4f-spin dynamics in La$_{2-x-y}$Sr$_x$Nd$_y$CuO$_4$

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We have performed inelastic magnetic neutron scattering experiments on La$_{2-x-y}$Sr$_x$Nd$_y$CuO$_4$ ($0 \leq x \leq 0.2$ and $0.1 \leq y \leq 0.6$) in order to study the Nd 4f-spin dynamics at low energies ($\hbar\omega \lesssim 1$ meV). In all samples we find at high temperatures a quasielastic line (Lorentzian) with a linewidth which decreases on lowering the temperature. The temperature dependence of the quasielastic linewidth $\Gamma/2(T)$ can be explained with an Orbach process, i.e., a relaxation via the coupling between crystal field excitations and phonons. At low temperatures the Nd-4f magnetic response $S(\mathbf{Q}, \omega)$ correlates with the electronic properties of the CuO$_2$ layers. In the insulator La$_{2-y}$Nd$_y$CuO$_4$ ($y = 0.1, 0.3$) the quasielastic line vanishes below 80 K and an inelastic excitation occurs. This directly indicates the splitting of the Nd^{3+} ground state Kramers doublet due to the static antiferromagnetic order of the Cu moments. In La$_{1.7-x}$Sr$_x$Nd$_{0.3}$CuO$_4$ with $x = 0.12, 0.15$ and La$_{1.4-x}$Sr$_x$Nd$_{0.6}$CuO$_4$ with $x = 0.1, 0.12, 0.15, 0.18$ superconductivity is strongly suppressed. In these compounds we observe a temperature independent broad quasielastic line of Gaussian shape below $T \approx 30$ K. This suggests a distribution of various internal fields on different Nd sites and is interpreted in the frame of the stripe model. In La$_{1.8-y}$Sr$_{0.2}$Nd$_y$CuO$_4$ ($y = 0.3, 0.6$) such a quasielastic broadening is not observed even at lowest temperature. [S0163-1829(99)04537-3]

I. INTRODUCTION

In the high-T_c superconductors a close interplay between superconductivity and magnetism exists. The parent compounds of the cuprates are antiferromagnetic insulators. Doping with holes or electrons destroys the long range order but antiferromagnetic correlations persist even in the superconducting region. *Inelastic* incommensurate magnetic peaks in superconducting La$_{2-x}$Sr$_x$CuO$_4$ (LSCO) show that superconductivity and magnetic correlations coexist.[1] This phenomenon has regained attraction since in 1995 *elastic* peaks at the same incommensurate positions were found in La$_{1.48}$Sr$_{0.12}$Nd$_{0.4}$CuO$_4$. These elastic peaks are interpreted in terms of hole-rich and spin-rich domains in the CuO$_2$ layers, i.e., the well known stripe picture.[2] In LSCO (T phase) doping with Nd induces a further low temperature phase transition for Nd$\geq$0.18. For $x = 0$ there is a transition from the low temperature orthorhombic LTO to the less orthorhombic Pccn phase[3] whereas for $x \gtrsim 0.1$ the transition is to the tetragonal LTT phase. In the latter superconductivity is strongly suppressed for certain Sr concentrations.[4] In the LTT phase the tilt of the CuO$_6$ octahedra can serve as pinning potential for the dynamical stripe correlations. Hence, the inelastic peaks become elastic indicating a formation of *static* antiphase antiferromagnetic domains in the CuO$_2$ planes which are separated by quasi-one-dimensional stripes containing the doped charge carriers. Recently, inelastic incommensu-

rate peaks have also been observed in superconducting YBa$_2$Cu$_3$O$_{6.6}$ (Ref. 5) giving rise to the question whether stripes are a general feature of cuprate based high-T_c superconductors.

In addition to the investigation of the Cu subsystem, the dynamics of the Nd spins in Nd$_{2-x}$Ce$_x$CuO$_4$ (NCCO, T' phase) has been examined extensively by several groups.[6–9] Henggeler *et al.*[10] performed neutron scattering experiments to examine the spin excitation spectrum at low temperatures. The same group explained the heavy-fermion-like large γ coefficient in low temperature specific heat measurements[11] of NCCO by the shift of spectral weight of the Nd modes to lower energies with increasing number of charge carriers.[12] Specific heat measurements show a Schottky anomaly in the parent compound Nd$_2$CuO$_4$.[13] This is explained by the presence of Nd-Cu interactions being responsible for the splitting of the Nd^{3+} ground state Kramers doublet as, e.g., observed in Raman and neutron scattering experiments.[14–17] At higher temperatures in neutron scattering experiments on powder samples of NCCO (Ref. 17) and on a single crystal of Nd$_2$CuO$_4$ (Ref. 8) a quasielastic (QE) Lorentzian is observed with a linewidth that increases almost linearly with increasing temperature. At lower temperatures the line shape turns into a Gaussian with an almost constant linewidth. Similar features have been presented for NdBa$_2$Cu$_3$O$_{7-\delta}$.[18] In this compound the QE Gaussian and the Lorentzian coexist.

In the present work we report on inelastic magnetic neu-

tron scattering experiments on $La_{2-x-y}Sr_xNd_yCuO_4$ at various temperatures. We have investigated the $4f$ magnetic response for samples with Sr concentrations $0 \leq x \leq 0.2$ and Nd concentrations $0.1 \leq y \leq 0.6$ at low energies (typically ~ 1 meV) in order to obtain information about the Cu magnetism in the CuO_2 layers via the Nd-Cu interaction. The paper is organized as follows. The next section describes the experimental technique. The presentation of our results and their discussion follows in Sec. III which is divided into two parts as justified by our experimental findings. In Sec. IV we will give a brief summary.

II. EXPERIMENTAL

We performed temperature dependent studies on $La_{1.9}Nd_{0.1}CuO_4$, $La_{1.7-x}Sr_xNd_{0.3}CuO_4$ ($x=0,0.12,0.15,0.2$), and $La_{1.4-x}Sr_xNd_{0.6}CuO_4$ ($x=0.1,0.12,0.15,0.18,0.2$) using the time-of-flight (TOF) spectrometers V3 NEAT (Ref. 19) (HMI Berlin), G6.2 MIBEMOL (LLB Saclay), and IN5 (ILL Grenoble). All spectrometers are located at cold neutron beam lines and use chopper systems for monochromatization which give very sharp and clean resolution functions. The energy chosen for the incident neutrons ranges from $E_i=3.15$ meV ($\equiv 5.1$ Å) to $E_i=1.28$ meV ($\equiv 8$ Å) resulting in energy resolutions between $\Delta E \approx 50$ μeV and $\Delta E \approx 20$ μeV (HWHM), respectively. Additionally, $La_{1.45}Sr_{0.15}Nd_{0.4}CuO_4$ was measured at the IN6 (ILL, Grenoble) with $E_i=3.15$ meV ($\Delta E \approx 45$ μeV).[20] This spectrometer uses Bragg diffraction on single crystals to monochromize the neutron beam. In all cases a Vanadium standard for calibration and an empty can measurement of the Al flat plate for background correction were carried out. For the experiments we used well characterized powder samples[21] of typically $m=20$ g and a standard cryostat for cooling. Details of the data analysis are described elsewhere.[22] Since we could not find any Q dependence of the magnetic signals $S(Q,\omega)$ is averaged over a broad Q window for all spectra to obtain a better statistics.

III. RESULTS AND DISCUSSION

A. 4f spin relaxation due to coupling of phonons and crystal field excitations

In all samples a quasielastic line of Lorentzian shape is observed at high temperatures. This is illustrated in Fig. 1 showing spectra of $La_{1.25}Sr_{0.15}Nd_{0.6}CuO_4$ for two different temperatures. The linewidth $\Gamma/2(T)$ of the Lorentzian decreases with decreasing temperature and shows the *same* temperature dependence in *all* compounds within experimental error. In Fig. 2 the temperature dependence of the QE linewidth for samples with different Sr and Nd concentrations is plotted. Above about 100 K the linewidth increases almost linearly with increasing temperature. Below this temperature the slope is drastically reduced although the linewidth decreases furthermore on lowering the temperature. The residual linewidth for $T \to 0$ K is below the resolution limit even in the experiments when an energy resolution of 20 μeV (HWHM) was chosen. Generally, it is hard to detect the QE Lorentzian for $T \lesssim 20$ K. However, when we do not include a QE Lorentzian in our fitting procedure we observe an increase of the elastic intensity in this temperature
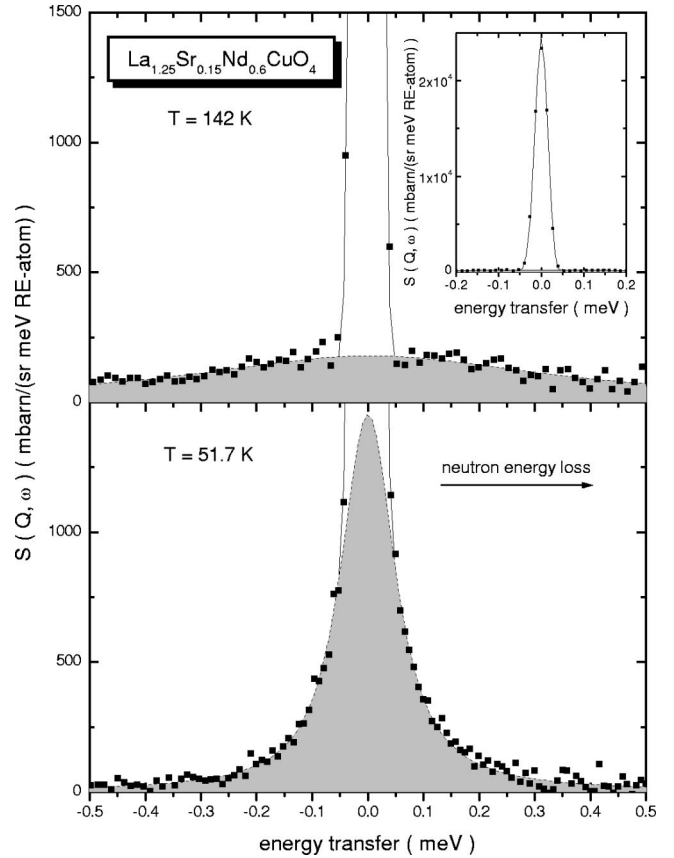

FIG. 1. Background corrected spectra of $La_{1.25}Sr_{0.15}Nd_{0.6}CuO_4$ (IN5). The full line is the best fit including the nuclear incoherent elastic scattering. The magnetic contribution (QE Lorentzian convolved with the instrumental resolution function) is illustrated by the shaded area. The inset shows the overall elastic scattering.

region. Since the coherent and incoherent elastic scattering intensity should be almost temperature independent for each sample we can conclude that this additional intensity originates from magnetic scattering. If the fit for the QE Lorentzian yielded a value smaller than half of the resolution (HWHM) we fixed the linewidth at $\Gamma/2=0$ for this temperature.
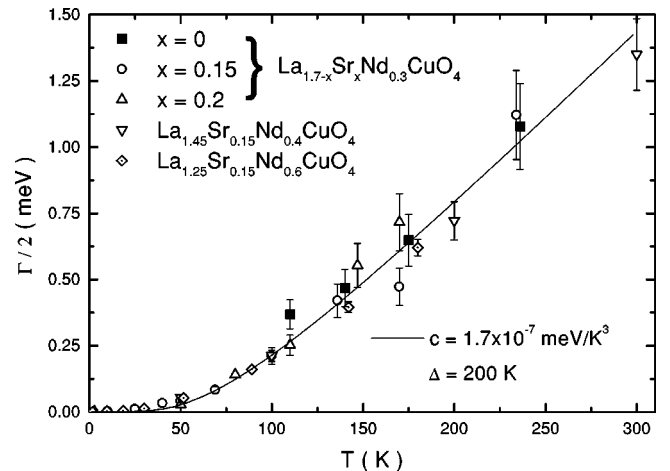

FIG. 2. Quasielastic linewidths of $La_{1.7-x}Sr_xNd_{0.3}CuO_4$ with $x=0,0.15,0.2$ (NEAT), $La_{1.45}Sr_{0.15}Nd_{0.4}CuO_4$ (IN6), and $La_{1.25}Sr_{0.15}Nd_{0.6}CuO_4$ (IN5). The full line is a function according to Eq. (1) with a mean value for parameter c (see text).
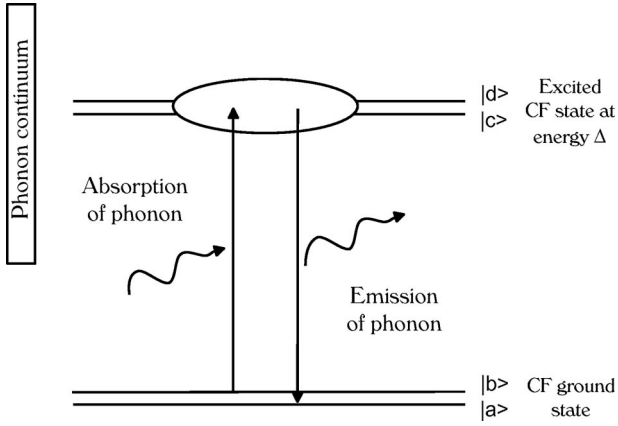
FIG. 3. Schematic diagram of the Orbach relaxation process. $|a\rangle$ and $|b\rangle$ denote the two states of the CF ground state, $|c\rangle$ and $|d\rangle$ of an excited state, respectively (for a Kramers ion like Nd^{3+}). The arrows indicate a possible transition with involvement of a phonon.

The QE linewidth is related to the spin fluctuation frequency via $\Gamma/2 = \hbar/\tau$. Thus the decrease of the linewidth with decreasing temperature is a direct evidence for the lowering of the 4f-spin fluctuation frequency.

The observation of a magnetic QE line in high-T_c superconductors and related materials has been a subject for discussion for over a decade.[17,18,23–25] During this time two attempts to describe the 4f spin relaxation, i.e., the temperature dependence of the QE linewidth, were suggested: (i) via the interaction of 4f spins with conduction electron spins and (ii) by exchange interaction between 4f spins themselves. In the first case the linewidth should increase linearly with temperature as $\Gamma/2 \propto [N(\epsilon_F) \cdot J_{ex}]^2 \cdot T$ (Korringa law) with $N(\epsilon_F)$ the density of electron states at the Fermi energy and J_{ex} the exchange integral between 4f- and conduction electron spins.[26] Such a temperature dependence was often found in intermetallic systems.[27] In the second case a power law is expected.[28]

From our data we can exclude both cases, since we observe the same temperature dependence of $\Gamma/2(T)$ in samples with $(x>0)$ and without charge carriers $(x=0)$ which rules out scenario (i). Again, the same temperature dependence of $\Gamma/2(T)$ is obtained for samples with high Nd content $(y=0.6)$ and low Nd content $(y=0.1)$. For a small Nd concentration the Nd-Nd exchange interaction should be weak, which rules out scenario (ii). After a detailed analysis of our data we found that the relaxation of the 4f spins can be well described with a two-phonon Orbach process[29] assuming a coupling between crystal field (CF) excitations and phonons. The Orbach process is sketched in Fig. 3. In this relaxation process a Nd ion which is in state $|b\rangle$ can absorb a phonon and hence is excited to state $|c\rangle$ (or $|d\rangle$). From this state it can fall down to state $|a\rangle$ by emitting a second phonon. The temperature dependence of the QE linewidth then follows

$$\Gamma/2(T) = \Gamma_0/2 + c \cdot \Delta_{CF}^3/(e^{\Delta_{CF}/T} - 1), \qquad (1)$$

where Δ_{CF} is the energy of an excited crystal field state and c is a factor which among others considers the coupling of the CF ground state with the excited CF state. In Eq. (1) only for $T \gg \Delta_{CF}$ the linewidth is proportional to the temperature.

We fit this function to the experimental data of many different samples and obtained values around 200 K for Δ_{CF}. Since the energy of the excited CF state should not differ much between our samples, we took a mean value of $\Delta_{CF} = 200$ K in the following. Unfortunately, the crystal field scheme for Nd doped LSCO has yet not been evaluated. However, $\Delta_{CF} \approx 200$ K coincides roughly with the energy of the first excited state ($\Delta_{CF} = 173$ K) in Nd$_2$CuO$_4$.[30]

A fit with Eq. (1) reveals $c = 1.54 \times 10^{-7}$ meV/K^3 and $c = 1.93 \times 10^{-7}$ meV/K^3 for La$_{1.25}$Sr$_{0.15}$Nd$_{0.6}$CuO$_4$ and La$_{1.5}$Sr$_{0.2}$Nd$_{0.3}$CuO$_4$, respectively. A residual linewidth of $\Gamma_0/2 = 10$ μeV has also been taken into account but does not influence the other parameters markedly.[31] The values for c are in the expected range[29] and are in good agreement with the reported value for LSCO probed with Er^{3+} spins.[32]

We now turn to a discussion of the relevance of the Orbach relaxation process in other systems. A comparison of the QE linewidths in La$_{2-x-y}$Sr$_x$Nd$_y$CuO$_4$ with the reported values in Nd$_2$CuO$_4$ (Refs. 8,17) and NdBa$_2$Cu$_3$O$_{7-\delta}$ (Ref. 18) shows that the absolute values of $\Gamma/2$ are of the same order of magnitude in all compounds. For Nd$_2$CuO$_4$ only a few data points of the QE linewidth exist. The agreement between the data of Casalta et al. for a single crystal and Loewenhaupt et al. for a powder sample is rather poor. A fit with the above function yields $\Gamma_0/2 = 0.24$ meV, $c = 1.42 \times 10^{-7}$ meV/K^3 and $\Gamma_0/2 = 0.17$ meV, $c = 8.5 \times 10^{-8}$ meV/K^3 for the data of Loewenhaupt et al. and Casalta et al., respectively (Δ_{CF} was fixed at 175 K). Note that in both cases a large residual linewidth is obtained from the fit. This broad Lorentzian line is probably masked by the broad Gaussian line. A large value of $\Gamma_0/2$ was directly observed in NdBa$_2$Cu$_3$O$_{7-\delta}$.[18] A fit of the data on NdBa$_2$Cu$_3$O$_6$ with Eq. (1) reveals $c = 3.35 \times 10^{-8}$ meV/K^3 ($\Gamma_0/2$ and Δ_{CF} fixed at 0.235 meV and 410 K,[33] respectively). Two things are worth mentioning. First, the choice of 410 K for Δ_{CF} has the consequence that the increase of the linewidth is suppressed up to higher temperatures. This is indeed observed in NdBa$_2$Cu$_3$O$_{7-\delta}$.[18] Second, c is roughly an order of magnitude smaller than in La$_{2-x-y}$Sr$_x$Nd$_y$CuO$_4$. This is in agreement with the observations of Shimizu et al. for an Er^{3+} spin probe in LSCO and YBCO.[32] The broad residual linewidth in the concentrated systems might be caused by a strongly enhanced Nd-Nd interaction.

A QE Lorentzian was also observed in Pb$_2$Sr$_2$TbCu$_3$O$_8$ (Ref. 34) and Pb$_2$Sr$_2$Tb$_{0.5}$Ca$_{0.5}$Cu$_3$O$_8$.[28] In both compounds Tb has a quasidoublet ground state, i.e., two singlets separated by only a few μeV. It was found that in Pb$_2$Sr$_2$Tb$_{0.5}$Ca$_{0.5}$Cu$_3$O$_8$ the temperature dependence of $\Gamma(T)$ obeys a power law t^ν with $\nu = 2.8$ [$t = (T - T_N)/T_N$]. Conclusively the authors claimed that the rare-earth exchange interaction might be the dominant process for the 4f-spin relaxation. The same group obtained similar results for Y$_{0.9}$Tb$_{0.1}$Ba$_2$Cu$_3$O$_7$.[35] Since the concentration of Tb in this compound is very low the above interpretation of a strong rare-earth exchange interaction seems rather unlikely. In contrast, a very recent reanalysis of the data showed that the temperature dependence of the QE linewidth also follows the two-phonon Orbach process.[36] Taking all these facts into account it seems reasonable to assume that the 4f spin relaxation in high-T_c superconductors and related materials is
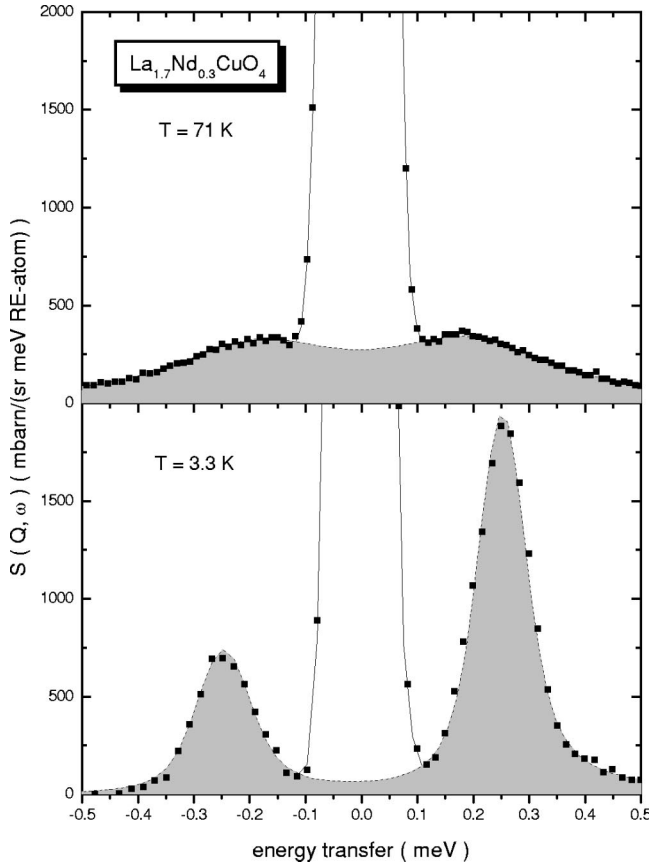
FIG. 4. Background corrected spectrum of $La_{1.7}Nd_{0.3}CuO_4$ at $T = 71$ K (MIBEMOL) and $T = 3.3$ K (NEAT). The full line is the best fit including the nuclear incoherent elastic scattering. The magnetic contribution (INE Lorentzian) is given by the shaded area.

caused by CF transitions assisted by phonons and *not* by interaction with conduction electrons. Moreover, the interpretation of the deviation from a linear temperature dependence of $\Gamma/2(T)$ as opening of a gap has to be reexamined.[37,38]

To conclude this section we want to mention that a coupling between these two elementary excitations was already reported in Raman scattering studies on several high-T_c superconductors and related compounds.[39] Furthermore, ESR data have been discussed in terms of an Orbach process.[32,40] These data are consistent with our interpretation of the temperature dependence of the QE linewidth (see also our previous work[41]).

B. Magnetic response due to Nd-Cu interaction

In contrast to the above described behavior the $4f$ magnetic response at low temperatures correlates with the electronic properties of the CuO_2 layers. Depending on the dopant concentration we find different features of the magnetic response which we will now discuss in detail.

1. x=0

In insulating $La_{2-y}Nd_yCuO_4$ with $y = 0.1, 0.3$ the $4f$ magnetic response changes from a QE Lorentzian to an inelastic (INE) excitation below about 80 K (Ref. 42) (see Fig. 4). This INE excitation clearly indicates the splitting of the
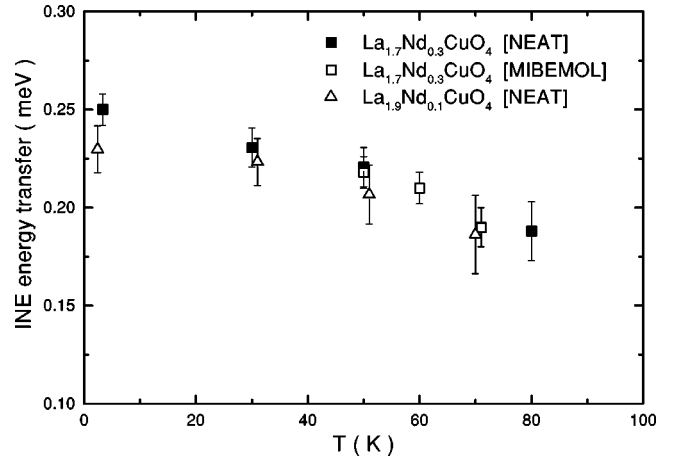

FIG. 5. Temperature dependence of the energy excitation in $La_{2-y}Nd_yCuO_4$ for $y = 0.1$ and $y = 0.3$.

Nd^{3+} ground state Kramers doublet due to the internal exchange field of ordered Cu moments and shows the strong interaction between the Cu and Nd subsystems. Although the Cu ordering temperature is much higher, this excitation becomes first detectable below 80 K since at higher temperature the linewidth is larger than the observed energy splitting. We mention that this is in contrast to neutron scattering results on Nd_2CuO_4 where the magnetic signal remains QE down to 5 K (Ref. 17) possibly due to Nd-Nd interactions. For the sample with smaller Nd content, namely, $La_{1.9}Nd_{0.1}CuO_4$, we find a similar behavior as for $y = 0.3$. In contrast to $y = 0.3$ in this sample only a minority fraction of about 20% changes to the $Pccn$ phase.[43]

The temperature dependence of the energy splitting for both compounds is plotted in Fig. 5. An increase of the splitting, i.e., of the internal exchange field at the Nd site, is clearly visible. Our findings agree roughly with the data of Chou *et al.*[44] who measured the internal field at the La site in La_2CuO_4 with ^{139}La NQR. According to their results the temperature dependence of the internal field can be described with a power law $(1 - T/T_N)^\beta$ ($T_N = 250$ K and $\beta = 0.41$). This means an increase of about 17% from 80 to 3 K and agrees roughly with our results in $La_{1.9}Nd_{0.1}CuO_4$. It is obvious that the exchange field at the Nd site is influenced by both, the staggered magnetization and the direction of the Cu spins. In $La_{1.7}Nd_{0.3}CuO_4$ the structural transition from LTO $\rightarrow Pccn$ is accompanied by a Cu spin reorientation.[3,45] Thus, the enhanced splitting in $y = 0.3$ compared to $y = 0.1$ for $T \rightarrow 0$ K might be due to the difference in the direction of the Cu spins. Finally, we want to mention that the value of ΔE coincides with that derived from the Schottky anomaly found in low temperature specific heat measurements.[46]

2. x=0.12

Static ordering of charges and spins was first reported in $La_{1.48}Sr_{0.12}Nd_{0.4}CuO_4$.[2] Similar results are obtained for $x = 0.15$ (Ref. 47) where magnetic order has also been observed with μ^+SR experiments[48] and Mössbauer experiments.[49]

We performed measurements on several compounds related to this composition and found similar properties at low temperatures. As a representative sample we chose
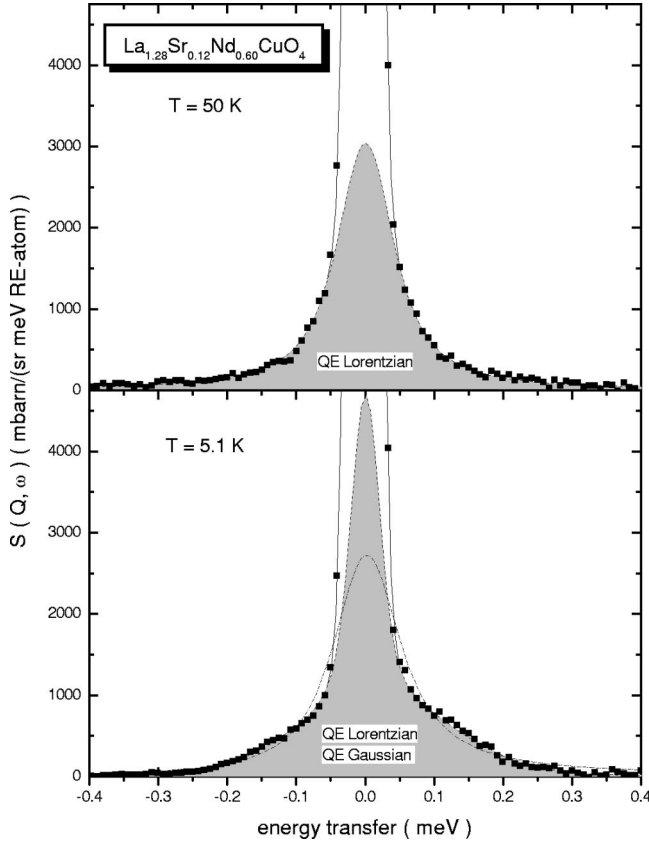
FIG. 6. Background corrected spectrum of La$_{1.28}$Sr$_{0.12}$Nd$_{0.6}$CuO$_4$ (MIBEMOL). The full line is the best fit including the nuclear incoherent elastic scattering. The magnetic contribution is illustrated by the shaded area. The dotted-dashed line shows the fit assuming a single Lorentzian.

La$_{1.28}$Sr$_{0.12}$Nd$_{0.6}$CuO$_4$ to explain the general features that are observed when the type of ordering changes from the well known spin structure for $x=0$ into an antiferromagnetic stripe pattern of ordered spins and charges in the CuO$_2$ layers. Above the antiferromagnetic ordering temperature ($T_N \approx 30$ K) a single QE Lorentzian line is found as discussed in Sec. III A. When the temperature is further lowered below T_N the linewidth does not become smaller as expected from the two-phonon Orbach process. Instead, a broad magnetic response of almost constant width centered around the elastic peak is visible (see Fig. 6). The analysis of our results shows that in addition to the Lorentzian a Gaussian line is necessary to accurately describe the data. The width of this Gaussian is almost temperature independent ($\Gamma_{Gaussian}/2 \approx 0.13$ meV, see Fig. 7).

The observation of a broad QE Gaussian line instead of a well resolved INE excitation as for $x=0$ infers a distribution of different energy splittings on different Nd sites. This is a direct hint on spatial inhomogeneities in the CuO$_2$ planes and is probably caused by the formation of stripes. In this picture the Kramers doublet of a Nd^{3+} ion, which is located ''near'' a charge stripe and thus is not split up, contributes to the Lorentzian signal which is still observable (at least the splitting must be small compared to the width of this Lorentzian). The comparison of the width of the Gaussian line in $x = 0.12$ with the energy excitation of the inelastic line ($x = 0$) reveals a reduced (average) splitting, which is related to
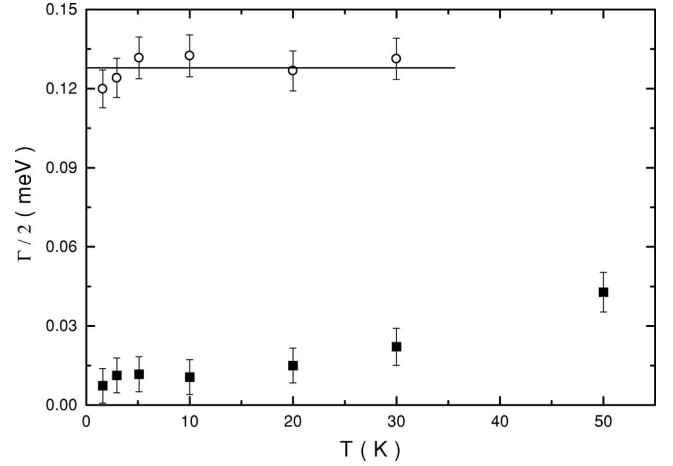

FIG. 7. Quasielastic linewidths (HWHM) in La$_{1.28}$Sr$_{0.12}$Nd$_{0.6}$CuO$_4$. Open circles are associated with the Gaussian line. Closed squares symbolize Lorentzian linewidths.

a reduced zero temperature staggered magnetization in the CuO$_2$ planes. This finding coincides with the μ^+SR experiments[48] on La$_{1.85-y}$Sr$_{0.15}$Nd$_y$CuO$_4$ where a decrease of the muon spin rotation frequency compared to La$_{1.7}$Nd$_{0.3}$CuO$_4$ (Ref. 50) was found. This was interpreted as a decrease of the average magnetic field at the muon site in Sr doped compounds.

In Fig. 8 the intensities of the Lorentzian and Gaussian signals are plotted versus temperature. Above 50 K the intensity of the single Lorentzian line raises with decreasing temperature due to an increase in the thermal occupation of the ground state (not shown). Below this temperature firstly the intensity of the Lorentzian decreases linearly for $T \gtrsim 10$ K whereas the intensity of the Gaussian raises. This can be interpreted as a decrease in the number of paramagnetic Nd ions (i.e., with a splitting smaller than the Lorentzian linewidth). This is expected since the Lorentzian linewidth decreases with decreasing temperature and therefore the number of Nd ions for which the above condition holds reduces. Secondly, between 10 and 3 K, a plateaulike level is reached. For $T \lesssim 10$ K the Lorentzian linewidth is well be-
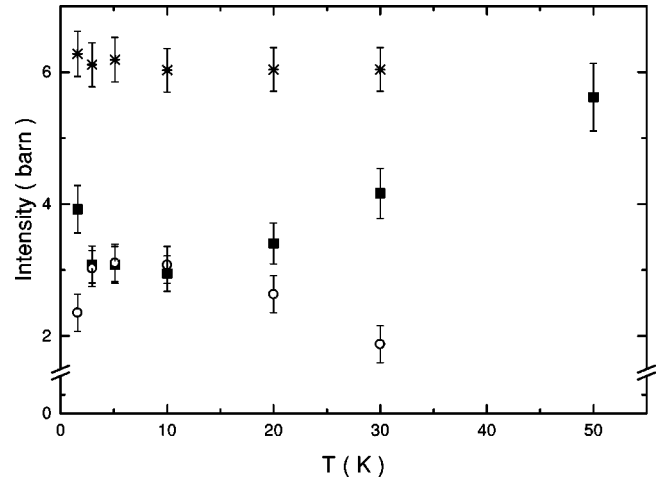

FIG. 8. Intensity of Gaussian (open circles) and Lorentzian (closed squares) line in La$_{1.28}$Sr$_{0.12}$Nd$_{0.6}$CuO$_4$. The asterisks are the overall magnetic intensity.
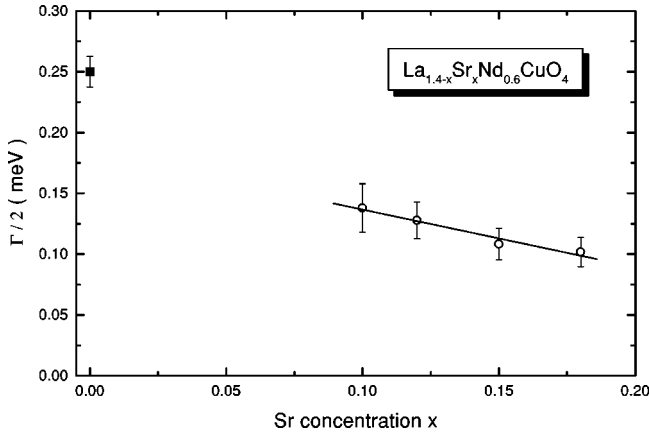
FIG. 9. Gaussian line widths (open circles) vs Sr concentration. The solid line is a guide to the eye. For comparison the INE energy splitting of $La_{1.7}Nd_{0.3}CuO_4$ at 3.3 K is also plotted (closed square).

FIG. 10. Background corrected spectrum of $La_{1.2}Sr_{0.2}Nd_{0.6}CuO_4$ at 1.6 K. The magnetic contribution (QE Lorentzian) is given by the shaded area. The open circles represent the magnetic response which is found in $La_{1.28}Sr_{0.12}Nd_{0.6}CuO_4$ at the same temperature. We compare these samples since both were measured on the same spectrometer (MIBEMOL). Due to the Bose factor the spectral weight is shifted asymmetrically to the neutron energy loss side.

low the resolution limit and so small that we cannot distinguish between a Lorentzian line and the elastic line. Finally, below 3 K a strong increase of the Lorentzian intensity (which we cannot distinguish from an increase of the elastic intensity) is observed. At a similar temperature a pronounced increase of the magnetic intensity due to ordering of the Nd moments was reported in $La_{1.48}Sr_{0.12}Nd_{0.4}CuO_4$.[2]

3. Sr dependence of the magnetic response

In addition to the abovementioned two Sr concentrations $x=0$ (with $y=0.1$, 0.3) and $x=0.12$ (with $y=0.6$) experiments with varying Sr compositions were carried out on $La_{1.7-x}Sr_xNd_{0.3}CuO_4$ with $x=0.12,0.15,0.2$ and $La_{1.4-x}Sr_xNd_{0.6}CuO_4$ with $x=0.1,0.15,0.18,0.2$. For clarity the data of the series with $y=0.3$ are not shown since the observations in these compounds are similar to the findings in the related samples with $y=0.6$.

To study the Sr dependence of the magnetic signal at low temperatures in detail we extended our measurements to samples with a Nd content of $y=0.6$ with $0.1 \leqslant x \leqslant 0.2$. In contrast to $La_{1.7-x}Sr_xNd_{0.3}CuO_4$ all of these samples lie in the region of the phase diagram where superconductivity is strongly suppressed[4] and thus a broad magnetic response at low temperatures is expected. Such a response was found in all samples except for $La_{1.2}Sr_{0.2}Nd_{0.6}CuO_4$. The linewidths of the QE Gaussians are plotted in Fig. 9 (these data points are fits under the assumption that $\Gamma_{Gaussian}/2$ is temperature independent for each compound). The decrease of the linewidth with increasing Sr content is related to a reduction of the average staggered magnetization in the CuO_2 planes. This observation is consistent with the findings in $La_{1.6-x}Sr_xNd_{0.4}CuO_4$ with $x=0.12$, 0.15, and 0.2.[47]

Unfortunately the spectra of $La_{1.3}Sr_{0.1}Nd_{0.6}CuO_4$ showed strongly enhanced background probably due the diffusion of air into the neutron flight path. Nevertheless, the analysis showed that a QE Gaussian is not consistent with the data in the whole temperature range 1.8–30 K although it is accurate enough at certain temperatures. A better agreement is obtained when we use an INE Gaussian line. This might reflect a mixture of both types of signals which we observe in $0 < x=0.1<0.12,0.15,0.18$, i.e., an INE excitation and QE Gaussian, respectively. Furthermore we note that this com-
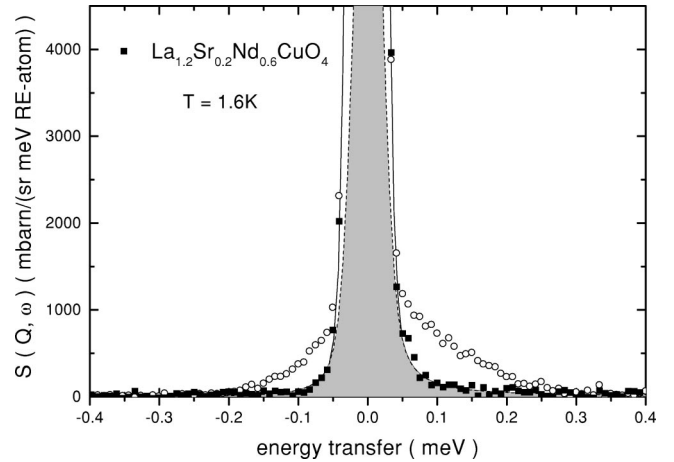
pound is closest to the *Pccn* phase.

We could not detect any QE broadening in $La_{1.8-y}Sr_{0.2}Nd_yCuO_4$ with $y=0.3$ and 0.6 at lowest temperature (see Fig. 10). For $y=0.3$ this behavior is expected since this compound is a bulk superconductor below $T_c \approx 25$ K and hence no magnetic order in the CuO_2 planes is expected. Indeed, in a superconductor with even higher Nd concentration ($La_{1.4}Sr_{0.2}Nd_{0.4}CuO_4$) Nachumi *et al.* could not find any hints for magnetic order in a recent μSR experiment.[51] In contrast to this, the absence of a QE broadening is surprising in the compound with $y=0.6$ because superconductivity is strongly suppressed and hence a broad magnetic response is expected. Our sample has also been studied in a recent μSR$^+$ experiment which shows magnetic order below about 15 K.[52] This finding seems to contradict our neutron scattering results. There might be two reasons why a broadening is not observable in the present neutron scattering experiment: (i) the splitting of the Nd Kramers ground state is too small or (ii) the intensity of the QE Gaussian is too low. From the energy resolution chosen in our experiment we can conclude that a possible average splitting of the Nd ground state must be below ≈ 20 μeV if case (i) were true. This contradicts the value which is obtained by extrapolating the Gaussian widths of Fig. 9 to $x=0.2$. Indeed, scenario (ii) is more likely since we observe a drastic drop of the intensity of the QE Gaussian in $y=0.18$ compared to $y=0.15$ at lowest temperature. At present we have no interpretation for this drop of the intensity. Further combined neutron and μSR studies are necessary in order to investigate and understand this pronounced concentration dependence of the intensity.

IV. SUMMARY

To summarize, we have presented inelastic magnetic neutron scattering experiments on Nd doped $La_{2-x}Sr_xCuO_4$. In all samples at higher temperatures a quasielastic line of Lorentzian shape is observed with a linewidth which decreases with decreasing temperature. The temperature depen-

dence of this width, i.e., the relaxation of the Nd $4f$ moments is dominated by the Orbach relaxation process via the coupling of phonons and CF excitations and does not depend on the charge carrier concentration in the CuO$_2$ planes. The low-temperature behavior of the magnetic response clearly correlates with the electronic properties of the CuO$_2$ layers. In the undoped samples ($x=0$) below about 80 K an inelastic excitation occurs which shows the splitting of the Nd^{3+} Kramers doublet ground state due to the Cu exchange field at the Nd site. In La$_{1.7-x}$Sr$_x$Nd$_{0.3}$CuO$_4$ with $x=0.12,0.15$ and La$_{1.4-x}$Sr$_x$Nd$_{0.6}$CuO$_4$ with $x=0.1,0.12,0.15,0.18$ superconductivity is strongly suppressed. In all these compounds we observe an additional quasielastic Gaussian below about 30

K. The width of this Gaussian is almost temperature independent and decreases with increasing Sr concentration. The observation of a Gaussian line infers a distribution of various Cu exchange fields on different Nd sites and is interpreted in terms of the stripe model. In La$_{1.8-y}$Sr$_{0.2}$Nd$_y$CuO$_4$ ($y=0.3,0.6$) no indication for a Nd-Cu interaction has been found, i.e., a single quasielastic Lorentzian is observed.

[1] S.-W. Cheong, G. Aeppli, T.E. Mason, H.A. Mook, S.M. Hayden, P.C. Canfield, Z. Fisk, K.N. Klausen, and J.L. Martinez, Phys. Rev. Lett. **67**, 1791 (1991); M. Matsuda, K. Yamada, Y. Endoh, T.R. Thurston, G. Shirane, R.J. Birgenau, M.A. Kastner, I. Tanaka, and H. Kojima, Phys. Rev. B **49**, 6958 (1994); K. Yamada, S. Wakimoto, C.H. Lee, M.A. Kastner, S. Hosaya, M. Greven, Y. Endoh, and R.J. Birgenau, Phys. Rev. Lett. **75**, 1626 (1995); K. Yamada, C.H. Lee, K. Kurahashi, J. Wada, S. Wakimoto, S. Ueki, H. Kimura, and Y. Endoh, Phys. Rev. B **57**, 6165 (1998); S. Petit, A.H. Moudden, B. Hennion, A. Vietkin, and A. Revcolevschi, Eur. Phys. J. B **3**, 163 (1998).

[2] J.M. Tranquada, B.J. Sternlieb, J.D. Axe, Y. Nakamura, and S. Uchida, Nature (London) **375**, 561 (1995); J.M. Tranquada, J.D. Axe, N. Ichikawa, Y. Nakamura, S. Uchida, and B. Nachumi, Phys. Rev. B **54**, 7489 (1996).

[3] M.K. Crawford, R.L. Harlow, E.M. McCarron, W.E. Farneth, and N. Herron, Phys. Rev. B **47**, 11 623 (1993).

[4] B. Büchner, M. Breuer, A. Freimuth, and A.P. Kampf, Phys. Rev. Lett. **73**, 1841 (1994).

[5] P. Dai, H.A. Mook, and F. Dogan, Phys. Rev. Lett. **80**, 1738 (1998).

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

[7] A.S. Ivanov, P. Bourges, D. Petitgrand, and J. Rossat-Mignod, Physica B **213 & 214**, 60 (1995).

[8] H. Casalta, P. Bourges, M. d'Astuto, D. Petitgrand, and A. Ivanov, Phys. Rev. B **57**, 471 (1998).

[9] 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).

[10] W. Henggeler, T. Chattopadhyay, B. Roessli, P. Vorderwisch, P. Thalmeier, D.I. Zhigunov, S.N. Barilo, and A. Furrer, Phys. Rev. B **55**, 1269 (1997).

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

[12] W. Henggeler, B. Roessli, A. Furrer, P. Vorderwisch, and T. Chatterji, Phys. Rev. Lett. **80**, 1300 (1998).

[13] J.T. Markert, E.A. Early, T. Bjornholm, S. Ghamaty, B.W. Lee, J.J. Neumeier, R.D. Price, C.L. Seaman, and M.B. Maple, Physica C **158**, 187 (1989).

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

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

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

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

[18] H. Drössler, H.D. Jostarndt, J. Harnischmacher, J. Kalenborn, U. Walter, A. Severing, W. Schlabitz, and E. Holland-Moritz, Z. Phys. B **100**, 1 (1996).

[19] R.E. Lechner, Neutron News **7**, 9 (1996).

[20] H. D. Jostarndt and U. Walter (unpublished).

[21] M. Breuer, B. Büchner, R. Müller, M. Cramm, O. Maldonado, A. Freimuth, B. Roden, R. Borowski, B. Heymer, and D. Wohlleben, Physica C **208**, 217 (1993).

[22] E. Holland-Moritz and G. Lander, *Handbook of the Chemistry and Physics of Rare Earths* (1994), Vol. 19, P. 1.

[23] U. Walter, S. Fahy, A. Zettl, S.G. Louie, M.L. Cohen, P. Tejedor, and A.M. Stacy, Phys. Rev. B **36**, 8899 (1987).

[24] P. Allenspach, A. Furrer, and F. Hulliger, Phys. Rev. B **39**, 2226 (1989).

[25] P. Allenspach, A. Furrer, P. Bruesch, and P. Unternaehrer, Physica B **156 & 157**, 864 (1989).

[26] K.W. Becker, P. Fulde, and J. Keller, Z. Phys. B **28**, 9 (1977).

[27] P. Fulde and M. Loewenhaupt, Adv. Phys. **34**, 589 (1986).

[28] U. Staub, L. Soderholm, S. Skanthakumar, S. Rosenkranz, C. Ritter, and W. Kagunya, Europhys. Lett. **34**, 447 (1996).

[29] A. Abragam and B. Bleaney, *Electron Paramagnetic Resonance of Transition Ions* (Clarendon, Oxford, 1970), Chap. 10.

[30] C.-K. Loong and L. Soderholm, Phys. Rev. B **48**, 14 001 (1993).

[31] We do not know if there is really a difference in the relaxation rates (and thereby in the fit parameters) of our samples. The data points at high temperatures $T>200$ K have a large uncertainty due to the fact that the signal to noise ratio is rather bad. Furthermore, the Nd content in La$_{1.7-x}$Sr$_x$Nd$_{0.3}$CuO$_4$ is low resulting in a poor statistics. Hence, the coincidence of the linewidths of La$_{1.7}$Nd$_{0.3}$CuO$_4$ and La$_{1.55}$Sr$_{0.15}$Nd$_{0.3}$CuO$_4$ at 240 K might be accidental. The most accurate results we obtained for La$_{1.25}$Sr$_{0.15}$Nd$_{0.6}$CuO$_4$.

[32] H. Shimizu, K. Fujiwara, and K. Hatada, Physica C **288**, 190 (1997).

[33] P. Allenspach, J. Mesot, U. Staub, M. Guillaume, A. Furrer, S.-I. Yoo, M.J. Kramer, R.W. McCallum, H. Maletta, H. Blank, H. Mutka, R. Osborn, M. Arai, Z. Bowden, and A.D. Taylor, Z. Phys. B **95**, 301 (1994).

[34] U. Staub, L. Soderholm, S. Skanthakumar, and M.R. Antonio, Phys. Rev. B **52**, 9736 (1995).

[35] U. Staub, F. Fauth, M. Gutmann, and W. Kagunya, Physica B **234-236**, 841 (1997).

[36] U. Staub (private communication).

[37] A.T. Boothroyd, A. Mukherjee, and A.P. Murani, Phys. Rev. Lett. **77**, 1600 (1996).

[38] J. Mesot, G. Böttger, P. Berastegui, H. Mutka, and A. Furrer, Physica C **282-287**, 1377 (1997).

[39] E.T. Heyen, R. Wegerer, and M. Cardona, Phys. Rev. Lett. **67**, 144 (1991); E.T. Heyen, R. Wegerer, E. Schönherr, and M. Cardona, Phys. Rev. B **44**, 10 195 (1991); J.A. Sanjurjo, C. Rettori, S. Oseroff, and Z. Fisk, *ibid.* **49**, 4391 (1994); R. Wegerer, C. Thomsen, T. Ruf, E. Schönherr, M. Cardona, M. Reedyk, J.S. Xue, J.E. Greedan, and A. Furrer, *ibid.* **48**, 6413 (1993).

[40] L. Kan, S. Elschner, and B. Elschner, Solid State Commun. **79**, 61 (1991).

[41] M. Roepke, E. Holland-Moritz, B. Büchner, R. Borowski, R. Kahn, R.E. Lechner, S. Longeville, and J. Fitter, J. Supercond. (to be published).

[42] M. Roepke, E. Holland-Moritz, G. Coddens, J. Fitter, and R.E. Lechner, Physica B **234-236**, 723 (1997); M. Roepke, E. Holland-Moritz, R. Müller, G. Coddens, R.E. Lechner, S. Longeville, and J. Fitter, J. Phys. Chem. Solids **59**, 2233 (1998).

[43] M. Cramm (private communication).

[44] F.C. Chou, F. Borsa, J.H. Cho, D.C. Johnston, A. Lascialfari, D.R. Torgeson, and J. Ziolo, Phys. Rev. Lett. **71**, 2323 (1993).

[45] B. Keimer, R.J. Birgenau, A. Cassanho, Y. Endoh, M. Greven, M.A. Kastner, and G. Shirane, Z. Phys. B **91**, 373 (1993).

[46] B. Büchner (private communication).

[47] J.M. Tranquada, J.D. Axe, N. Ichikawa, A.R. Moodenbaugh, Y. Nakamura, and S. Uchida, Phys. Rev. Lett. **78**, 338 (1997).

[48] W. Wagener, H.H. Klauss, M. Hillberg, M.A.C. de Melo, M. Birke, and F.J. Litterst, Phys. Rev. B **55**, 14 761 (1997).

[49] M. Breuer, B. Büchner, H. Micklitz, E. Baggio-Saitovitch, I. Souza Azevedo, R. Scorzelli, and M.M. Abd-Elmeguid, Z. Phys. B **92**, 331 (1993).

[50] W. Wagener, H.H. Klauss, M. Hillberg, M.A.C. de Melo, M. Birke, F.J. Litterst, E. Schrier, B. Büchner, and H. Micklitz, Hyperfine Interact. **105**, 107 (1997).

[51] B. Nachumi, Y. Fudamoto, A. Keren, K.M. Kojima, M. Larkin, G.M. Luke, J. Merrin, O. Tchernyshyov, Y.J. Uemura, N. Ichikawa, M. Goto, H. Takagi, S. Uchida, M.K. Crawford, E.M. McCarron, D.E. McLaughlin, and R.H. Heffner, Phys. Rev. B **58**, 8760 (1998).

[52] H. H. Klauss (private communication).