Magnetic ordering in electronically phase-separated $La_{2-x}Sr_xCuO_{4+y}$: Neutron diffraction experiments

L. Udby,[1] N. H. Andersen,[1] F. C. Chou,[2] N. B. Christensen,[1,3,4] S. B. Emery,[5] K. Lefmann,[1,4,6] J. W. Lynn,[7] H. E. Mohottala,[5] Ch. Niedermayer,[3] and B. O. Wells[5]

[1]*Materials Research Division, Risø DTU, Technical University of Denmark, DK-4000 Roskilde, Denmark*

[2]*Center for Condensed Matter Sciences, National Taiwan University, Taipei 10617, Taiwan*

[3]*Laboratory for Neutron Scattering, ETHZ and PSI, CH-5232 Villigen PSI, Switzerland*

[4]*Nanoscience Center, Niels Bohr Institute, University of Copenhagen, DK-2100 Copenhagen, Denmark*

[5]*Department of Physics, University of Connecticut, 2152 Hillside Road, U-3046, Storrs, Connecticut 06269-3046, USA*

[6]*ESS-Scandinavia, University of Lund, Stora Algatan 4, 22350 Lund, Sweden*

[7]*NIST Center for Neutron Research, National Institute of Standards and Technology, Gaithersburg, Maryland 20899, USA*

(Received 31 March 2009; revised manuscript received 8 June 2009; published 9 July 2009)

We present results of magnetic neutron diffraction experiments on the codoped superoxygenated $La_{2-x}Sr_xCuO_{4+y}$ ($LSCO_{+O}$) system with $x=0.09$. We find that the magnetic phase is long-range ordered incommensurate antiferromagnetic with a Néel temperature T_N coinciding with the superconducting ordering temperature $T_c=40$ K. The incommensurability value is consistent with a hole doping of $n_h \approx \frac{1}{8}$ but in contrast to nonsuperoxygenated $La_{2-x}Sr_xCuO_4$ with hole doping close to $n_h \approx \frac{1}{8}$ the magnetic-order parameter is not field dependent. We attribute this to the magnetic order being fully developed in $LSCO_{+O}$ as in the spin and charge ordered "stripe" compounds $La_{1.48}Nd_{0.40}Sr_{0.12}CuO_4$ and $La_{7/8}Ba_{1/8}CuO_4$.

DOI: 10.1103/PhysRevB.80.014505 PACS number(s): 74.25.Ha, 74.72.Dn, 75.25.+z

I. INTRODUCTION

The presence of an inhomogeneous charge concentration in cuprate superconductors has become increasingly obvious in recent years. The most dramatic experiments showing local density of state variations have been performed using scanning tunneling microscopy or spectroscopy.[1–3] There has also been an increasing number of other experiments that are best explained by invoking an inhomogeneous electronic structure.[4–7] For most of these experiments the charge variations appear to be short ranged, associated with a length scale of only a few nanometers at most. However, for the special cases of oxygen-doped La_2CuO_{4+y} (LCO_{+O}) or oxygen-codoped $La_{2-x}Sr_xCuO_{4+y}$ ($LSCO_{+O}$), muon and superconducting quantum interference techniques suggest that the electronic inhomogeneity moves beyond such local variations to form fully phase-separated regions.[8,9] For both cases, with hole concentrations (n_h) between 0.125 and 0.16 per Cu site, samples at low temperatures spontaneously form separate regions of (i) a nonsuperconducting, magnetically ordered phase (with physical properties indicating $n_h=1/8$), and (ii) an optimally doped superconductor (with $n_h=0.16$) that is not magnetically ordered. The driving force for this phase separation appears to be interactions between the doped holes themselves rather than any specific O or Sr chemistry.[9]

The full implications of this complete phase separation are still to be determined both theoretically and empirically. One area where phase separation should certainly be important is for effects associated with competing order parameters. A pronounced case of such an effect is the large magnetic field enhancement of the ordered moment in underdoped $La_{2-x}Sr_xCuO_4$ (LSCO) superconductors as measured by neutron diffraction. A series of experiments have shown that samples with $x<1/8$ have an incommensurate (IC) antiferromagnetic (AFM) elastic diffraction peak that grows substantially with the application of a magnetic field.[10,11] Samples with $x \gtrsim 0.14$ have no elastic magnetic peak in zero applied field, but such a peak appears at a critical field H_c and then grows in intensity as the field increases above that.[10,12] For samples doped very close to $x=1/8$, and for which suppression of the superconducting T_c is also observed, a strong magnetic peak exists at zero field with less enhancement from the application of a field. Samples of $La_{1.88}Sr_{0.12}CuO_4$ still show a small field enhancement[13] while samples of $La_{1.48}Nd_{0.4}Sr_{0.12}CuO_4$ (LNSCO) and $La_{7/8}Ba_{1/8}CuO_4$ (LBCO) have a fully developed magnetic moment and no or very small field enhancement.[10,14,15] We will hereafter refer to samples with fully developed magnetic order and no field enhancement within 13.5 T applied field as true 1/8 samples.

A widely used theory for the intensity enhancement by application of an external field has been developed by Demler, Sachdev, and Zhang (DSZ).[16] This theory (DSZ) describes the cuprates as having coexisting but competing magnetic and superconducting order parameters. The functional form for the magnetic peak intensity versus field appears to fit existing data well and the predicted phase diagram appears to qualitatively match measurements for samples which are not true 1/8. However, the observation that true 1/8 samples have no field enhancement is not predicted by the DSZ paper. Of natural interest is how this theory of competing but coexisting order parameters might be adapted for related samples which appear to have fully separated order parameters that do not coexist at the same location in the sample.

In the present work we study the details of the elastic magnetic scattering of a $LSCO_{+O}$ single crystal by neutron diffraction. The phase separation, phase fractions, zero-field-ordered moment, and flux pinning have been carefully mea-

sured previously using muon spin resonance (μSR) and bulk susceptibility measurements.[9,17] Here we establish that the magnetic phase is IC AFM and long-range ordered with the same incommensurability as the true 1/8 samples of LNSCO and LBCO. This is surprising since the superconducting transition temperature is not suppressed in our sample but instead is very high (T_c=40 K) and coinciding with the ordering temperature of the IC AFM. Field and temperature dependences of the IC AFM peak intensity are also presented. We discuss the field dependence in relation to the DSZ model, pointing out that some development is necessary to account for samples where the magnetic and the superconducting phases fully separate rather than coexist.

II. METHODS

Our sample is a codoped single crystal with Sr content x=0.09 ($LSCO_{+O}^{x=0.09}$). It has mass m=0.429 g. It was grown by the traveling solvent floating-zone method in a mirror furnace. Additional oxygen was introduced using wet electrochemical methods as presented previously.[18] Previous studies of this particular crystal showed onset for both superconductivity and magnetism at 40 K.[9] Only one superconducting and one magnetic phase were detected and each of these two phases occupy close to 50% of the volume as measured by μSR.

The neutron diffraction measurements were performed at the cold neutron triple-axis spectrometers RITA-II at the Paul Scherrer Institute, Switzerland (PSI) and SPINS at the National Institute of Standards and Technology, Maryland, USA (NIST). In both experiments we used 5 meV neutrons, 40′ collimation before the sample, and a Be filter before the analyzer. Error bars in this paper are statistical in nature and represent one standard deviation.

RITA-II has the special feature of a seven blade PG(002) analyzer making it possible to simultaneously monitor seven different reciprocal space points and energy transfer, the so-called monochromatic imaging mode.[19–21] This enables simultaneous measurements of peak and background, which have proven very useful since the weak magnetic signal requires very long counting times on the order of hours. The size of the sample and the width of the analyzer blades result in an effective horizontal collimation of 40′ between the sample and each analyzer blade.

The LSCO system is subjected to twinning when in the low-temperature orthorhombic (LTO) state. In terms of the $F4/mmm$ setting of the high-temperature tetragonal (HTT) structure the twinning is along (110) and $(1\bar{1}0)$. The orthorhombic axes are almost parallel to the $F4/mmm$ axes and the twinning gives up to four peaks in the LTO phase for each peak in the HTT phase.[22] In our crystal even at low temperatures in the LTO state,[23] the difference between lattice constants a=5.318(4) Å and b=5.337(6) Å gives rise to only a tiny transversal splitting of α=0.10(4)° across the H and the K axes. All references to crystallography are in the LTO $Bmab$ notation unless explicitly stated otherwise.

III. RESULTS

Previous μSR studies[9] of our $LSCO_{+O}^{x=0.09}$ crystal have shown a strongly damped oscillatory behavior with ν =3.33(8) MHz. This corresponds to an internal field of 24.7 mT, which is about 2/3 of the value observed in the undoped compound La_2CuO_4 (LCO).[24]

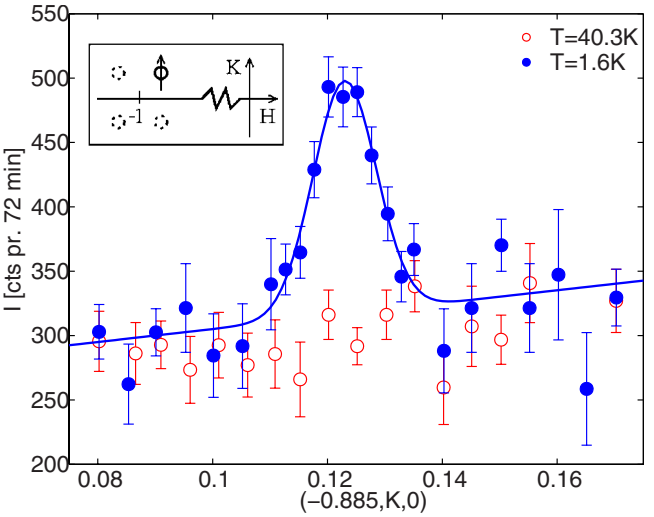

FIG. 1. (Color online) Scans in reciprocal space through (−0.885,0.123,0) along the K direction. Scans are taken above and below T_N in zero applied field; the solid line is a fit to the data as explained in Sec. IV. The inset shows the peak position and scan direction.

Our neutron diffraction studies find IC AFM elastic peaks at the same scattering vectors as in superoxygenated LCO_{+O} crystals.[25,26] Scans through the incommensurate point are shown in Fig. 1 at base temperature and just above the magnetic transition temperature T_N. The peak incommensurabilities are δ_H=0.115(2) r.l.u. and δ_K=0.123(1) r.l.u., respectively, which gives a distance δ=0.198(3) $Å^{-1}$ from the AFM point. This corresponds to an incommensurability of δ_T=0.119(2) r.l.u. in pseudotetragonal notation,[27] which is consistent with a hole doping of $n_h \approx 1/8$ according to the Yamada plot[28] and consistent with $\delta \approx 0.12$ of LBCO (Ref. 29) and LNSCO.[10] The incommensurate peaks are similar to or sharper than the previously reported instrumentally resolved ones in LCO_{+O}.[25,26,30,31] It has been checked by multiple tests that the peak width, the position, and the amplitude do not depend on cooling rate or cycle.

As shown in Fig. 2, the application of a field does not shift or broaden the IC peak within the error bars, making it possible to monitor the intensity as a function of applied field in few-point scans. In imaging mode at RITA-II the central blade is used for measuring the peak amplitude whereas the other blades measure the background which is found to be field independent. Data for applied fields up to 13.5 T are shown in Fig. 3. Simply fitting the peak intensity to a constant describes our data quite accurately meaning that we observe no field effect within this field range. This is similar to the anomalous behavior of the other true 1/8 samples such as LNSCO and LBCO. In addition, the $LSCO_{+O}$ system also has the same μSR response as LBCO and LNSCO, which is a strongly damped oscillatory behavior with a frequency $\nu \approx 3.5$ MHz corresponding to a local-ordered moment of $\sim 0.35 \mu_B$.[9,10,32] The neutron-scattering data are proportional to the ordered spin-moment squared. Therefore, in order to

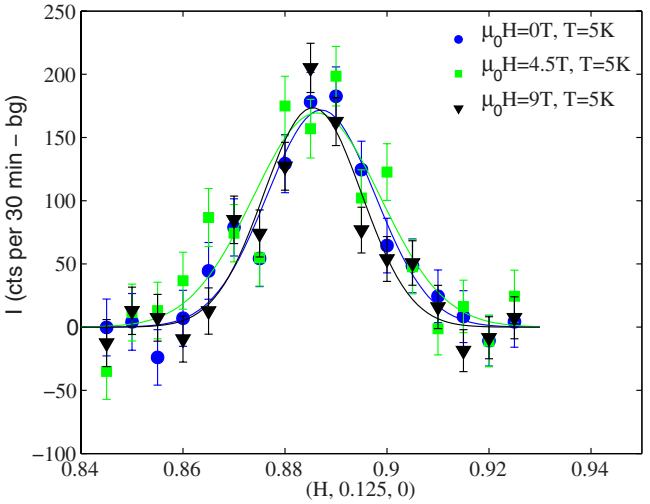

FIG. 2. (Color online) H scans through the IC peak position at various fields. The experimental data have been fit to a Gaussian on sloping background. In the figure the data points and their Gaussian fits (solid lines) are shown after subtracting their sloping background (940 cts/30 min at the peak position). All fitted Gaussian parameters are the same within error bars for the different fields.

compare with μSR results, our data have been presented after taking the square root of the background-subtracted measured intensities and scaling to LNSCO muon data in Ref. 10.

The temperature dependence of the IC spin-density wave (SDW) peak intensity is shown in Fig. 4 for both 0 and 13.5 T applied fields. From a linear mean-field fit we find a magnetic-ordering temperature of $T_N=40(4)$ K for both the 0 and the 13.5 T data. The magnetic transition temperature is

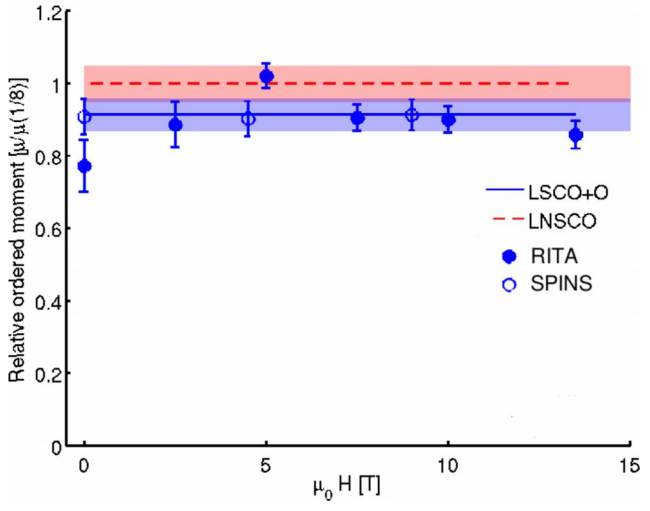

FIG. 3. (Color online) Internal magnetic moment (square root of SDW peak intensities) as a function of applied field. Closed symbols are data from RITA-II (measuring one point at the peak position) and open symbols are from SPINS (full-momentum scan fitted to a Gaussian) scaled to weighted average of RITA-II data. A constant fit to the data (solid line) is shown relative to the internal moment of LNSCO from Ref. 10 (dashed line). Shaded areas indicate the error related to the determination of the local-ordered moment from μSR.

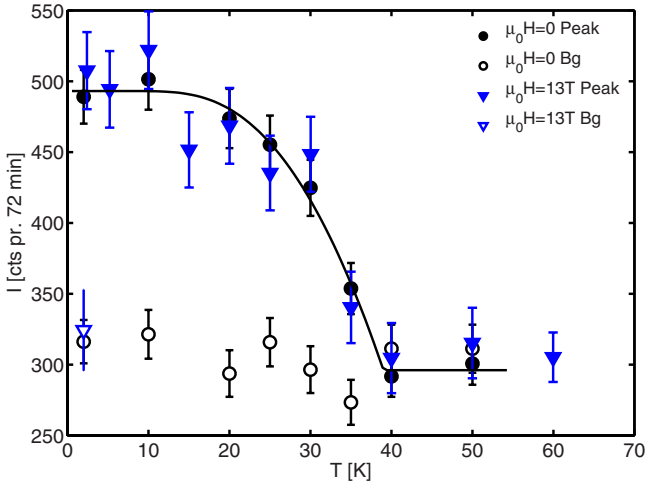

FIG. 4. (Color online) Temperature dependence of the IC SDW peak intensity at $\mu_0 H=0$ (in cryostat) and $\mu_0 H=13.5$ T (in magnet, after field cooling), respectively. The $\mu_0 H=13.5$ T data are scaled (from the ratio of Bragg-peak intensity in the cryostat and magnet, respectively) and subtracted a constant background. The solid curve is a guide to the eyes.

well within the experimental uncertainties of the measured bulk superconducting transition temperature.

IV. DISCUSSION

We first consider the IC AFM peak of Fig. 1. We find that the peak has $\mathrm{FWHM_{IC}}=0.0130(14)$ r.l.u. by a simple Gaussian fit on sloping background, which is slightly broader than the resolution at the $(-1,0,0)$ position having $\mathrm{FWHM_{res}}=0.0101(1)$ r.l.u..[33] However, even if we take the small excess width with respect to the resolution width to be due to finite-size broadening, the SDW correlation length will still exceed 400 Å.[34] Hence it is reasonable to conclude that the IC peak is close to resolution limited and expressing long-range SDW order. The intrinsic (i.e., corrected for resolution broadening) width of our IC AFM peak is the same as the width of the IC AFM peak of LSCO $x=0.12$ and LNSCO (Ref. 10), which are resolution limited. Thus the correlation length is maximum near the 1/8 state whereas it decreases in LSCO when doping departs from $x=1/8$.[10,35]

Imposing a field does not change the intensity, the correlation length, the incommensurability, or the transition temperature of the magnetic phase, and it is hence reasonable to conclude that the magnetic state in the part of the sample with $n_h=1/8$ is already fully developed in zero field.

This is in contrast to LSCO $x=0.12$ for which neutron diffraction studies have shown that the field enhancement matches the functional form of the DSZ theory. In this case the superconductivity (SC) and the SDW coexist but the local-ordered magnetic moment is not fully developed in zero applied field. Imposing a field, however, pushes it toward the local-ordered moment of the true 1/8 state.[10]

Enhancement up to a factor of 2 of the elastic IC AFM peak at moderate fields (<8 T) has also been observed by neutron diffraction in non-Sr-doped $\mathrm{LCO_{+O}}$ crystals when

they were cooled slowly enough for the excess oxygen to order.[26,36] The oxygen ordering is observable by the concomitant staging superstructure. At the time of writing the authors of Ref. 26 did not consider their crystal to be macroscopically phase separated. Given more recent developments, and the fact that the reported magnetic and superconducting properties of that crystal are very similar to ours, it seems likely that the sample used in that report was indeed phase separated in a manner similar to the sample we present here. Hence in the following discussion we will assume that this is the case. It is however important to bear in mind that the value of the magnetic volume fraction of otherwise similar LCO_{+O} crystals can vary significantly.[37]

One possible explanation for the increase in the magnetic signal in Ref. 26 could be that the local magnetic moment in this sample was not saturated in zero field and after slow cooling. However, from our previous μSR work we know that highly oxygenated LCO_{+O} crystals have a fully developed local magnetic moment at zero applied field,[9] so the field effect of LCO_{+O} is probably not explained by an unsaturated magnetic moment. We speculate instead that it is due to the ability of LCO_{+O} to convert SC regions into SDW in the case where the excess oxygen is ordered. The mechanism behind this might be similar to what is found in $YBa_2Cu_3O_{6+x}$ (YBCO). In YBCO (Refs. 38 and 39) oxygen ordering facilitates itinerant-doped holes thereby favoring SC, whereas oxygen disorder does not favor SC. In this scenario SC would be favored in slowly cooled oxygen-ordered LCO_{+O} at least at zero applied field. Applying moderate fields hereafter allows SDW regions to grow to a plateau volume. In LCO_{+O} oxygen disorder can be introduced by fast cooling. In $LSCO_{+O}$, which can be viewed as doping LCO_{+O} with Sr, the homogeneously distributed Sr anticorrelates to the excess oxygen, creating an increasingly disordered oxygen distribution with increasing Sr content.[23] Thus following this scenario, in $LSCO_{+O}^{x=0.09}$ and fast-cooled LCO_{+O}, SC regions are not particularly favored over SDW regions even at zero applied field. This can explain why we see little or no enhancement of the magnetic signal by the application of a field in the $LSCO_{+O}$ system.

Let us now consider the volume of our sample with $n_h=0.16$ separately and treat it within the DSZ frame. Then keeping our high T_c of 40 K in mind, probably the critical field needed to actually enhance the magnetic signal in $LSCO_{+O}$ would be at least as large as that of LSCO $x=0.16$ (optimally doped). According to the DSZ phase diagram, the critical field increases rapidly with increasing x. The fact that the critical field of $x=0.145$ is already 7 T (Ref. 10) suggests that the critical field for $x=0.16$ would not be within our experimental reach. The total outcome considering both phases would be that neither of them would show significant field enhancement of the magnetic signal in moderate fields, which is indeed what we observe.

There was no evidence in our codoped $LSCO_{+O}$ crystal of any superconducting phase with T_c different from 40 K,[9] nor did we observe any signs of the Néel antiferromagnetic order observed in LCO (Ref. 24) as well as in the hole-poor phase of nonSr-doped LCO_{+O}.[40] This is corroborating evidence for the suggestion[9] that in the region of the $LSCO_{+O}$ phase diagram to which our sample belongs, there exists only two stable ground states: the optimally doped superconducting phase and the "true 1/8" magnetically ordered SDW phase.

V. CONCLUSION

We conclude that the magnetic phase in our $LSCO_{+O}$ crystal consists of fully developed long-range SDW order corresponding to the SDW of the 1/8 compounds LBCO and LNSCO with the same incommensurability $\delta \approx 0.12$, local Cu^{2+} moment $\sim 0.35\mu_B$, and lack of field effect. The regions occupied by the SDW are large, at least 400 Å, meaning that below 40 K large SDW islands (or patches correlated over large distances) form simultaneously with the optimally doped SC in the rest of the sample.

Since there is no enhancement of the IC AFM peak with the application of external field, we conclude that the $LSCO_{+O}$ system has a fully developed magnetic phase, which cannot be expanded at least below 13.5 T applied field. In the slow-cooled highly oxygen-doped LCO_{+O} system with the same magnetic structure there is clearly a (large) enhancement of the SDW peak with field. We speculate that the discrepancy with respect to our system is due to the ability of the (slow-cooled) LCO_{+O} system to convert SC volume into SDW volume by application of an external field. This ability might be related to the degree of oxygen ordering in the sample. Our neutron-scattering measurements support that codoping facilitates long-range electronic phase separation below $T_N=T_c=40$ K in two phases, 40 K SC and true 1/8 SDW, whose relative amounts are only determined by the total hole content.

ACKNOWLEDGMENTS

We would like to thank Brian M. Andersen for many helpful discussions. This work was supported by the Danish Agency for Science Technology and Innovation under the Framework Programme on Superconductivity and the Danish Research Council FNU through the instrument center DANSCATT. Work at the University of Connecticut was supported by the U.S. Department of Energy under Contract No. DE-FG02-00ER45801. Part of this work was performed at the Swiss Spallation Neutron Source, Paul Scherrer Institute, Villigen, Switzerland. Work on SPINS is supported in part by the National Science Foundation under Agreement No. DMR-0454672.

[1] K. M. Lang, V. Madhavan, J. E. Hoffman, E. W. Hudson, H. Eisaki, S. Uchida, and J. C. Davis, Nature (London) **415**, 412 (2002).

[2] C. Howald, P. Fournier, and A. Kapitulnik, Phys. Rev. B **64**, 100504(R) (2001).

[3] Y. Kohsaka, C. Taylor, K. Fujita, A. Schmidt, C. Lupien, T. Hanaguri, M. Azuma, M. Takano, H. Eisaki, H. Takagi, S. Uchida, and J. C. Davis, Science **315**, 1380 (2007).

[4] M. v. Zimmermann, A. Vigilante, T. Niemöller, N. Ichikawa, T. Frello, J. Madsen, P. Wochner, S. Uchida, N. H. Andersen, J. Tranquada, D. Gibbs, and J. R. Schneider, EPL **41**, 629 (1998).

[5] S. V. Dordevic, S. Komiya, Y. Ando, and D. N. Basov, Phys. Rev. Lett. **91**, 167401 (2003).

[6] P. M. Singer, A. W. Hunt, and T. Imai, Phys. Rev. Lett. **88**, 047602 (2002).

[7] S. Wakimoto, K. Yamada, J. M. Tranquada, C. D. Frost, R. J. Birgeneau, and H. Zhang, Phys. Rev. Lett. **98**, 247003 (2007).

[8] A. T. Savici, Y. Fudamoto, I. M. Gat, T. Ito, M. I. Larkin, Y. J. Uemura, G. M. Luke, K. M. Kojima, Y. S. Lee, M. A. Kastner, R. J. Birgeneau, and K. Yamada, Phys. Rev. B **66**, 014524 (2002).

[9] H. E. Mohottala, B. O. Wells, J. I. Budnick, W. A. Hines, C. Niedermayer, L. Udby, C. Bernhard, A. R. Moodenbaugh, and F. C. Chou, Nature Mater. **5**, 377 (2006).

[10] J. Chang, C. Niedermayer, R. Gilardi, N. B. Christensen, H. Rønnow, D. F. McMorrow, M. Ay, J. Stahn, O. Sobolev, A. Hiess, S. Pailhes, C. Baines, N. Mornono, M. Oda, M. Ido, and J. Mesot, Phys. Rev. B **78**, 104525 (2008).

[11] B. Lake, H. M. Rønnow, N. B. Christensen, G. Aeppli, K. Lefmann, D. F. McMorrow, P. Vorderwisch, P. Smeibild, N. Mangkorntong, T. Sasagawa, M. Nohara, H. Takagi, and T. E. Mason, Nature (London) **415**, 299 (2002).

[12] B. Khaykovich, S. Wakimoto, R. J. Birgeneau, M. A. Kastner, Y. S. Lee, P. Smeibidl, P. Vorderwisch, and K. Yamada, Phys. Rev. B **71**, 220508(R) (2005).

[13] S. Katano, M. Sato, K. Yamada, T. Suzuki, and T. Fukase, Phys. Rev. B **62**, R14677 (2000).

[14] M. Fujita, M. Matsuda, H. Goka, T. Adachi, Y. Koike, and K. Yamada, J. Phys.: Conf. Ser. **51**, 510 (2006).

[15] J. Wen, Z. Xu, G. Xu, J. M. Tranquada, G. Gu, S. Chang, and H. J. Kang, Phys. Rev. B **78**, 212506 (2008).

[16] E. Demler, S. Sachdev, and Y. Zhang, Phys. Rev. Lett. **87**, 067202 (2001).

[17] H. E. Mohottala, B. O. Wells, J. I. Budnick, W. A. Hines, C. Niedermayer, and F. C. Chou, Phys. Rev. B **78**, 064504 (2008).

[18] B. O. Wells, R. J. Birgeneau, F. C. Chou, Y. Endoh, D. C. Johnston, M. A. Kastner, Y. S. Lee, G. Shirane, and J. M. Tranquada, Z. Phys. B: Condens. Matter **100**, 535 (1996).

[19] K. Lefmann, D. F. McMorrow, H. M. Rønnow, K. Nielsen, K. N. Clausen, B. Lake, and G. Aeppli, Physica B **283**, 343 (2000).

[20] C. R. H. Bahl, P. Andersen, S. N. Klausen, and K. Lefmann, Nucl. Instrum. Methods Phys. Res. B **226**, 667 (2004).

[21] C. R. H. Bahl, K. Lefmann, A. B. Abrahamsen, H. M. Rønnow, F. Saxild, T. B. S. Jensen, L. Udby, N. H. Andersen, N. B. Christensen, H. S. Jacobsen, T. Larsen, P. S. Hafliger, S. Streule, and Ch. Niedermayer, Nucl. Instrum. Methods Phys. Res. B **246**, 452 (2006).

[22] M. Braden, G. Heger, P. Schweiss, Z. Fisk, K. Gamayunov, I. Tanaka, and H. Kojima, Physica C **191**, 455 (1992).

[23] L. Udby *et al.* (unpublished).

[24] J. I. Budnick, B. Chamberland, D. P. Yang, C. Niedermayer, A. Golnik, E. Recknagel, M. Rossmanith, and A. Weidinger, EPL **5**, 651 (1988).

[25] Y. S. Lee, R. J. Birgeneau, M. A. Kastner, Y. Endoh, S. Wakimoto, K. Yamada, R. W. Erwin, S. H. Lee, and G. Shirane, Phys. Rev. B **60**, 3643 (1999).

[26] Y. S. Lee, F. C. Chou, A. Tewary, M. A. Kastner, S. H. Lee, and R. J. Birgeneau, Phys. Rev. B **69**, 020502(R) (2004).

[27] $a_T = b_T = \frac{a+b}{2\sqrt{2}} = \frac{5.328(4)}{\sqrt{2}} = 3.767(3)$ Å

[28] K. Yamada, C. H. Lee, K. Kurahashi, J. Wada, S. Wakimoto, S. Ueki, H. Kimura, Y. Endoh, S. Hosoya, G. Shirane, R. J. Birgeneau, M. Greven, M. A. Kastner, and Y. J. Kim, Phys. Rev. B **57**, 6165 (1998).

[29] M. Fujita, H. Goka, K. Yamada, J. M. Tranquada, and L. P. Regnault, Phys. Rev. B **70**, 104517 (2004).

[30] B. Khaykovich, Y. S. Lee, R. W. Erwin, S.-H. Lee, S. Wakimoto, K. J. Thomas, M. A. Kastner, and R. J. Birgeneau, Phys. Rev. B **66**, 014528 (2002).

[31] B. Khaykovich, R. J. Birgeneau, F. C. Chou, R. W. Erwin, M. A. Kastner, S.-H. Lee, Y. S. Lee, P. Smeibidl, P. Vorderwisch, and S. Wakimoto, Phys. Rev. B **67**, 054501 (2003).

[32] 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. MacLaughlin, and R. H. Heffner, Phys. Rev. B **58**, 8760 (1998).

[33] This was measured by second order scattering of the structural $(-2,0,0)$ peak (without Be filter) but the resolution is similar in angular space.

[34] If the peak is fitted to a Gaussian the combined width is the sum of the internal width σ and the resolution in quadrature giving $\sigma = \sqrt{(0.0130\frac{2\pi}{5.318})^2 - (0.0101 * \frac{2\pi}{5.337})^2} = 0.009\,72$ Å^{-1}. The estimate of the correlation length can be either $2\pi/\sigma = 646$ Å or by the Scherrer formula $\frac{K\lambda}{B\cos\theta} = \frac{0.9(4.045)}{0.00972(0.9404)} = 414$ Å with $B = 2\tan^{-1}\frac{\sigma}{2q_{IC}}$. The Scherrer dimensionless constant K is dependent on domain geometry but the value of 0.9 used here is correct within 10% error. The error on the Lorentzian width on fitting to a Voigt is as large as the value itself; hence, it has not been considered fitting to a Voigt in the main text.

[35] M. Fujita, K. Yamada, H. Hiraka, P. M. Gehring, S. H. Lee, S. Wakimoto, and G. Shirane, Phys. Rev. B **65**, 064505 (2002).

[36] S. B. Emery *et al.* (unpublished).

[37] As an example the LCO_{+O} crystals of Refs. 9 and 36 were prepared by a similar electrochemical method and both have $T_c = T_M = 40$ K but the first had a magnetic volume fraction of 66(5)% whereas the second only had 30(5)% measured in zero applied field and by the same μSR method after similar cooling conditions. We should mention that the first crystal is very small ($m = 0.025$ g) and hence no attempt has been made so far to measure the field dependence of the magnetic signal.

[38] M. v. Zimmermann, J. R. Schneider, T. Frello, N. H. Andersen, J. Madsen, M. Käll, H. F. Poulsen, R. Liang, P. Dosanjh, and W. N. Hardy, Phys. Rev. B **68**, 104515 (2003).

[39] G. Uimin, Phys. Rev. B **50**, 9531 (1994).

[40] E. J. Ansaldo, J. H. Brewer, T. M. Riseman, J. E. Schirber, E. L. Venturini, B. Morosin, D. S. Ginley, and B. Sternlieb, Phys. Rev. B **40**, 2555 (1989).