathrm{Nd}})B_{\mathrm{eff}}^{\mathrm{Nd}}$, we derive the value $\Delta E/k_BB_{\mathrm{eff}}^{\mathrm{Nd}}=(m_{\mathrm{low}}^{\mathrm{Nd}}-m_{\mathrm{high}}^{\mathrm{Nd}})/k_B=1.84$ K/T found before. In our analysis, a formula similar to Eq. (6.1) is now derived as follows:

$$\Delta E = \frac{\Delta E}{B_{\rm eff}^{\rm Nd}}\,(B_{\rm mol}^{\rm Nd\text{-}Cu} + B_{\rm mol}^{\rm Nd\text{-}Nd}) = \frac{\Delta E}{B_{\rm eff}^{\rm Nd}}\,(B_{\rm mol}^{\rm Nd\text{-}Cu} + n_{\rm RR}^{\rm dif} m^{\rm Nd})$$
$$= \frac{\Delta E}{B_{\rm eff}^{\rm Nd}}\left\{ B_{\rm mol}^{\rm Nd\text{-}Cu} + n_{\rm RR}^{\rm dif}\left[\frac{1}{2}\,(m_{\rm low}^{\rm Nd} + m_{\rm high}^{\rm Nd}) + \frac{1}{2}\,(m_{\rm low}^{\rm Nd} - m_{\rm high}^{\rm Nd})\tanh\left(\frac{1}{2}\,\frac{\Delta E}{k_B T}\right)\right]\right\}$$
$$= \frac{\Delta E}{B_{\rm eff}^{\rm Nd}}\left[B_{\rm mol}^{\rm Nd\text{-}Cu} + \frac{1}{2}\,n_{\rm RR}^{\rm dif}(m_{\rm low}^{\rm Nd} + m_{\rm high}^{\rm Nd})\right] + \frac{1}{2}\,\frac{\Delta E}{B_{\rm eff}^{\rm Nd}}\,n_{\rm RR}^{\rm dif}(m_{\rm low}^{\rm Nd} - m_{\rm high}^{\rm Nd})\tanh\left(\frac{1}{2}\,\frac{\Delta E}{k_B T}\right). \qquad (6.2)$$

Like in Eq. (6.1) we see two terms. Unlike in the treatment of Adelmann *et al.*, in our analysis the first term in the right-hand side of Eq. (6.2) contains a small, weakly temperature-dependent contribution originating from the Nd-Nd interaction. Using the values given before, this contribution turns out to be negligibly small. For the first term we obtain a value of 5.0 K, which is to be compared with the value 4.5 K Adelmann *et al.* found for Δ_0. For the coefficient of tanh( ), we find -0.93 K, to be compared with the value -0.8 K Adelmann *et al.* found for $2B^{\rm Nd}m_0^{\rm Nd}$. Adelmann *et al.* applied the same approach to fit the specific-heat data obtained in an applied field of 4.4 T. Although the authors claim to reproduce reasonably well the experimental data, they found values for the parameters that differ from those found at fitting the zero-field data. Moreover, they found a value of only 0.85 μ_B for the Nd moment, which is significantly smaller than $m_{\rm low}^{\rm Nd} = 1.39\ \mu_B$, found in our calculations, and than 1.3 μ_B, reported in neutron-diffraction studies.[2,34] In our view this result of Adelmann *et al.* for nonzero applied field is not satisfactory. We explain this by considering that (i) Adelmann *et al.* did not take into account the anisotropy of the effect of an effective field on the splitting of the ground-state doublet, and (ii) in contrast to other experiments[4,5] and to our approach, the authors did not base their calculation on the fundamental assumption that at such a high applied field, in any part of the material the Cu moments orient themselves in such a way that $\boldsymbol{B}_{\rm mol}^{\rm Nd\text{-}Cu}$ and $\boldsymbol{B}_{\rm appl}$ are perpendicular.

The Nd-Nd interaction in the undoped compound has also been studied by Thalmeier[36] and by Henggeler *et al.*[37] In these studies, a spin-wave theory was developed in which four different Nd-Nd interactions were considered rather than three, as in our approach. The authors claim that the spin-wave theory could describe their INS experiments quite well. However, the splitting of the ground-state doublet, as far as it is due to the Nd-Cu interaction, was found to be 0.59 meV, which corresponds to 6.8 K and differs substantially from our result of 5.0 K. The values they reported for the four Nd-Nd exchange constants cannot be compared with the three constants in our treatment, because they considered anisotropic exchange, described by tensors.

B. The doped crystal ($x = 0.15$)

As mentioned in Sec. V B, we have in our fitting procedure to account for the existence of inequivalent Nd ions in $Nd_{1.85}Ce_{0.15}CuO_{4-y}$. Based on that assumption we considered the three types of Nd ions with the three corresponding CEF-parameter sets proposed recently by Jandl *et al.*[18]

As is clear from Figs. 7, 8(a), and 8(b), the calculations with the parameters derived in Sec. V B, give a rather satisfactory description of the zero-field specific-heat data. The field effect on the specific heat, however, is not reproduced satisfactorily. In case the field is applied along the c axis, we note that the calculated specific-heat maxima are slightly higher than observed in the experiments. This is in contrast to the results for fields applied in the ab plane where the calculated specific-heat maxima are substantially lower. For both field directions, all the calculated curves are essentially narrower than the corresponding experimental ones. We suggest that at least part of the difference between the experimental and calculated specific heat is due to contributions of the electronic and lattice specific heat that might be larger in the doped than in the undoped crystal. Also, cerium might be in an intermediate valence state, see, e.g., Ref. 38, or a trivalent state, which could lead to an electronic specific heat or a contribution similar to the one of Nd^{3+} ions, respectively. There is still another explanation for the difference mentioned above. One should realize that the results reported by Jandl *et al.*[18] might be sample-dependent. Indeed, our crystal has been prepared by another method than that used by these authors. We point out, however, that calculations for the field of 8 T applied in the ab plane, with different sets of values for the fitting parameters $B_{\rm mol}^{\rm Nd\text{-}Cu}$, $n_{\rm RR}^{\rm dif}$, and $n_{\rm RR}^{\rm sum}$, resulted in essentially the same curve, even at varying the weight factors for the three types of Nd ions.

By now, there exist theories for the electronic specific heat in the doped compound.[39] On the one hand, Tornow and co-workers[39] pointed out that their model is not able to describe the anisotropy in the field effect in single crystals. On the other hand, we cannot expect to derive conclusions from our results in the present paper concerning the validity of such theories. This is because, as we explained in the previous Sec. V B, we could not give a correct treatment of the magnetic specific heat connected with the system of Nd and Cu ions, considering the large number of inequivalent Nd ions that occur in the doped compound. Furthermore, our experiments were carried out in a temperature range which is not low enough to observe the high electronic contribution to the specific heat (the ''linear term'') in the compound as reported in Ref. 23 for the polycrystalline material.

We point out again that at increasing applied field, the detailed features of the Nd-Nd interaction and the weights of the occupancy of the different inequivalent Nd ions become less and less important for the calculated curves. In Fig. 9, we show for both compounds the calculated specific heat at 8 T in the ab plane. It can be noticed that the calculated curves essentially coincide for the entire temperature range of consideration at this high value of the magnetic field. For completeness, we present in Fig. 9 also the experimental specific-heat curves for both compounds at fields of 4 and 8 T applied in the ab plane. At 4 T, the specific-heat curve of the doped compound is substantially shifted to higher temperatures and broadened, and the maximum is lower than that of the undoped compound. At 8 T, however, the experimental curves essentially coincide in the temperature range below 3 K, and the specific-heat maximum of the doped compound is about

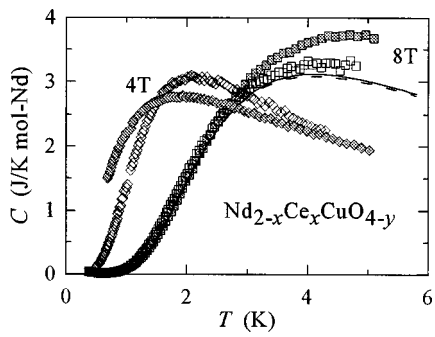

FIG. 9. Calculated specific heat of single-crystalline $Nd_{2-x}Ce_xCuO_{4-y}$ (with $x=0$: dashed line, and $x=0.15$: solid line) at a magnetic field of 8 T applied perpendicular to the crystallographic c axis. Also shown is the experimental specific heat ($x=0$: open symbols, and $x=0.15$: hatched symbols) at 4 T and 8 T applied in the same direction.

10% higher than that of the undoped one. These experimental results show clearly that the effect of an applied magnetic field on the specific heat is quite different in the doped and undoped compound.

VII. CONCLUDING REMARKS

We have measured the low-temperature specific heat of single-crystalline $Nd_{2-x}Ce_xCuO_{4-y}$ with $x=0$ and 0.15 in magnetic fields up to 8 T, applied both parallel and perpendicular to the crystallographic c axis. The results exhibit a remarkable anisotropy of the field effect on the specific heat of Nd_2CuO_{4-y}, which is less pronounced for the $x=0.15$ compound.

We arrived at an adequate description of the specific-heat data measured on the Nd_2CuO_{4-y} single crystal in different magnetic fields, applied along different symmetry axes. This was achieved (Sec. IV) by using a model in which the division of the Nd system into subsystems with different orientations of the Nd moment, is different for the two orientations of the applied field considered: 4 and 2 Nd subsystems for $\boldsymbol{B}_{\mathrm{appl}} \parallel c$ axis and $\boldsymbol{B}_{\mathrm{appl}} \perp c$ axis, respectively. Apart from CEF parameters the model contains three additional fitting parameters: one describing the Nd-Cu interaction and two, denoted by $n_{\mathrm{RR}}^{\mathrm{dif}}$ and $n_{\mathrm{RR}}^{\mathrm{sum}}$, being certain linear combinations of the three Nd-Nd exchange constants we introduced for the description of the interaction between the different Nd sublattices in Nd_2CuO_{4-y}. By using the CEF-parameter set proposed by Jandl *et al.*,[17] we could find a single set of values for the other three fitting parameters, which could describe simultaneously all experimental curves except the one measured for 2 T applied in the ab plane.

In Sec. VI we considered the nonconvergence of our computer program for small values (i.e., below about 4 T) of the applied field in the ab plane. We pointed out that this is consistent with the experimental findings of Cherny *et al.*,[5] who observed a first-order phase transition at 4.4 T in magnetization measurements in applied fields along the a direction.

We want to emphasize that our model is such that measuring the specific heat in zero field does not give all information on the Nd-Nd exchange: it only yields the parameter $n_{\mathrm{RR}}^{\mathrm{dif}}$. Measurements in nonzero field (in the ab plane) turn out to be necessary to obtain additional information, i.e., the parameter $n_{\mathrm{RR}}^{\mathrm{sum}}$. Even this is not all: the analysis we performed of our specific-heat measurements, determines only *two* relations between the *three* Nd-Nd exchange constants. It is quite possible that a third relation would be given by an analysis of the curve measured at 2 T in the ab plane, which curve has not been analyzed in the present paper by lack of an appropriate model. As may be inferred from the discussion in Sec. VI, such a model should describe configurations with 4 rather than 2 different directions of the Nd moments.

It should be noted that we applied many models, which were simpler than the model discussed in the present paper. Among these simpler models was, for example, a model with the same single-ion Hamiltonian for all Nd ions, i.e., truly the same, not only apart from symmetry transformations. For such simpler models we could find combinations of parameters that describe well the specific-heat curve at 0 T, and also the very similar curves for fields along the c axis. However, the same parameters turned out to be completely inadequate to give a correct description for the curves at fields in the ab plane.

The nature of the anisotropy of Nd_2CuO_{4-y} is a crucial point in understanding this material. In contrast to the analysis given in the present paper in which only Heisenberg exchange interaction is considered, in the spin-wave treatment of Thalmeier[36] and of Henggeler *et al.*[37] anisotropic exchange is assumed. Indeed, our measurements indicate a strong anisotropy in the effect of a magnetic field on the splitting of the ground-state doublet of the Nd ion. We assume in our model that this anisotropy should be ascribed to the peculiar form of the CEF Hamiltonian rather than to an anisotropy in the exchange interactions. [Our description implies that the specific heat is determined (mainly) by nondispersive modes in a spin-wave treatment.] In Sec. VI A we pointed out that the approach of Adelmann *et al.*[22] is not valid for the specific-heat measured at nonzero fields, since the anisotropy was not considered in their treatment.

We could not find a satisfactory theoretical description for the experimental specific-heat data of the $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ single crystal. We have attempted to account for the many different environments expected for the Nd ions—with different numbers of Ce neighbors which, moreover, can occupy different positions with respect to the Nd ions. For zero applied field we could well reproduce the experimental data (Fig. 7). For nonzero fields, however, all test distributions of inequivalent types of Nd ions we used, led to calculated specific-heat peaks that were significantly narrower than the ones found experimentally.

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[1] Y. Tokura, H. Takagi, and S. Uchida, Nature (London) **337**, 345 (1989); H. Takagi, S. Uchida, and Y. Tokura, Phys. Rev. Lett. **62**, 1197 (1989).

[2] M. Matsuda, Y. Endoh, K. Yamada, H. Kojima, I. Tanaka, R. J. Birgeneau, M. A. Kastner, and G. Shirane, Phys. Rev. B **45**, 12 548 (1992), and references therein.

[3] S. Skanthakumar, J. W. Lynn, J. L. Peng, and Z. Y. Li, J. Appl. Phys. **73**, 6326 (1993), and references therein.

[4] D. Petitgrand, A. H. Moudden, P. Galez, and P. Boutrouille, J. Less-Common Met. **164&165**, 768 (1990).

[5] A. S. Cherny, E. N. Khats'ko, G. Chouteau, J.-M. Louis, A. A. Stepanov, P. Wyder, S. N. Barilo, and D. I. Zhigunov, Phys. Rev. B **45**, 12 600 (1992).

[6] D. Petitgrand, L. Boudarène, P. Bourges, and P. Galez, J. Magn. Magn. Mater. **104-107**, 585 (1992).

[7] G. M. Luke, L. P. Le, B. J. Sternlieb, Y. J. Uemura, J. H. Brewer, R. Kadono, R. F. Kiefl, S. R. Kreitzman, T. M. Riseman, C. E. Stronach, M. R. Davis, S. Uchida, H. Takagi, Y. Tokura, Y. Hidaka, T. Murakami, J. Gopalakrishnan, A. W. Sleight, M. A. Subramanian, E. A. Early, J. T. Markert, M. B. Maple, and C. L. Seaman, Phys. Rev. B **42**, 7981 (1990); T. R. Thurston, M. Matsuda, K. Kakurai, K. Yamada, Y. Endoh, R. I. Birgeneau, P. M. Gehring, Y. Hidaka, M. A. Kastner, T. Murakami, and G. Shirane, Phys. Rev. Lett. **65**, 263 (1990); S. Skanthakumar, J. W. Lynn, J. P. Peng, and Z. Y. Li, J. Magn. Magn. Mater. **104-107**, 519 (1992).

[8] P. Hoffmann, M. Loewenhaupt, S. Horn, P. v. Aaken, and H.-D. Jostarndt, Physica B **163**, 271 (1990).

[9] U. Staub, P. Allenspach, A. Furrer, H. R. Ott, S.-W. Cheong, and Z. Fisk, Solid State Commun. **75**, 431 (1990); A. Furrer, P. Allenspach, J. Mesot, and U. Staub, Physica C **168**, 609 (1990).

[10] A. T. Boothroyd, S. M. Doyle, D. McK. Paul, D. S. Misra, and R. Osborn, (a) Physica C **165**, 17 (1990); (b) Phys. Rev. B **45**, 10 075 (1992).

[11] V. Nekvasil, Physica C **170**, 469 (1990).

[12] Y. U. Muzichka, E. A. Goremychkin, I. V. Sashin, M. Divis, V. Nekvasil, M. Nevriva, and G. Fillion, Solid State Commun. **82**, 461 (1992).

[13] C.-K. Loong and L. Soderholm, J. Alloys Compd. **181**, 241 (1992); Phys. Rev. B **48**, 14 001 (1993).

[14] W. Henggeler, T. Chattopadhyay, B. Roessli, D. I. Zhigunov, S. N. Barilo, and A. Furrer, Z. Phys. B **99**, 465 (1996).

[15] S. Jandl, M. Iliev, C. Thomsen, T. Ruf, M. Cardona, B. M. Wanklyn, and C. Chen, Solid State Commun. **87**, 609 (1993).

[16] P. Dufour, S. Jandl, C. Thomsen, M. Cardona, B. M. Wanklyn, and C. Changkang, Phys. Rev. B **51**, 1053 (1995).

[17] S. Jandl, P. Dufour, T. Strach, T. Ruf, M. Cardona, V. Nekvasil, C. Chen, and B. M. Wanklyn, Phys. Rev. B **52**, 15 558 (1995).

[18] S. Jandl, P. Dufour, T. Strach, T. Ruf, M. Cardona, V. Nekvasil, C. Chen, B. M. Wanklyn, and S. Piñol, Phys. Rev. B **53**, 8632 (1996).

[19] M. F. Hundley, J. D. Thompson, S.-W. Cheong, Z. Fisk, and S. B. Oseroff, Physica C **158**, 102 (1989).

[20] S. Ghamaty, B. W. Lee, J. T. Markert, E. A. Early, T. Björnholm, C. L. Seaman, and M. B. Maple, Physica C **160**, 217 (1989).

[21] M. B. Maple, N. Y. Ayoub, T. Björnholm, E. A. Early, S. Ghamaty, B. W. Lee, J. T. Markert, J. J. Neumeier, and C. L. Seaman, Physica C **162-164**, 296 (1989).

[22] P. Adelmann, R. Ahrens, G. Czjzek, G. Roth, H. Schmidt, and C. Steinleitner, Phys. Rev. B **46**, 3619 (1992).

[23] T. Brugger, T. Schreiner, G. Roth, P. Adelmann, and G. Czjzek, Phys. Rev. Lett. **71**, 2481 (1993).

[24] V. B. Barbeta, P. Allenspach, M. B. Maple, and R. F. Jardim, J. Magn. Magn. Mater. **145**, 391 (1995).

[25] K. Oka, M. J. V. Menken, Z. Tarnawski, A. A. Menovsky, A. M. Moe, T. S. Han, H. Unoki, T. Ito, and Y. Ohashi, J. Cryst. Growth **137**, 479 (1994).

[26] N. T. Hien, thesis, University of Amsterdam, 1997.

[27] M. Loewenhaupt, A. Metz, N. M. Pyka, D. McK. Paul, J. Martin, V. H. M. Duijn, J. J. M. Franse, H. Mutka, and W. Schmidt, Ann. Phys. (Leipzig) **5**, 197 (1996).

[28] M. Loewenhaupt (private communication).

[29] V. H. M. Duijn, thesis, University of Amsterdam (unpublished).

[30] A. J. Kassman, J. Chem. Phys. **53**, 4118 (1970).

[31] G. Balakrishnan, S. K. Malik, C. K. Subramaniam, D. McK. Paul, C. Piñol, and R. Vijayaraghavan, J. Magn. Magn. Mater. **104-107**, 469 (1992).

[32] M. Matsuda, Y. Endoh, K. Yamada, H. Kojima, I. Tanaka, R. J. Birgeneau, M. A. Kastner, and G. Shirane, Phys. Rev. B **45**, 12 548 (1992).

[33] M. Loewenhaupt, P. Fabi, S. Horn, P. V. Aken, and A. Severing, J. Magn. Magn. Mater. **140-144**, 1293 (1995).

[34] T. Chattopadhyay, P. J. Brown, and U. Köbler, Physica C **177**, 294 (1991).

[35] T. Chattopadhyay, and K. Siemensmeyer, Europhys. Lett. **29**, 579 (1995).

[36] P. Thalmeier, Physica C **266**, 89 (1996).

[37] W. Henggeler, T. Chattopadhyay, P. Thalmeier, P. Vorderwisch, and A. Furrer, Europhys. Lett. **34**, 537 (1996).

[38] T. C. Hoang, E. Moran, A. I. Nazzal, J. B. Torrance, and P. W. Wang, Physica C **159**, 625 (1989).

[39] S. Tornow, V. Zevin, and G. Zwicknagl, Ann. Phys. (Leipzig) **5**, 501 (1996), and references therein.