Spin gap in the single spin-$\frac{1}{2}$ chain cuprate $Sr_{1.9}Ca_{0.1}CuO_3$

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We report ^{63}Cu nuclear magnetic resonance and muon spin rotation measurements on the $S = 1/2$ antiferromagnetic Heisenberg spin chain compound $Sr_{1.9}Ca_{0.1}CuO_3$. An exponentially decreasing spin-lattice relaxation rate T_1^{-1} indicates the opening of a spin gap. This behavior is very similar to what has been observed for the cognate zigzag spin chain compound $Sr_{0.9}Ca_{0.1}CuO_2$, and it confirms that the occurrence of a spin gap upon Ca doping is independent of the interchain exchange coupling J'. Our results therefore suggest that the appearance of a spin gap in an antiferromagnetic Heisenberg spin chain is induced by a local bond disorder of the intrachain exchange coupling J. A low-temperature upturn of T_1^{-1} evidences growing magnetic correlations. However, zero-field muon spin rotation measurements down to 1.5 K confirm the absence of magnetic order in this compound, which is most likely suppressed by the opening of the spin gap.

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I. INTRODUCTION

Quantum effects in low-dimensional magnets lead to a diversity of novel ground states, thereby attracting much experimental and theoretical attention in order to understand the impact of these effects on the properties of low-dimensional systems. Among others, the integrable one-dimensional (1D) antiferromagnetic $S = 1/2$ Heisenberg spin chain is a particularly interesting model system. It is characterized by a quantum critical ground state lacking any long-range order and by a gapless spectrum of elementary $S = 1/2$ excitations (spinons). Although it has been known for more than 80 years, it is still a very interesting model system from both a theoretical and an experimental point of view. This was demonstrated by a recent report on highly precise measurements of the spinon excitation spectrum of a 1D $S = 1/2$ antiferromagnetic Heisenberg chain material by means of inelastic neutron scattering [1]. The chain cuprates Sr_2CuO_3 and $SrCuO_2$ are very good realizations of the 1D antiferromagnetic $S = 1/2$ Heisenberg model. They exhibit static magnetism only at low temperatures, namely at $T_N = 5.4$ and 2 K, respectively [2–4]. For $SrCuO_2$, inelastic neutron scattering (INS) measurements reported the short-ranged nature of the static magnetism below T_N and the gapless spinon excitation spectrum down to the lowest measured energy transfer of 0.5 meV [5]. The ^{63}Cu nuclear magnetic resonance (NMR) spin-lattice relaxation rates T_1^{-1} of both compounds display the theoretically predicted temperature-independent behavior of an $S = 1/2$ antiferromagnetic Heisenberg chain [6–9]. Spin-charge separation, which is an essential feature of the 1D Hubbard model [10], was already experimentally observed 15 years ago for both compounds [11,12]. Recently, a spin-orbital separation was reported for Sr_2CuO_3 by means of resonant inelastic x-ray spectroscopy (RIXS) [13].

Apart from the experimental evidence for the good one-dimensionality of these compounds, up to now only a few investigations focused on the effects of disorder introduced by impurities in these systems. However, such studies can reveal important information. For instance, by increasing the purity of the primary chemicals for the crystal growth, a very large spinon heat conductivity could be observed in highly pure $SrCuO_2$, indicating ballistic transport, which is a consequence of the integrability of the Heisenberg model [14]. Direct magnetic (Ni) and nonmagnetic (Pd) impurity doping on the Cu sites within the spin-chain structure of Sr_2CuO_3 led to an expected increase of the Curie contribution in the magnetic susceptibility and to the theoretically predicted reduction of T_N [15,16]. For $SrCuO_2$ it has been shown that the increase of the Curie contribution is the same for magnetic (Ni) and nonmagnetic (Zn, Ga) dopants, suggesting that its origin is the general chain break effect of the impurities, while Ni remains in a low spin ($S = 0$) state [17]. However, the observation of only a slight increase of the Curie contribution upon Ni doping could also be linked to the appearance of a pseudo spin gap in the spinon excitation spectrum of $SrCu_{0.99}Ni_{0.01}O_2$, as recently reported by inelastic neutron scattering [18]. Unexpected strong effects have also been observed in $Sr_{0.9}Ca_{0.1}CuO_2$. The introduction of nonmagnetic Ca dopants on the Sr sites outside the zigzag chain structures drastically suppressed the spinon heat transport [19] and led to an exponential decrease of the ^{63}Cu NMR spin-lattice relaxation rate T_1^{-1} below 100 K, pointing toward the opening of a spin gap [9]. Density-matrix renormalization-group (DMRG) calculations suggested that Ca induces a weak bond disorder in the spin chains leading to a small alternation δ of the intrachain exchange coupling J along the chains, which causes the opening of the spin gap Δ [9]. These calculations also showed that the value of Δ does not depend on the strength and the sign of the interchain coupling J', but rather on the strength of the alternation δ of the intrachain coupling J.

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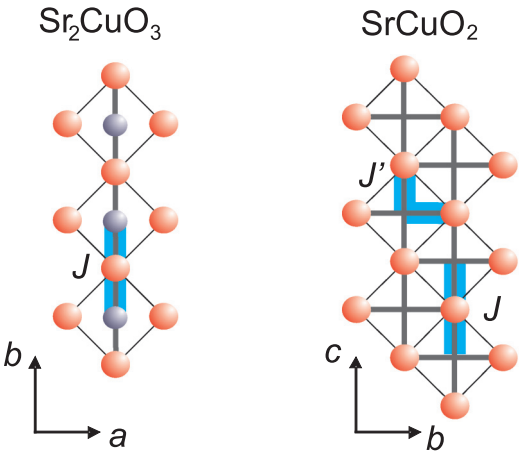

FIG. 1. (Color online) Chain structure of Sr_2CuO_3 and double-chain structure of $SrCuO_2$ including the main exchange couplings J (intrachain) and J' (interchain). Gray spheres represent the copper ions, red spheres represent the oxygen ions.

Here, we report measurements of the ^{63}Cu NMR spin-lattice relaxation rate T_1^{-1} on the single chain compound $Sr_{1.9}Ca_{0.1}CuO_3$, where we observe a very similar decrease of the ^{63}Cu NMR T_1^{-1}, confirming that the interchain coupling constant J' is indeed negligible. Additionally, we performed zero-field (ZF) muon spin rotation (μSR) measurements to search for static magnetism. Down to 1.5 K, we did not observe any evidence for the onset of a magnetic order in $Sr_{1.9}Ca_{0.1}CuO_3$.

II. SAMPLE DETAILS, SAMPLE PREPARATION, AND EXPERIMENTAL DETAILS

In contrast to the zigzag spin chains of $SrCuO_2$, the crystal structure of Sr_2CuO_3 contains only single chains of Cu^{2+} ($3d^9$, $S = 1/2$) ions, running along the crystallographic b direction (see Fig. 1). The nearest-neighbor (NN) intrachain exchange interaction between adjacent Cu moments is antiferromagnetic and its coupling constant amounts to $J \approx (2200 \pm 200)$ K, which is of the same order as in the zigzag chain compound $SrCuO_2$ [20]. The only difference between the two compounds is the existence of a weak, ferromagnetic interchain coupling with $|J'/J| \approx 0.1$–0.2 in $SrCuO_2$ [21], which, due to the linear arrangement of Cu ions, is absent in Sr_2CuO_3.

Single crystals of $Sr_{1.9}Ca_{0.1}CuO_3$ were prepared using the traveling-solvent floating zone technique [22]. The crystals cleave readily along $(h,0,0)$ and were verified to be free from secondary phases by x-ray diffraction measurements. The stoichiometry was confirmed using EDX and the crystals were oriented using the Laue x-ray backscattering method. Several previous refinement studies confirm that Ca indeed occupies the Sr sites in the $(Sr_{1-x}Ca_x)_2CuO_3$ series [23,24].

^{63}Cu NMR spectra have been measured with the standard Hahn spin echo method at a constant frequency of 75.742 MHz by sweeping the magnetic field parallel to the crystallographic b axis. Since ^{63}Cu has a nuclear spin $I = 3/2$, its NMR spectrum consists of three quadrupolar split lines, where the central line is affected by first-order magnetic hyperfine and second-order quadrupolar interactions, while the two satellites are subjects to first-order magnetic hyperfine and first-order quadrupolar effects [25]. ^{63}Cu NMR ($I = 3/2$) spin-lattice relaxation rate measurements were carried out on the central line of the quadrupolar split ^{63}Cu NMR spectrum (corresponding to the transition $I_z = -1/2 \leftrightarrow I_z = +1/2$) in a slightly higher magnetic field of $\mu_0 H = 7.0493$ T, with H being parallel to the crystallographic a and b axes. The inversion recovery method was used to measure T_1^{-1}. The relaxation function for a magnetic relaxation of an $I = 3/2$ nuclear spin system measured at the central transition is given by [26]

$$M_z(t) = M_0\left[1 - f\left(0.9e^{-(6t/T_1)^{\lambda}} + 0.1e^{-(t/T_1)^{\lambda}}\right)\right], \quad (1)$$

where M_0 is the saturation value of the nuclear magnetization, the parameter f is ideally 2 for a complete inversion, and $\lambda < 1$ accounts for a distribution of spin-lattice relaxation times around the characteristic value T_1^{-1}. At high temperatures, $\lambda \approx 1$ confirms a well-defined spin-lattice relaxation rate T_1^{-1}. At lower temperatures, λ starts to decrease, indicating a distribution of T_1^{-1} (see the inset of Fig. 2).

Zero-field (ZF) μSR measurements down to 1.5 K have been performed at the GPS instrument of the πM3 beamline at Paul Scherrer Institute (PSI) in Villigen (CH). To reduce the background, two single crystals of dimensions $5 \times 4 \times 1$ mm^3 each were used, aligned with their crystallographic c axis parallel to the incident muon beam.

III. EXPERIMENTAL RESULTS AND DISCUSSION

Figure 2 shows the temperature dependence of T_1^{-1} for $Sr_{1.9}Ca_{0.1}CuO_3$ in comparison to the data of the zigzag chain compound $Sr_{0.9}Ca_{0.1}CuO_2$ [9]. At high temperatures, both compounds display an almost constant T_1^{-1}, as it is theoretically expected for 1D antiferromagnetic $S = 1/2$ Heisenberg chains [6,7] and has been reported for the undoped parent compounds [8,9]. Below $T \approx 90$ K, T_1^{-1} of $Sr_{1.9}Ca_{0.1}CuO_3$ decreases exponentially. This decrease is identical to the one observed in the Ca-doped zigzag chain compound $Sr_{0.9}Ca_{0.1}CuO_2$ and points toward the opening of a spin gap of the same order of magnitude ($\Delta = 50$ K) as in $Sr_{0.9}Ca_{0.1}CuO_2$ [9].

On the one hand, this result indicates that the double chain structure of $SrCuO_2$ and the associated interchain coupling constant J' are not necessary prerequisites for the appearance of a gap in the spin-excitation spectrum of the spin chains. This has also been reported by our previous DMRG calculations, which showed that the value of the spin gap does not depend either on the value or on the sign of the interchain coupling J' [9]. On the other hand, these DMRG calculations also showed that the spin gap is a consequence of a subtle bond disorder induced by the Ca doping, and that the value of the spin gap Δ depends sensitively on the value of the alternation δ of the intrachain coupling J. In this regard, the perfect agreement between the two data sets pointing toward spin gaps of equal sizes is surprising, since δ should depend delicately on the structural background (e.g., elastic constants), which is supposed to differ between the two systems. The observation of the very same decrease of T_1^{-1} upon cooling might thus either be a coincidence or point toward an intrinsic energy scale defined by a collective ordering phenomenon in disordered

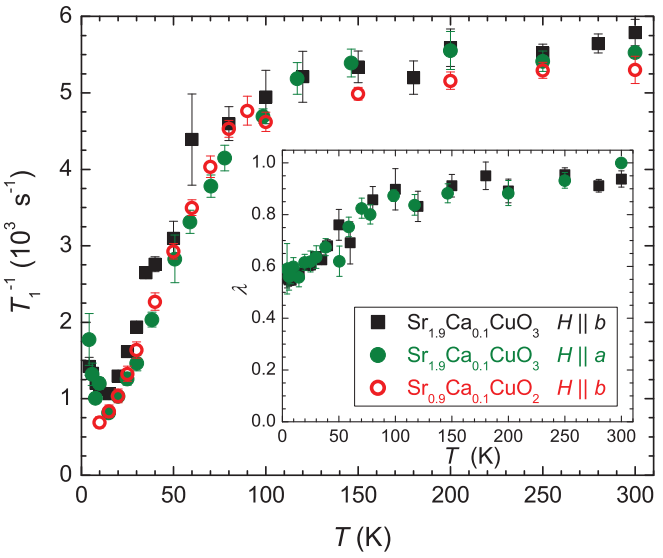

FIG. 2. (Color online) Temperature-dependent T_1^{-1} for $Sr_{1.9}Ca_{0.1}CuO_3$ measured along the b (black squares) and a (green dots) direction, in comparison to T_1^{-1} of $Sr_{0.9}Ca_{0.1}CuO_2$ measured along b (open red dots, taken from Ref. [9]). Note that the b direction of $SrCuO_2$ is crystallographically equivalent to a of Sr_2CuO_3 (see Fig. 1). The inset shows the temperature evolution of the stretching exponent λ of the relaxation function [see Eq. (1)].

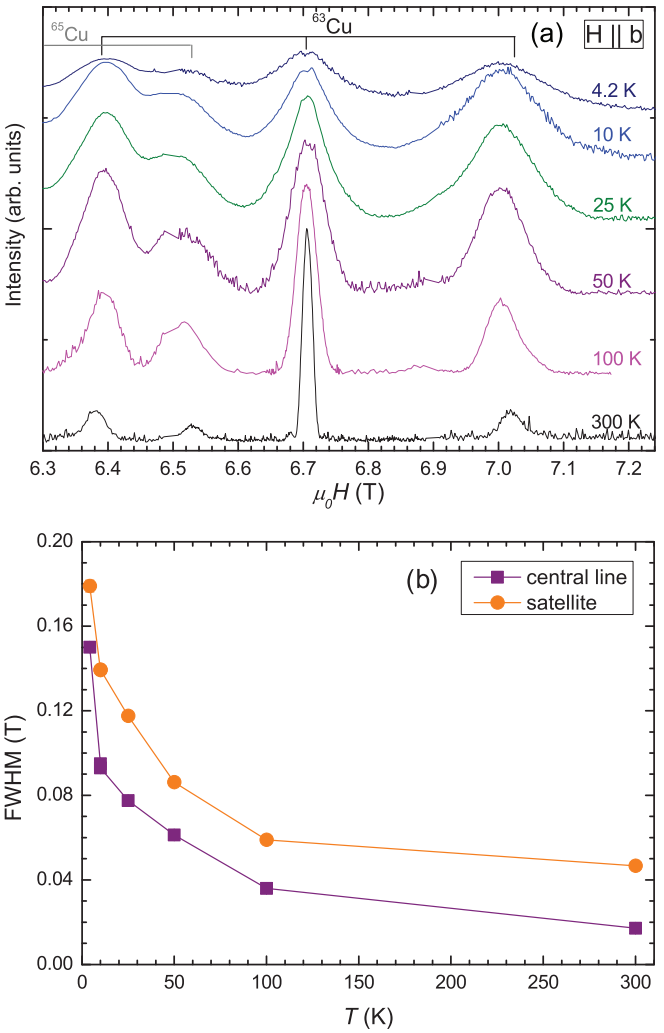

FIG. 3. (Color online) (a) ^{63}Cu NMR spectra of $Sr_{1.9}Ca_{0.1}CuO_3$ for $H \parallel b$ measured at different temperatures (see labels). All three resonance lines of the quadrupolar split ^{63}Cu NMR spectrum are visible. Additionally, the high field satellite of the ^{65}Cu isotope is visible at ∼6.52 T. (b) Temperature-dependent full width at half-maximum (FWHM) of the ^{63}Cu central line and the ^{63}Cu satellite at high field values, which is not disturbed by an overlapping ^{65}Cu resonance line. The almost parallel temperature dependence of the FWHM of the satellite and the central transition evidences a magnetic broadening effect.

spin chains. Therefore, further studies (e.g., of different doping levels) are envisaged.

Apart from the very similar decrease of T_1^{-1}, there are also slight differences between the two data sets of $Sr_{0.9}Ca_{0.1}CuO_2$ and $Sr_{1.9}Ca_{0.1}CuO_3$. At first, T_1^{-1} of $Sr_{1.9}Ca_{0.1}CuO_3$ starts to increase again below $T \approx 15$ K (see Fig. 2). This upturn could indicate a slowing down of magnetic fluctuations at least in some parts of the chains. Secondly, below $T \sim 100$ K the relaxation rates become more distributed, indicated by a decrease of the stretching exponent λ [see Eq. (1) and the inset of Fig. 2]. Such a stretching exponent was not needed to fit the data of Ca-doped $SrCuO_2$, where a similar gap feature was observed [9]. This suggests that the stretched exponent, which indicates a distribution of spin-lattice relaxation rates, is not linked to the gap, but rather to enhanced magnetic fluctuations in some parts of the chains. Thirdly, the ^{63}Cu NMR spectra broaden significantly upon lowering the temperature (see Fig. 3). This broadening is much stronger than what has been observed in the pure compound Sr_2CuO_3 [27,28]. There, a subtle structure of shoulders, broadening, and splitting evolves at low temperatures and has been assigned to a field-induced local staggered magnetization developing at broken chain ends [27,28]. While these chain ends might also be present in the investigated $Sr_{1.9}Ca_{0.1}CuO_3$, the associated subtle structure of the spectra cannot be observed. Already at room temperature, the spectrum is broader than the one of Sr_2CuO_3. This is due to the impact of disorder induced by the Ca doping, which is especially demonstrative at the satellites. Disorder mainly affects the quadrupolar interaction between the quadrupolar moment of the nuclei and the disordered electrical field gradient. However, the further NMR line broadening upon lowering the temperature evolves simultaneously for all the resonance lines [see Fig. 3(b)], suggesting a magnetic origin. In line with the upturn of T_1^{-1} and the decrease of λ, this broadening points toward slowly fluctuating magnetic moments.

These results suggest that the spin excitations in the chains of $Sr_{1.9}Ca_{0.1}CuO_3$ are not simply subject to a spin gap, but that there could be regions in the sample that are closer to a magnetic order. Therefore, the spin gap should rather be regarded as a pseudo spin gap, where the term "pseudo" indicates that there is either a distribution of gaps [18], or the opening of the gap is not complete in a sense that there are still magnetic excitations in some regions of the sample. This observation is in line with our previous DMRG

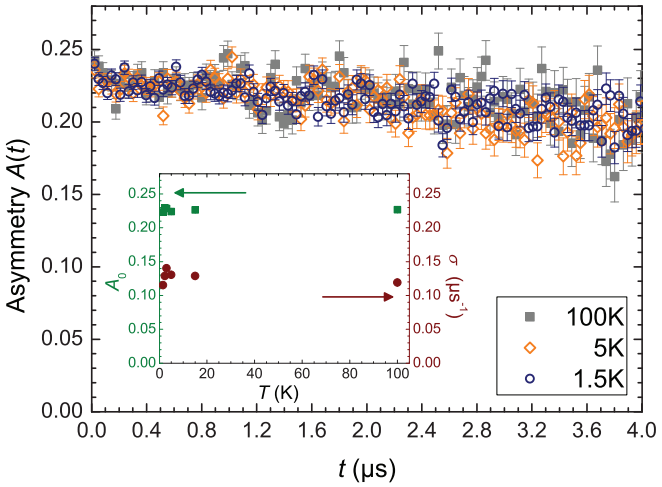

FIG. 4. (Color online) ZF μSR asymmetries of $Sr_{1.9}Ca_{0.1}CuO_3$ measured at 100 K (gray squares), 5 K (orange diamonds), and 1.5 K (blue dots), respectively. The inset shows the temperature dependence of the initial asymmetry A_0 (green squares) and the Gaussian decay rate σ (brown dots) obtained by fitting the asymmetry time spectra to Eq. (2).

calculations [9] as well as with recent inelastic neutron scattering measurements on $SrCu_{0.99}Ni_{0.01}O_2$ [18].

Let us recall that the parent compound Sr_2CuO_3 exhibits a 3D magnetic order below $T_N \sim 5$ K [2]. On the other hand, $SrCuO_2$ orders only below $T_N \sim 2$ K. This could suggest that the low-temperature upturn of T_1^{-1} in $Sr_{1.9}Ca_{0.1}CuO_3$ points toward an onset of magnetic order.

To search for such a magnetic order at low temperatures, we performed ZF μSR measurements on $Sr_{1.9}Ca_{0.1}CuO_3$ down to 1.5 K. Due to the extreme sensitivity of the muons to small magnetic fields, μSR is ideally suited to study possibly weak and local magnetism. Some representative ZF μSR asymmetry spectra are shown in Fig. 4. For all temperatures (from 100 K down to 1.5 K), we observe only a simple Gaussian decay due to nuclear dipolar interaction. The decay rate σ and the initial asymmetry A_0, obtained by fitting the ZF asymmetry time dependence $A(t)$ to

$$A(t) = A_0 e^{-(\sigma t)^2}, \tag{2}$$

do not change upon cooling down (see the inset of Fig. 4). Additionally, we performed transverse field (TF) μSR measurements in 30 G, and we could observe the whole initial asymmetry also at the lowest measured temperature 1.5 K (not shown). These results suggest that no magnetic order sets in in $Sr_{1.9}Ca_{0.1}CuO_3$ down to 1.5 K in zero external field. This observation is in line with measurements of the specific heat, which also did not show any evidence of a magnetic order down to 2 K [29]. Nonetheless, the upturn of the ^{63}Cu NMR T_1^{-1}, together with the decrease of the stretching exponent λ and the strong NMR line broadening, are pointing toward growing magnetic correlations, at least in a strong magnetic field at low temperatures. These results indicate an approach toward a magnetic order at lower temperatures than the experimentally accessed ones, or in high magnetic fields.

The suppression of long-range magnetic order by the opening of a gap is, however, a remarkable observation. To the best of our knowledge, only the reverse effect has been observed so far, namely the appearance of magnetic order in a nominally gapped system induced by impurity doping [30–32].

IV. CONCLUSION

We have used NMR measurements to investigate the low-energy spin dynamics of the $S = 1/2$ antiferromagnetic Heisenberg spin-chain compound $Sr_{1.9}Ca_{0.1}CuO_3$. We found an exponential decrease of the ^{63}Cu NMR T_1^{-1} upon decreasing temperature, indicating the opening of a pseudo spin gap of the same order of magnitude as in the double-chain compound $Sr_{0.9}Ca_{0.1}CuO_2$. The observation of the spin gap *per se* suggests that it is solely induced by a local bond disorder causing a small alternation of the intrachain exchange coupling J in Ca-doped $SrCuO_2$ and Sr_2CuO_3, and that it does not depend on the existence of an interchain coupling J'. The observation of the very same spin-gap value Δ in both compounds is surprising and might indicate an intrinsic energy scale defined by a collective ordering phenomenon in disordered spin-chain systems, which should be investigated in more detail in the future. At low temperatures, we observe an upturn of T_1^{-1}, accompanied by a growing distribution of T_1^{-1}, expressed in a decreasing stretching exponent λ and a simultaneous broadening of all the ^{63}Cu NMR resonance lines. While we could prove the absence of any kind of static magnetic order by ZF μSR measurements down to 1.5 K, these NMR features indicate growing magnetic correlations at low temperatures in a strong magnetic field. Nevertheless, the evidence for a suppression of magnetic order and the concomitant appearance of a spin gap remain unambiguous.

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[1] M. Mourigal, M. Enderle, A. Klöpperpieper, J.-S. Caux, A. Stunault, and H. M. Ronnow, Nat. Phys. **9**, 435 (2013).

[2] A. Keren, L. P. Le, G. M. Luke, B. J. Sternlieb, W. D. Wu, Y. J. Uemura, S. Tajima, and S. Uchida, Phys. Rev. B **48**, 12926 (1993).

[3] K. M. Kojima, Y. Fudamoto, M. Larkin, G. M. Luke, J. Merrin, B. Nachumi, Y. J. Uemura, N. Motoyama, H. Eisaki, S. Uchida *et al.*, Phys. Rev. Lett. **78**, 1787 (1997).

[4] M. Matsuda, K. Katsumata, K. M. Kojima, M. Larkin, G. M. Luke, J. Merrin, B. Nachumi, Y. J. Uemura, H. Eisaki, N. Motoyama *et al.*, Phys. Rev. B **55**, R11953 (1997).

[5] I. A. Zaliznyak, C. Broholm, M. Kibune, M. Nohara, and H. Takagi, Phys. Rev. Lett. **83**, 5370 (1999).
[6] S. Sachdev, Phys. Rev. B **50**, 13006 (1994).
[7] A. W. Sandvik, Phys. Rev. B **52**, R9831 (1995).
[8] M. Takigawa, N. Motoyama, H. Eisaki, and S. Uchida, Phys. Rev. Lett. **76**, 4612 (1996).
[9] F. Hammerath, S. Nishimoto, H.-J. Grafe, A. U. B. Wolter, V. Kataev, P. Ribeiro, C. Hess, S.-L. Drechsler, and B. Büchner, Phys. Rev. Lett. **107**, 017203 (2011).
[10] T. Giamarchi, *Quantum Physics in One Dimension* (Clarendon, Oxford, 2004).
[11] C. Kim, A. Y. Matsuura, Z.-X. Shen, N. Motoyama, H. Eisaki, S. Uchida, T. Tohyama, and S. Maekawa, Phys. Rev. Lett. **77**, 4054 (1996).
[12] R. Neudert, M. Knupfer, M. S. Golden, J. Fink, W. Stephan, K. Penc, N. Motoyama, H. Eisaki, and S. Uchida, Phys. Rev. Lett. **81**, 657 (1998).
[13] J. Schlappa *et al.*, Nature (London) **485**, 82 (2012).
[14] N. Hlubek, P. Ribeiro, R. Saint-Martin, A. Revcolevschi, G. Roth, G. Behr, B. Büchner, and C. Hess, Phys. Rev. B **81**, 020405 (2010).
[15] A. V. Mahajan and N. Venkataramani, Phys. Rev. B **64**, 092410 (2001).
[16] K. M. Kojima, J. Yamanobe, H. Eisaki, S. Uchida, Y. Fudamoto, I. M. Gat, M. I. Larkin, A. Savici, Y. J. Uemura, P. P. Kyriakou *et al.*, Phys. Rev. B **70**, 094402 (2004).
[17] S. Chattopadhyay, S. Giri, and S. Majumdar, J. Phys.: Condens. Matter **23**, 216006 (2011).
[18] G. Simutis, S. Gvasaliya, M. Månsson, A. L. Chernyshev, A. Mohan, S. Singh, C. Hess, A. T. Savici, A. I. Kolesnikov, A. Piovano *et al.*, Phys. Rev. Lett. **111**, 067204 (2013).
[19] N. Hlubek, P. Ribeiro, R. Saint-Martin, S. Nishimoto, A. Revcolevschi, S.-L. Drechsler, G. Behr, J. Trinckauf, J. E. Hamann-Borrero, J. Geck *et al.*, Phys. Rev. B **84**, 214419 (2011).
[20] N. Motoyama, H. Eisaki, and S. Uchida, Phys. Rev. Lett. **76**, 3212 (1996).
[21] T. Rice, Europhys. Lett. **23**, 445 (1993).
[22] A. Revcolevschi, U. Ammerahl, and G. Dhalenne, J. Cryst. Growth **198–199**, Part 1, 593 (1999).
[23] D. Lines, M. Weller, D. Currie, and D. Ogbome, Mater. Res. Bull. **26**, 323 (1991).
[24] M. Xu, E. Voiles, L. Chumbley, A. Goldman, and D. Finnemore, J. Mater. Res. **7**, 1283 (1992).
[25] M. Takigawa, N. Motoyama, H. Eisaki, and S. Uchida, Phys. Rev. B **57**, 1124 (1998).
[26] A. F. Mcdowell, J. Magn. Res., Ser. A **113**, 242 (1995).
[27] M. Takigawa, N. Motoyama, H. Eisaki, and S. Uchida, Phys. Rev. B **55**, 14129 (1997).
[28] J. P. Boucher and M. Takigawa, Phys. Rev. B **62**, 367 (2000).
[29] N. S. Beesetty (private communication).
[30] M. Azuma, Y. Fujishiro, M. Takano, M. Nohara, and H. Takagi, Phys. Rev. B **55**, R8658 (1997).
[31] B. Grenier, P. Monod, M. Hagiwara, M. Matsuda, K. Katsumata, S. Clément, J.-P. Renard, A. L. Barra, G. Dhalenne, and A. Revcolevschi, Phys. Rev. B **65**, 094425 (2002).
[32] F. Casola, T. Shiroka, S. Wang, K. Conder, E. Pomjakushina, J. Mesot, and H.-R. Ott, Phys. Rev. Lett. **105**, 067203 (2010).