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Strong enhancement of spin fluctuations in the low-temperature tetragonal phase of antiferromagnetically ordered $La_{2-x-y}Eu_ySr_xCuO_4$

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Abstract. Measurements of the static magnetization, susceptibility and electron spin resonance (ESR) of Gd spin probes have been performed to study the properties of antiferromagnetically ordered $La_{2-x-y}Eu_ySr_xCuO_4$ ($x \leqslant 0.02$) with the low-temperature tetragonal structure. According to the static magnetic measurements, the CuO_2 planes are magnetically decoupled in this structural phase. The ESR study reveals strong magnetic fluctuations at the ESR frequency which are not present in the orthorhombic phase. It is argued that this drastic enhancement of the spin fluctuations is due to a considerable weakening of the interlayer exchange and a pronounced influence of hole motion on the antiferromagnetic properties of lightly hole-doped La_2CuO_4. No evidence for the stripe phase formation at small hole doping is obtained in the present study.

The structural phase transition from the low-temperature orthorhombic (LTO) phase to the low-temperature tetragonal (LTT) phase (hereafter the LT transition) in the rare-earth- (RE-) doped $La_{2-x}Sr_xCuO_4$ high-temperature superconductors [1, 2] is known to influence strongly the electronic properties of these compounds at elevated levels of Sr (i.e. hole) doping. It is found that the buckling of the CuO_6 octahedra controls the occurrence of superconductivity [3] and leads to a static order of spins and charges in the form of stripes which has been observed in Nd-doped crystals [4, 5]. In Eu-doped $La_{2-x}Sr_xCuO_4$ anomalous slowing of the spin dynamics [6] and reappearance of magnetic order at low temperatures [7] for $x \geqslant 0.08$ have been found in ESR and μSR experiments, respectively.

While spatially modulated spin correlations in $La_{2-x}Sr_xCuO_4$ are well established at elevated hole concentrations, the situation is controversial for the low-hole-doping regime. Neutron scattering experiments do not reveal incommensurate spin correlations for $x \leqslant 0.05$ [8]. However, static magnetic measurements and results from several local resonance techniques evidence the formation of hole-free antiferromagnetic (AF) domains separated by charged domain walls [9–12]. The properties of the ordered Néel state of the stripe phase in the doped cuprates are intensively discussed theoretically [13, 14]. Predictions of the stripe model agree well with the observed suppression of the ordering temperature $T_N(x)$ with hole doping [13]. The effects of doped holes on the AF ordered La_2CuO_4 with LTO structure have been thoroughly studied by means of NQR [10–12]. The experiments reveal magnetic

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fluctuations in the ordered state at small hole doping which freeze out at low T. It is argued that these fluctuations result from a collective charge (stripe) motion in the planes thus giving evidence for a stripe phase formation at very low hole doping [11, 12]. The relevance of this scenario to the underdoped La_2CuO_4 can be verified by causing a structural phase transition to the LTT phase by RE doping [1, 2]. One may anticipate a diminished influence of holes on the spin dynamics in the LTT phase because of the pinning of charge stripes which is known to occur in compounds with large hole concentration [4]. However, this expectation is not met in the recent NQR experiments on $La_{2-x-y}Eu_ySr_xCuO_4$ which evidence a more dynamic behaviour of Cu spins in the LTT phase [15].

To obtain an insight into the stripe phase formation and to study the interplay between structure and magnetism in lightly hole-doped lanthanum copper oxide we have studied the magnetic properties of Eu-doped $La_{2-x}Sr_xCuO_4$ at low Sr concentrations by means of the static susceptibility, magnetization and electron spin resonance (ESR) of Gd spin probes. We find striking changes of the magnetism due to the LT transition:

(i) the Gd^{3+} ESR spectrum, which exhibits a partially resolved fine structure in both the paramagnetic and the magnetically ordered state in the LTO phase, collapses into a single narrow line for $x > 0$ and $T < T_{LT}$, signalling strong fluctuating magnetic fields in the hole-doped samples at the ESR frequency;

(ii) a spin-flip transition in the magnetization curves $M(H)$ clearly visible at $T > T_{LT}$ is strongly reduced for $T < T_{LT}$ suggesting a considerable decrease of the interlayer coupling in the LTT phase.

We argue that the strong enhancement of spin fluctuations in the hole-doped samples is related to the magnetic decoupling of the CuO_2 planes with LTT structure. Our data give no indication as to the relevance of charge stripes at very low hole concentration.

Polycrystalline $La_{2-x-y}Eu_ySr_xCuO_4$ samples with $0 \leqslant x \leqslant 0.03$ and $0 \leqslant y \leqslant 0.20$ were prepared as described elsewhere [16]. To remove the excess oxygen the samples were annealed in N_2 atmosphere at 625 °C for three days. The samples for ESR measurements were additionally doped with 0.5% of Gd ions. The ESR experiments were carried out at 9.3 GHz. The static magnetic susceptibility $\chi(T)$ was measured using a Faraday balance in a magnetic field of 10 kOe. Measurements of the magnetization $M(H, T)$ in fields up to 140 kOe were performed with a vibrating-sample magnetometer. The LT transition temperatures $T_{LT} \sim 120$–140 K were verified by x-ray diffraction.

In figure 1 we show a set of Gd^{3+} ESR spectra (field derivatives of the absorption) of $La_{1.99-x-y}Sr_xGd_{0.01}Eu_yCuO_4$ samples. We find that in the paramagnetic state, i.e. above the ordering temperature T_N, the spectra of all samples are structured and most of the weight of the spectra is below 2000 Oe (see e.g. spectra (1) in figures 1(a) and 1(b)). At low T the shapes of the spectra change. For the samples without Sr this concerns both the number and the positions of the peaks. Nevertheless, the spectra remain structured for both Eu-free and Eu-doped cases (figure 1(a), spectra (2) and (3)). However, doping of La_2CuO_4 with *both* Eu and Sr leads to a qualitatively new and striking effect: the ESR spectrum narrows into a single line at $T < T_{LT}$ with a width less than the extent of the resolved spectrum. Moreover, the line shifts considerably to higher fields. We illustrate this in figure 1(b) which shows a representative set of spectra for the sample with $x = 0.008$, $y = 0.20$. For $T > T_{LT}$ the resonance curves (figure 1(b), spectra (1) and (2)) resemble the respective spectra for the Sr-doped samples without Eu. However, below the LT transition the spectrum acquires a shape close to a single Lorentzian line (see figure 1(b), spectrum (3)). Its spectral weight shifts to much higher fields ($\gtrsim$3000 Oe). The peak-to-peak width of this line becomes smaller than the total extent of the structured spectra. Eventually at low temperatures the structure in the spectrum of the Eu-doped sample partially

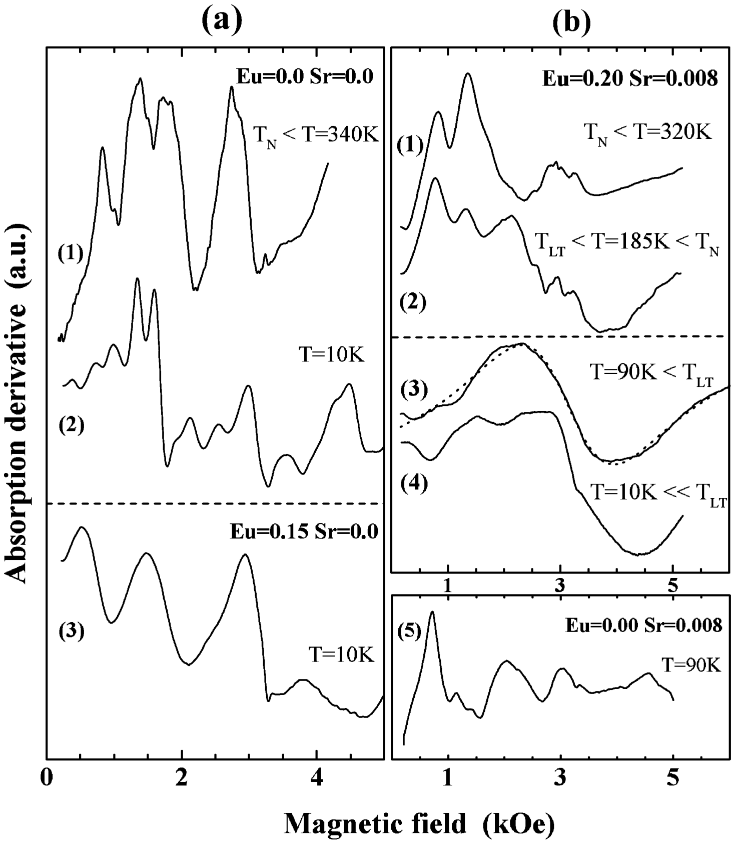

Figure 1. (a) Gd^{3+} ESR spectra of insulating $La_{1.99-y}Eu_yGd_{0.01}CuO_4$. Curves (1) and (2) correspond to the sample without Eu in the paramagnetic state and in the AF state at 10 K. Curve (3) at 10 K corresponds to the sample doped with Eu ($y = 0.15$). (b) Gd^{3+} ESR spectra of $La_{1.99-x-y}Sr_xEu_yGd_{0.01}CuO_4$. Curves (1)–(4) show the evolution of the spectrum of the sample with $x = 0.008$ and $y = 0.20$ with decreasing temperature. Note the collapse of the spectrum into a nearly single line below the LT transition. The dotted line represents a Lorentzian fit. In the bottom panel a respective spectrum (5) of the sample without Eu is shown for comparison.

reappears and the centre of weight of the spectrum shifts back to lower fields (figure 1(b), spectrum (4)). With increasing Sr content in $La_{2-x-y}Eu_ySr_xCuO_4$ the collapse of a structured ESR spectrum into a single line occurs at progressively lower temperatures. The width of the line gets smaller. The shaded areas in the T–x phase diagram in figure 2 map the regions where the collapsed ESR spectrum is observed. We note that the collapse of the ESR spectrum does not occur for the samples with the LTO structure (with and without Eu) throughout the whole temperature range studied (8 K $\leqslant T \leqslant$ 300 K). In particular, a respective spectrum for a hole-doped sample without Eu (i.e. with the LTO structure) shows a well resolved structure (figure 1(b), spectrum (5)).

A free Gd^{3+} ion has a pure spin ground state ($S = 7/2$) with eightfold degeneracy. This degeneracy is lifted in a magnetic field which allows ESR transitions between the states $|S_z\rangle$ and $|S_z \pm 1\rangle$. All of them require the same energy quantum $g_{spin}\mu_B H_{res} = h\nu$. Therefore a single absorption line is expected at the field $H_{res} \approx$ 3300 Oe for $\nu \approx$ 9.3 GHz and a g-factor $g_{spin} \simeq 2$. However, the crystalline electrical field (CEF) causes a small initial splitting of Gd^{3+}

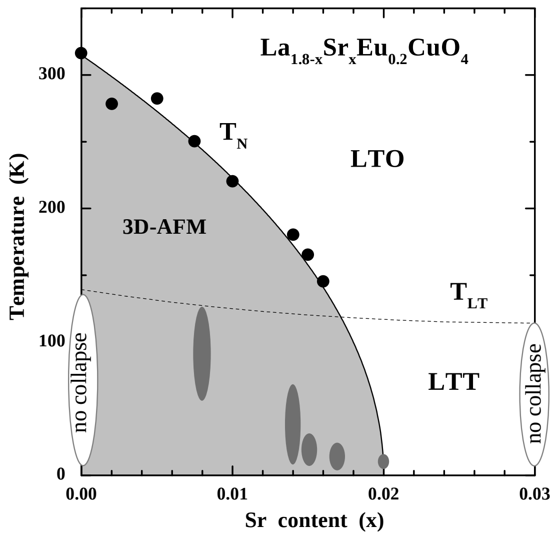

Figure 2. The structural (T_{LT}) and magnetic (T_N) phase diagram of $La_{1.8-x}Sr_xEu_{0.2}CuO_4$. The $T_{LT}(x)$ phase boundary is obtained by x-ray diffraction (dotted line). Solid circles are the T_N-values obtained from the susceptibility data. The solid line shows the $T_N(x)$ dependence for $La_{2-x}Sr_xCuO_4$. The shaded ellipses are the regions where the Gd^{3+} ESR spectra of the samples with $x = 0.008, 0.014, 0.015, 0.017$ and 0.020 collapse into a single line. The collapse is not observed for the other two Eu-doped samples studied in this work ($x = 0.0$ and 0.03), or for any samples without Eu.

energy levels. As a result, resonance transitions between different levels occur at different H and the respective spectrum consists of more than one line (the so-called fine-structure (FS) ESR spectrum) [17]. For La_2CuO_4 the Gd^{3+} ESR spectrum is rather complex [18,19]. It comprises a set of absorption lines which partially overlap. The spectrum is highly asymmetric and its weight is considerably shifted from $H \approx 3300$ Oe to fields lower than 2000 Oe.

Gd^{3+} ESR studies of single crystals of $La_{2-x}Sr_xCuO_4$ ($x \leqslant 0.024$) show that in the AF state the spectrum additionally splits due to a static internal field [19]. At $T \ll T_N$ the components of the FS spectrum are well resolved only in the insulating case. The presence of holes broadens the spectrum; nevertheless it remains structured [19]. Qualitatively similar features are observed in our experiments too. Also doping of insulating La_2CuO_4 with Eu does not change the behaviour qualitatively. It causes only an additional broadening and partial overlapping of individual components of the FS spectra (figure 1, spectrum (3)) due to distortions of the CEF and due to a spatially inhomogeneous Eu^{3+} Van Vleck magnetization. Thus one can conclude that the striking collapse of the structured Gd^{3+} ESR spectrum observed for $La_{1.99-x-y}Sr_xGd_{0.01}Eu_yCuO_4$ is due to both the structural LT transition induced by Eu doping *and* the presence of holes in the CuO_2 planes.

The narrowing of the Gd^{3+} ESR spectrum into a single line resembles the effect of exchange narrowing in magnetic resonance. This similarity strongly suggests a *dynamic* cause for the collapse of the Gd^{3+} ESR spectrum. In our case the Gd spins are highly diluted in the lattice and therefore Gd–Gd exchange is apparently too small to be responsible for the narrowing of the spectrum. However, studies of ESR in conventional metals give evidence for a *relaxation* narrowing of the FS spectrum, which is a single-ion process [20]. If the rate of a certain ESR transition between FS split levels of a Gd^{3+} ion is strong enough to couple it with the neighbouring transitions, then eventually the FS spectrum may collapse into a single narrow line. In the case of lightly Sr-doped $La_{1.99-x-y}Sr_xGd_{0.01}Eu_yCuO_4$ the only thinkable source of strong Gd spin relaxation is a fluctuating field produced by copper spins. Therefore, from the ESR data we conclude that the LT transition results in strong spin fluctuations in the CuO_2 planes with frequencies of the order of the ESR frequency for some $x > 0$ and T (see figure 2).

To understand the drastic changes of the spin dynamics in the CuO_2 planes due to the LT transition we have measured the static magnetic properties of $La_{2-x-y}Eu_ySr_xCuO_4$. Representative susceptibility curves $\chi(T)$ for the samples with $x = 0, 0.010(1)$ and $y = 0, 0.2$ are shown in figures 3(a) and 3(b). Note that for the Eu-doped samples a Van Vleck contribution of Eu^{3+} ions has been subtracted from the raw susceptibility and magnetization data [21]. Furthermore, the data have been corrected for the core diamagnetism ($\chi_{core} = -0.99 \times 10^{-4}$ emu mol^{-1}) and the Van Vleck paramagnetism of the copper ions ($\chi_{VV} = 0.43 \times 10^{-4}$ emu mol^{-1}) [22] . The $\chi(T)$ curves for $y = 0$ and 0.2 show a very similar behaviour for $T > T_{LT}$. There is a well defined Néel peak at the AF ordering temperature T_N whose position shifts to lower T with increasing Sr concentration x. The ordering temperatures as a function of x are plotted in figure 2. It is apparent that T_N for Eu-doped samples follows the same dependence on x as is observed for $La_{2-x}Sr_xCuO_4$ (the solid line in figure 2). Below T_{LT} the Néel peak is not seen for the Eu-doped compounds with $T_N < T_{LT}$. However, according to recent μSR measurements [7] the T_N-values of these samples are similar to those of the respective samples with the LTO structure as well. Remarkably, the susceptibility of the Eu-doped samples exhibits a pronounced step-like increase which coincides with the LT transition ($T_{LT} \approx 120$–140 K). The amplitude of the step is almost the same for all samples with $x \leqslant 0.016$. For higher values of x the anomaly rapidly decreases and it vanishes at $x = 0.020$.

In figure 3(c) we show representative magnetization curves $M(H)$ of Eu-doped La_2CuO_4 acquired at temperatures slightly above and below the LT transition. At $T > T_{LT}$ a step-like change of $M(H)$ at $H_c \sim 40$ kOe is seen similar to that observed for pure La_2CuO_4 crystals [23]. Remarkably, below T_{LT} the step-like change of $M(H)$ is strongly reduced. Similar changes of $M(H)$ due to the LT transition are apparent also for Sr-doped samples with $T_N > T_{LT}$.

In the CuO_2 planes with the LTO structure the spins are slightly canted out of the planes because of the tilting of the CuO_6 octahedra and the Dzyaloshinsky–Moriya (DM) anti-symmetric anisotropic exchange. Therefore a net out-of-plane ferromagnetic moment (DM moment) arises in every layer [23]. In the LTT structure the tilting of the Cu–O–Cu bonds is still present in one crystallographic direction [1, 2] and allows for the DM interaction too [24]. In principle, two kinds of spin arrangements are possible in the latter structure: one with and another without the DM moments [24]. Which of these two configurations should be the ground state of the LTT phase of La_2CuO_4 is still a question under intense theoretical discussion (see e.g. [25–27]). In the following we show that our static magnetic data give strong evidence that the DM moments are present in the LTT phase of Eu-doped $La_{2-x}Sr_xCuO_4$.

It is well established that in the LTO phase, below T_N, the DM moments are ordered antiferromagnetically due to the interplane coupling of the Cu spins [23]. Because the DM moments produce an additional contribution χ_{DM} to the total susceptibility, the measured χ in the LTO phase is significantly larger than that expected for an isotropic two-dimensional $S = 1/2$ Heisenberg antiferromagnet χ_{2DHAF} (see the respective curves in figure 3 and reference [28]). Our experimental data clearly show that at the LT structural transition the difference between χ_{2DHAF} and the measured susceptibility of $La_{2-x-y}Eu_ySr_xCuO_4$ at 10 kOe *even increases* (see figures 3(a), 3(b)), thus suggesting that the DM moments are present also in the LTT phase. The same conclusion emerges from the analysis of the field dependence of the magnetization $M(H)$. As one can see in figure 3(c), at all fields it is larger than that of a 2DHAF

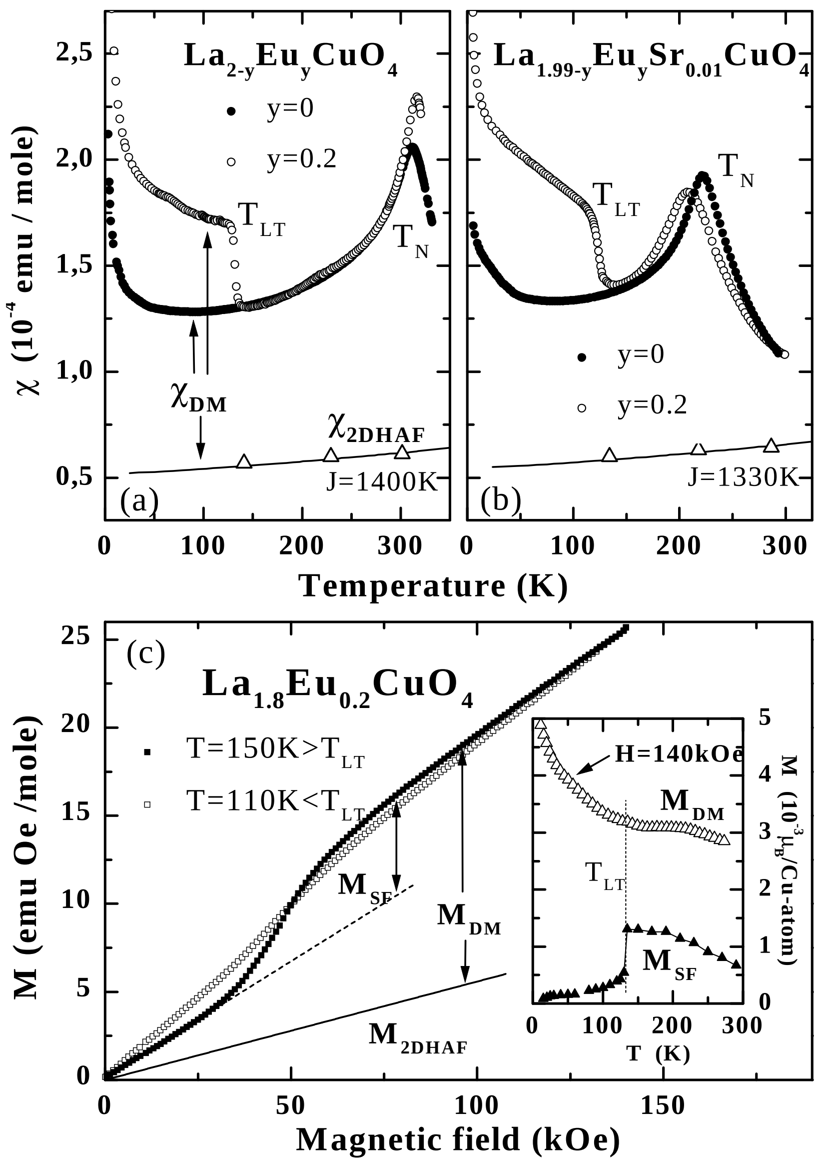

Figure 3. The static magnetic susceptibility $\chi(T)$ at $H = 10$ kOe (a), (b) and the magnetization $M(H)$ (c) of $La_{2-x-y}Eu_ySr_xCuO_4$. Solid lines and triangles are predictions for the $S = 1/2$ 2DHAF from the Schwinger boson mean-field theory [29] and Monte Carlo simulation [30], respectively, with an in-plane exchange $J \sim 1400$ K. χ_{DM} and M_{DM} denote contributions due to DM exchange. M_{SF} is the spin-flip magnetization. The inset shows the T-dependences of M_{DM} at $H = 140$ kOe and of M_{SF}. (For details see the text.)

by the amount M_{DM} which is the out-of-plane magnetization due to the DM moments. The main feature of the magnetization curves $M(H)$ for $T > T_{LT}$, the step-like increase by M_{SF}, is related to the spin flip (SF) of these moments at the critical field H_c [23]. Above H_c, due to the spin flip, M_{DM} makes the dominant contribution to the measured $M(H)$. Remarkably, the value of M_{SF} which measures the staggered part of the DM magnetization, drops down at the LT transition (see the inset of figure 3(c)), and the critical field H_c is not defined for the LTT phase [31]. Nevertheless, at fields $H > 100$ kOe, $M(H)$ curves measured slightly above and below the LT transition come very close together, i.e. the high-field magnetization which is mostly due to the DM exchange obviously does not change at the LT transition. This is also evident from the T-dependence of M_{DM} measured at $H = 140$ kOe (see the inset of figure 3(c)). Thus, one may conclude that CuO_2 planes with the LTT structure carry the same net DM moments as in the LTO phase. Hence, the strong reduction of the spin-flip transition evidences the considerable weakening of the interlayer exchange in the LTT phase which is responsible for the AF arrangement of the DM moments.

The reason for such a dramatic effect of the structural transition on the magnetic coupling of the CuO_2 planes has been suggested in reference [32] where neutron diffraction (ND) studies of $La_{2-y}Nd_yCuO_4$ crystals with the LTT structure are presented. For the LTT phase these studies reveal spin structures with a weakened and frustrated interlayer exchange which leads to the coexistence of magnetic domains with effective AF and ferromagnetic interlayer coupling. In accordance with the ND results our static magnetic measurements evidence that the magnetic decoupling of the CuO_2 planes due to such a frustration takes place in insulating as well as in hole-doped $La_{2-y}Eu_yCuO_4$ and results macroscopically in the absence of the critical spin-flip field and a considerable increase of the susceptibility and magnetization at low fields.

It is straightforward to relate a strong decoupling of the layers due to the LT transition with the strong magnetic fluctuations observed in the hole-doped $La_{2-x-y}Eu_ySr_xCuO_4$ samples at ESR frequencies. Magnetic fluctuations in AF ordered *hole-doped* La_2CuO_4 with the LTO structure were observed by NQR in the MHz-frequency window [10–12]. Our ESR data suggest the presence of strong magnetic fluctuations in hole-doped $La_{2-x-y}Eu_ySr_xCuO_4$ with the LTT structure on a much higher frequency scale. A possible reason for such a drastic enhancement of the magnetic dynamics is an effective reduction of the dimensionality of the AF correlations in the CuO_2 planes due to interlayer decoupling. As a result the magnetic correlations should be less stable against perturbations produced by moving holes. Consequently the frequency of the spin fluctuations may be considerably enhanced compared to that for the LTO phase. In a certain temperature range $\Delta T < T_{LT}$ it may approach the ESR frequency window and thus provide a channel for a strong spin relaxation of the Gd probe. Thus our data strongly suggest that the magnetic dynamics due to the charge motion is not inhibited in the LTT structure at small hole doping.

The NQR data on the LTO $La_{2-x}Sr_xCuO_4$ samples were explained assuming that holes form stripes or loops in the layers [11, 12]. It is argued that the motion of these charge objects serving as antiphase boundaries for the hole-free magnetic domains causes flips of the Cu spins. This motion freezes out at low T leading to a recovery of the sublattice magnetization and to a peak in the T-dependence of the ^{139}La spin–lattice relaxation rate $(1/T_1)^{La}$ at $T_f \sim 10$–16 K [10–12]. In this scenario, changing the structure from LTO to LTT should lead to weakening of magnetic fluctuations in the AF ordered CuO_2 planes since charge stripes are pinned in the LTT phase [4].

This expectation is in striking contrast with our experimental observations. For a qualitative understanding of our data not even an assumption of collective charge motion is necessary. In particular, we find no evidence for a pinning of charge stripes which is known to exist in the LTT phase at higher levels of hole doping. Remarkably, in a recent ^{139}La NQR study of lightly Sr-doped $La_{2-x-y}Eu_ySr_xCuO_4$ ($x = 0.010, 0.015$) [15] the peak in $(1/T_1)^{La}$ was found to occur at a considerably lower temperature ($T_f \sim 6$ K) than in compounds with LTO structure [10–12]. As this peak signals freezing of the spin fluctuations on the NQR timescale, its shift to lower T is also suggestive of a more dynamic magnetic behaviour of the

LTT phase. Therefore for a better understanding of the spin dynamics it would be desirable to perform a systematic NQR study of lightly hole-doped $La_{2-x-y}Eu_ySr_xCuO_4$.

In summary, we have measured the susceptibility, magnetization and ESR of Gd spin probes in order to study the magnetic properties of the LTT phase of $La_{2-x-y}Eu_ySr_xCuO_4$ with small hole doping $x \leqslant 0.03$. The static magnetic measurements evidence a drastic reduction of the interlayer coupling in the LTT phase. In the ESR measurements of the hole-doped samples we observe a collapse of the Gd spectrum into a single narrow line at $T < T_{LT}$ which signals strong magnetic fluctuations at the ESR frequency. We conclude from the data that the distinct differences in the properties of the AF ordered LTT phase of $La_{2-x-y}Eu_ySr_xCuO_4$ at low levels of hole doping arise due to the effective interlayer magnetic decoupling, which enhances the influence of mobile holes on the antiferromagnetism of this compound. In particular, the data give no evidence for charge stripes at small hole concentrations.

Acknowledgments

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References

[1] Büchner B *et al* 1991 *Physica* C **185–189** 903
[2] Crawford M K, Harlow R L, McCarron E M, Farneth W E, Axe J D, Chou H and Huang Q 1991 *Phys. Rev.* B **44** 7749
[3] Büchner B, Breuer M, Freimuth A and Kampf A P 1994 *Phys. Rev. Lett.* **73** 1841
[4] Tranquada J M, Axe J D, Ichikawa N, Moodenbaugh A R, Nakamura Y and Uchida S 1997 *Phys. Rev. Lett.* **78** 338 and references therein
[5] von Zimmermann M *et al* 1998 *Europhys. Lett.* **41** 629
[6] Kataev V, Rameev B, Büchner B, Hücker M and Borowski R 1997 *Phys. Rev.* B **55** R3394
Kataev V, Rameev B, Validov A, Büchner B, Hücker M and Borowski R 1998 *Phys. Rev.* B **58** R11 876
[7] Wagener W, Klauß H-H, Hillberg M, Kopmann W, Walf H, Litterst F J, Hücker M and Büchner B 1999 to be published
[8] Yamada K *et al* 1998 *Phys. Rev.* B **57** 6165
[9] Cho J H, Chou F C and Johnston D C 1993 *Phys. Rev. Lett.* **70** 222
[10] Chou F C, Borsa F, Cho J H, Johnston D C, Lascialfari A, Torgeson D R and Ziolo J 1993 *Phys. Rev. Lett.* **71** 2323
[11] Borsa F *et al* 1995 *Phys. Rev.* B **52** 7334
[12] Suh B J, Hammel P C, Yoshinari Y, Thompson J D, Sarrao J L and Fisk Z 1998 *Phys. Rev. Lett.* **81** 2791
[13] Castro Neto A H and Hone D 1996 *Phys. Rev. Lett.* **716** 2165
Stojković B P, Yu Z G, Bishop A R, Castro Neto A H and Gronbech-Jensen N 1998 *e-print* cond-mat/9805367
[14] van Duin C N A and Zaanen J 1998 *Phys. Rev. Lett.* **80** 1513
[15] Suh B J, Hammel P C, Hücker and Büchner B 1999 *Phys. Rev.* B **59** R3952
[16] Breuer M *et al* 1993 *Physica* C **208** 217
[17] Abragam A and Bleaney B 1970 *Electron Paramagnetic Resonance of Transition Ions* (Oxford: Clarendon)
[18] Kataev V, Greznev Yu, Kukovitskiĭ E F, Teitel'baum G, Breuer M and Knauf N 1992 *JETP Lett.* **56** 385
Kataev V, Greznev Yu, Teitel'baum G, Breuer M and Knauf N 1993 *Phys. Rev.* B **48** 13 042
[19] Rettori C *et al* 1993 *Phys. Rev.* B **47** 8156
[20] Barnes S E 1981 *Adv. Phys.* **30** 801 and references therein
[21] Hücker M, Pommer J, Büchner B, Kataev V and Rameev B 1997 *J. Supercond.* **10** 451
[22] Allgeier C and Schilling J S 1993 *Phys. Rev.* B **48** 9747
[23] Thio T *et al* 1988 *Phys. Rev.* B **38** 905
Kastner M A *et al* 1988 *Phys. Rev.* B **38** 6636
[24] Coffey D, Rice T M and Zhang F C 1991 *Phys. Rev.* B **44** 10 112

[25] Koshibae W, Ohta Y and Maekawa S 1994 *Phys. Rev.* B **50** 3767
[26] Viertiö H E and Bonesteel N E 1994 *Phys. Rev.* B **49** 6088
[27] Stein J, Entin-Wohlman O and Aharony A 1996 *Phys. Rev.* B **53** 775
[28] Johnston D C 1991 *J. Magn. Magn. Mater.* **100** 218
[29] Auerbach A and Arovas D P 1988 *Phys. Rev. Lett.* **61** 617
[30] Okabe Y, Kikuchi M and Nagi A D S 1988 *Phys. Rev. Lett.* **61** 2971
[31] We mention here that the small amount of M_{SF} still visible in the $M(H)$ curves below T_{LT} rapidly decreases with temperature. We attribute this to the rest of the LTO phase whose volume fraction, according to x-ray diffraction, shows a similar temperature dependence.
[32] Shamoto S, Kiyokura T, Sato M, Kakurai K, Nakamura Y and Uchida S 1992 *Physica* C **203** 7
Keimer B, Birgeneau R J, Cassanho A, Endoh Y, Greven M, Kastner M A and Shirane G 1993 *Z. Phys.* B **91** 373
Crawford M K, Harlow R L, McCarron E M, Farneth W E, Herron N, Chou H and Cox D E 1993 *Phys. Rev.* B **47** 11 623