Orbital Control of Effective Dimensionality: From Spin-Orbital Fractionalization to Confinement in the Anisotropic Ladder System CaCu_{2}O_{3}

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Orbital control of effective dimensionality: from spin-orbital fractionalization to confinement in the anisotropic ladder system $CaCu_2O_3$

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Fractionalization of an electronic quasiparticle into spin, charge and orbital parts is a fundamental and characteristic property of interacting electrons in one dimension. However, real materials are never strictly one-dimensional and the fractionalization phenomena are hard to observe. Here we studied the spin and orbital excitations of the anisotropic ladder material $CaCu_2O_3$, whose electronic structure is not one-dimensional. Combining high-resolution resonant inelastic x-ray scattering experiments with theoretical model calculations we show that: (i) spin-orbital fractionalization occurs in $CaCu_2O_3$ along the leg direction x through the xz orbital channel as in a 1D system; and (ii) no fractionalization is observed for the xy orbital, which extends in both leg and rung direction, contrary to a 1D system. We conclude that the directional character of the orbital hopping can select different degrees of dimensionality. Using additional model calculations, we show that spin-orbital separation is *generally* far more robust than the spin-charge separation. This is not only due to the already mentioned selection realized by the orbital hopping, but also due to the fact that spinons are faster than the orbitons.

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An electron in a Mott insulator is characterized by fundamental quantum numbers, representing its charge, its spin and the orbital it occupies. But when electrons are confined to one dimension, these basic properties of the electron can break apart (fractionalize), forming three independently propagating excitations called spinon, holon and orbiton. While this very fundamental property of one-dimensional systems is both experimentally and theoretically well-established [1–5], its relevance in higher dimensions is heavily debated, in particular in relation to the high temperature superconductivity in two-dimensional cuprates [6–8].

The basic conceptual difference between one-dimensional (1D) and higher dimensional systems, such as ladders or two-dimensional materials (2D), can be illustrated by taking away an electron, having spin and charge, from either an antiferromagnetic (AF) chain or an AF plane [see Fig. 1(a) and (b)], and subsequently consider the propagating hole [9] in these two different situations. In the 1D case, the hole can start propagating freely after exciting only one spinon (a magnetic domain wall in the chain) and can consequently separate into a holon and a spinon [Fig. 1(a)] [1]. But when hopping between sites in an AF plane or ladder, the hole leaves behind a long trail of wrongly aligned spins [Fig. 1(b)] [10–12]. This trail acts as a string-potential that tends to confine the hole to its starting position and thereby to bind the spin and charge of the hole [10–13].

These basic examples illustrate that whereas spin-charge separation is a hallmark of 1D systems, any system beyond strictly 1D is much more complex because of the coupling of the charge carriers to the inherently strongly fluctuating quantum spin-background [1, 10]. It actually remains to be established if and how any of the peculiar 1D physics carries over to higher dimensions [14]. In this paper, we take a pragmatic approach to this problem: we try to verify whether a

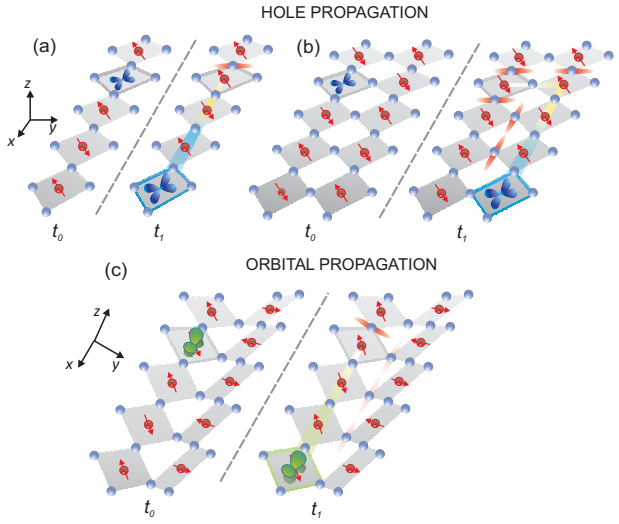

FIG. 1. (Color online) Schematic view of the hole/orbiton propagation in an S=1/2 environment. (a) A hole with $3d_{x^2-y^2}$ symmetry moves from its original position (marked in blue in the left chain) at time t_0 and while propagating at a later time t_1 it has created only one magnetic domain wall at the first hop (red cigar in the right chain). (b) In a 2D lattice, or equivalently in a two-leg ladder, the moving $3d_{x^2-y^2}$ hole creates a trail of magnetic domain walls. (c) In the buckled two-leg ladder $CaCu_2O_3$ studied here, when the $3d_{xz}$ *orbiton* moves (marked in green), it creates just one magnetic domain wall as in (a) not only because interleg domain walls (light red) can be neglected (due to very weak spin interaction along the rung) but also because it can move *solely* along one of the legs of the ladders (due to the directional hopping of the $3d_{xz}$ orbital).

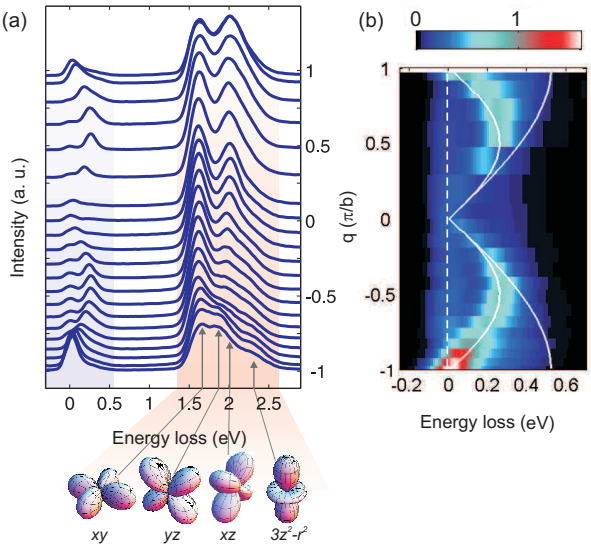

FIG. 2. (Color Online) RIXS spectra of $CaCu_2O_3$ measured at the maximum of the Cu L_3 resonance, for in-plane polarization of the incident x-rays, at 40 K. (a) The spectra in the vertical cascade correspond to different momenta $\mathbf{q}$, transferred along the leg direction x ($b \simeq 4.1$ Å). At low energies (below 0.3 eV), the spinon dispersion clearly emerges [19]. At high energies, orbital excitations dominate the spectra. The arrows identify the corresponding orbital symmetry of the hole in the RIXS final state, with the following energy splittings: $3d_{x^2-y^2}$ at 0 eV (ground state), $3d_{xy}$ at 1.62 eV, $3d_{yz}$ at 1.84 eV, $3d_{xz}$ at 2.03 eV and $3d_{3z^2-r^2}$ at 2.35 eV (see supp. mat. [20] and Tab. S1). (b) RIXS intensity map of the dispersing magnetic excitation. The solid white lines represent the lower and the upper boundaries of the multi-spinon continuum of a S=1/2 Heisenberg chain as in [21]. The dashed white line marks the zero energy loss.

realistic system that is not strictly 1D – and thus for example spin-charge separation is hard to observe – might nevertheless show fractionalization phenomena for some quantum numbers, because of the reduced effective dimensionality of these 'specific' quantum numbers.

In this Letter, we present a combined experimental and theoretical study of the AF anisotropic spin-ladder system $CaCu_2O_3$, cf. Figure 1(c). We look for signatures of spin-orbital separation, which is conceptually analogous to the spin-charge separation described above [3, 4]. To this purpose, we use the resonant inelastic X-ray scattering (RIXS) method that is inherently sensitive to both spin and orbital excitations in cuprate materials [4, 15–18]. Our experimental results together with theoretical model calculations show that spin-orbital fractionalization occurs in $CaCu_2O_3$ for the xz orbital channel – as in an ideal 1D system – while confinement is found for the xy orbital channel. From this result, we convey that diverse orbital symmetries select different degrees of dimensionality in the same system. Moreover, we demonstrate that spin-orbital separation is in general much more robust than spin-charge separation.

$CaCu_2O_3$ is a buckled two-leg spin ladder system composed of corner-sharing CuO_4 plaquettes [Fig. 1(c)] [21, 22] building a system of coupled spin chains parallel to the ladder leg direction x with a Cu-O-Cu angle of 123° along the ladder rung direction y. Just as the well-known 2D cuprate compounds, the ground state configuration of this system is dominated by Cu^{2+} sites in each plaquette with S=1/2 and one hole [9] localized in the $3d_{x^2-y^2}$ orbital.

In order to investigate the spin and orbital dynamics of $CaCu_2O_3$, we performed high-resolution RIXS experiments at the Cu L_3 edge (for more details, cf. Ref. [20]). RIXS enables to map out the dispersion of these excitations across the first Brillouin zone of $CaCu_2O_3$. The experiments were performed using the state-of-the-art SAXES spectrometer [23] installed at the ADRESS beamline of the Swiss Light Source at the Paul Scherre Institute [24].

The RIXS spectra, shown in Figure 2(a) as a vertical cascade for increasing momentum $\mathbf{q}$ transferred along the leg direction x, reveal the presence of strongly dispersing magnetic excitations at energies between 0 eV and 0.5 eV. As indicated in Figure 2(b), their momentum dependence exactly tracks the well-known two-spinon dispersion [19], which is in perfect agreement with previous neutron scattering data [21] and confirms the prevalent 1D nature of the *spin* system. It

is important to point out however that the low-dimensional magnetism of $CaCu_2O_3$ is not reflected in the electronic band structure, which exhibits a pronounced 2D character [22].

In the energy region between 1.5 eV to 2.6 eV we observe orbital excitations, corresponding to the hole [9] in the $3d_{x^2-y^2}$ ground state being excited into a different orbital. We can unambiguously identify these excitations and their energies by combining state-of-the-art quantum chemical calculations [25], which provide realistic estimates for the orbital sequence and the energy splittings, with local calculations of the RIXS cross-section based on single ion model (also referred to as 'local model' in what follows) [26], cf. Figure 2 and [20].

The good agreement of this local model and the experimental data (see suppl. mat. [20]) implies that the orbital excitations into the $3d_{xy}$, $3d_{yz}$, and $3d_{3z^2-r^2}$ are in fact local excitations within a CuO_4 plaquette. However, we observe that in case of the $3d_{xz}$ orbital excitation such local description fails [20] and a propagating *orbiton* is observed, see Figure 3(a): by normalizing the spectra for each $\mathbf{q}$ to its maximum value, a clear momentum dependence is revealed, which consists of a peak shift of 50 meV towards lower energies together with a strong increase in width by almost a factor of 2 for increasing $\mathbf{q}$. Such momentum dispersion is unusual and is in fact a direct indication of spin-orbital separation [3, 4].

In order to verify that spin-orbital separation indeed takes place in $CaCu_2O_3$, we analyzed the spin-orbital dynamics of this material in terms of an effective t–J model. Firstly, the various hopping integrals in the model are determined using density functional theory calculations [27]. Based on these parameters, the effective t–J model [3] for $CaCu_2O_3$ can be constructed, which takes into account all possible superexchange processes with virtual states both on copper and on oxygen sites [28] [see Fig. S2(a) in [20] for details]:

$$\mathcal{H} = -J^{xz}_{\rm leg}\sum_{\langle \mathbf{i},\mathbf{j}\rangle||x,\sigma}(o^{\dagger}_{\mathbf{i}\sigma,xz}o_{\mathbf{j}\sigma,xz}+h.c.)+E^{xz}_0\sum_{\mathbf{i}}n_{\mathbf{i},xz} + J_{\rm rung}\sum_{\langle \mathbf{i},\mathbf{j}\rangle||y}\mathbf{S_i}\cdot\mathbf{S_j}+J_{\rm leg}\sum_{\langle \mathbf{i},\mathbf{j}\rangle||x}\mathbf{S_i}\cdot\mathbf{S_j}, \qquad (1)$$

Here $o^{\dagger}_{\mathbf{i}\sigma}$ creates a hole in the excited $3d_{xz}$ orbital on site $\mathbf{i}$ with spin σ, $n_{\mathbf{i},xz}$ counts the number of holes in the $3d_{xz}$ orbital on site $\mathbf{i}$, and $\mathbf{S_i}$ is a spin $S = 1/2$ operator on site $\mathbf{i}$. The values of the parameters are (for further details see suppl. mat. [20]): the on-site energy of the $3d_{xz}$ orbital excitation $E^{xz}_0 = 2.03$ eV, $J^{xz}_{\rm leg} \simeq 51$ meV for the $3d_{xz}$ orbital superexchange, $J_{\rm leg} \simeq 134$ meV, and $J_{\rm rung} \simeq 11$ meV for the spin superexchange along the leg and along the rung, respectively. We underline that, in the present case, the orbital part of the Hamiltonian has by itself a 1D character: the reason behind this lies in the intrinsic one-dimensionality of the $3d_{xz}$ orbital hopping, which projects solely along x (within the ladder plane) and is completely unaffected by the neighboring ladders [refer to Fig. S2(a) in [20]].

Using Lanczos exact diagonalization, we determined the orbiton spectral function [3] for a single $3d_{xz}$ orbital excitation introduced into the AF ladder, which propagates via the Hamiltonian $\mathcal{H}$ on a 14×2-site ladder lattice. The obtained spectral function is shown in Figure 3(b). In order to facilitate the comparison to the experiment, we normalize the theoretical spectra from Figure 3(b) in the same manner as the experimental data and, after including the instrumental broadening, we present the results in Figure 3(c). Excellent agreement between theory and experiment is observed. This is further confirmed by comparing calculated and measured RIXS responses at two fixed $\mathbf{q}$-values [see Fig. 3(d) and Ref. [20]].

The comparison between the theoretical spectral function in Figure 3(b) with the one obtained for the purely 1D case [3] reveals that the $3d_{xz}$ excitation in $CaCu_2O_3$ exhibits all typical features of spin-orbital separation. This observation is also verified by a detailed analysis of Equation (1), which at energy scales $E \gg J_{\rm rung}$ effectively describes a 1D t–J model giving qualitatively the same spectral response as for the 1D case: having a 1D spin superexchange in the relevant energy scale is in fact a necessary condition for observing fractionalization. Higher dimensional effects become important only at energy scales $E \ll J_{\rm rung}$ [21], which are presently not accessible by the experimentally available RIXS energy resolution.

In a pure 1D system, however, spin-orbital separation manifests also for the $3d_{xy}$ orbital [4]. In the present case instead, the good agreement between the $3d_{xy}$ orbital excitation and the local model [see Fig. 3(d) at around 1.6 eV and Ref. [20]] seems not to support fractionalization for this orbital channel.

To investigate the possibility of spin-orbital fractionalization for the $3d_{xy}$ orbital in $CaCu_2O_3$, a similar analysis to the $3d_{xz}$ case has been done. Two finite paths for the orbital superexchange are found in this case, respectively along the leg and the rung directions as the hole can hop both along x and y [see Fig. S2(b) and [20] for more details]. Nevertheless, it occurs that the hopping along the leg of the ladder is *blocked* due to a peculiar interplay of the Pauli principle and a strong inter-ladder hopping between the $2p_y$ oxygen orbital in the leg and the $3d_{x^2-y^2}$ copper orbital in the neighboring ladder [cf. Fig. S2(b) in [20]]. Because of that, there is a nonzero probability of having the $2p_y$ oxygen orbital in the leg occupied by a hole coming from the neighboring ladder. Considering moreover that the spin of the *traveling* hole from the $3d_{xy}$ orbital is randomly oriented with respect to the spin of the hole in the $2p_y$ orbital, the $3d_{xy}$ hopping along x can be blocked in order not to violate the Pauli principle. In this way the coherent travel of the $3d_{xy}$ orbital along the leg is suppressed: the $3d_{xy}$ orbital excitation thus results in a localized excitation.

The above discussion of the $3d_{xy}$ case shows the importance of the directionality of the orbital motion in establishing the fractionalization phenomenon. We observe spin-orbital separation for the strictly 1D $3d_{xz}$ orbital excitation, though, at the same time the separation is not possible for the $3d_{xy}$ orbital excitation, which is strongly affected by inter-ladder couplings and, hence, behaves as in a 2D system. Note that the spin exchange is effectively 1D through both orbitals.

Finally, to quantitatively compare the spin-orbital separa-

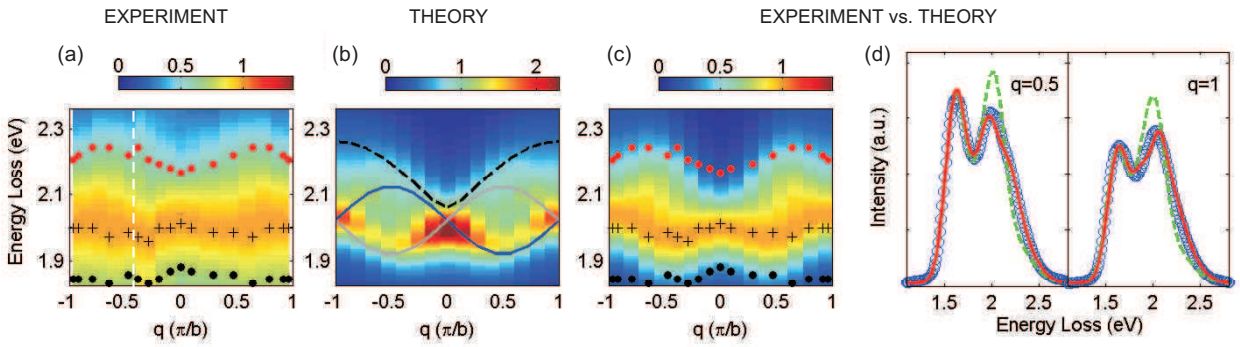

FIG. 3. (Color Online) Experimental and theoretical $3d_{xz}$ orbital dispersion. (a) Color map of the $3d_{xz}$ excitation lineshape. Each RIXS spectrum, in the range 1.8-2.35 eV, is normalized to the $3d_{xz}$ orbital excitation maximum. Data between $-1 <\mathbf{q}< -0.6$ are masked by the close $3d_{yz}$ excitation, which is strong at small $\mathbf{q}$, and have thus been replaced by the mirrored data from the positive $\mathbf{q}$ range. The red and the black dots define the 50% and 35% intensity drop with respect to the peak position. The latter is marked by black crosses. (b) Color map of the exact diagonalization solution for the spin-orbital separation model of $CaCu_2O_3$ [see Eq. (1)]. The lines follow from the orbital-spin separation Ansatz [4] applied to $CaCu_2O_3$ which shows the pure orbiton dispersion (solid line) and the edge (dashed line) of the spinon-orbiton continuum. (c) Normalized theoretical map (b), after broadening by experimental resolution (ΔE=130 meV). Dots are taken from the experimental map (a). (d) Comparison between RIXS experimental lineshapes (blue dots), local model for all orbital excitations (dashed green), and combination of local model (for $3d_{xy}$, $3d_{yz}$ and $3d_{3z^2-r^2}$) and spin-orbital separation model for $3d_{xz}$ (red) [20].

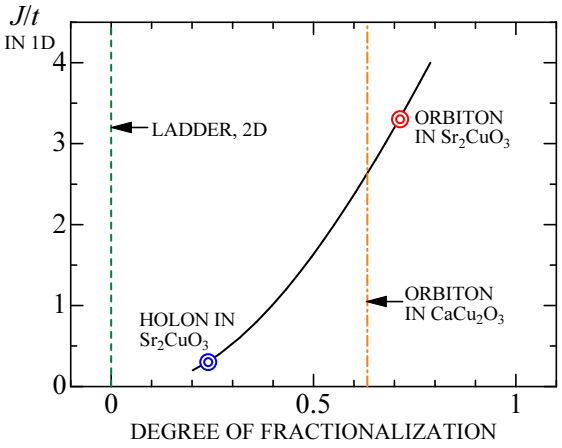

FIG. 4. DOF in various effective t–J models as obtained using the finite size scaled exact diagonalization: for the isotropic ladder t–J model (dashed line, DOF=0), for the anisotropic ladder t–J model Eq. (1) describing the spin-orbital separation in $CaCu_2O_3$ (dot dashed line, DOF$\simeq$0.62). These cases are compared with the ideal 1D t–J model, for which the DOF is calculated as a function of J/t (solid line): $J/t \simeq 0.4$ ($J/t \simeq 3.32$) describes the spin-charge (spin-orbital) separation in Sr_2CuO_3.

tion observed in $CaCu_2O_3$ and in other systems exhibiting spin-orbital or spin-charge separation, we consider here the "degree of fractionalization" (DOF), see Fig. 4: this index expresses how well the spinon and the orbiton or holon are separated from each other, and it ranges from 0 (not separated) to 1 (fully separated). Namely, given the correlation between the orbital (or charge) degree of freedom at site $\mathbf{i} = \mathbf{1}$ and the spin at site $\mathbf{i} + \mathbf{r}$, the DOF is the ratio between the correlation for r=∞ and r=1 (see supp. mat. [20] for more details). Despite the presence of finite inter-leg interaction, we have verified that the spin-orbital separation in $CaCu_2O_3$ is almost as strong as the one in Sr_2CuO_3. Moreover, surprisingly, it is much stronger than the spin-charge separation in the strictly 1D system (Fig. 4). Besides the directional character of the orbital, the reason behind this behavior lies in the different ratio of the spinon / orbiton and spinon / holon velocities. In fact the spinon can move away much quicker from the orbiton than from the holon, i.e. from the quasiparticle at which we look in the experiment, allowing for an 'easier' separation from the spinon in the spin-orbital case.

In conclusion, the present RIXS experiments and theoretical analysis thus demonstrate spin-orbital separation for the $3d_{xz}$ orbital in the anisotropic ladder material $CaCu_2O_3$. The spin-orbital separation is therefore not limited to ideal 1D systems only, but can also survive in systems with the electronic structure being not strictly 1D. This robustness of the fractionalization of the orbital excitation is related to the incidence of three features: (i) the spin excitations in the weakly coupled spin ladder $CaCu_2O_3$ are essentially spinons on the here relevant energy scale, (ii) the motion of the orbital excitation *can be* 1D due to the typical directional character of orbital hoppings (*note* here the $3d_{xy}$ orbiton case for which the hopping is not 1D and the fractionalization does not take place) [29, 30], and (iii) spinons are much faster than orbitons (typically $J_{\rm leg} \gg J^{xz}_{\rm leg}$, cf. [3]) which allows for an easy separation of the two. While the first condition requires 1D spin exchange interactions and is similarly valid for the spin-charge separation phenomenon, the other two conditions are generic to the spin-orbital separation phenomenon only and are rather easy to achieve in a number of systems whose band structure is not 1D. This therefore suggests that the spin-orbital separation can be realized in many other correlated

systems that lack spin-charge separation because they are *not* strictly enough 1D.

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[1] J. Voit, Reports on Progress in Physics **58**, 977 (1995).

[2] 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).

[3] K. Wohlfeld, M. Daghofer, S. Nishimoto, G. Khaliullin, and J. van den Brink, Physical Review Letters **107**, 147201 (2011).

[4] J. Schlappa, K. Wohlfeld, K. J. Zhou, M. Mourigal, M. W. Haverkort, V. N. Strocov, L. Hozoi, C. Monney, S. Nishimoto, S. Singh, A. Revcolevschi, J. Caux, L. Patthey, H. M. Rnnow, J. v. d. Brink, and T. Schmitt, Nature **485**, 82 (2012).

[5] A. Moreno, A. Muramatsu, and J. M. P. Carmelo, Phys. Rev. B **87**, 075101 (2013).

[6] P. W. Anderson, Science **235**, 1196 (1987).

[7] T. Senthil and M. P. A. Fisher, Phys. Rev. Lett. **86**, 292 (2001).

[8] M. Holt, J. Oitmaa, W. Chen, and O. P. Sushkov, Phys. Rev. Lett. **109**, 037001 (2012).

[9] In the introductory paragraph and in the caption of Fig. 1, the term *hole* refers to the hole formed in the S=1/2 spin background, i.e. the removal of the electron with its spin from the $3d_{x^2-y^2}$ orbital. Note that in the rest of the paper this term refers to the hole as defined by 'hole notation' widely used to describe the copper oxides: in that notation the one hole in the ground state $3d_{x^2-y^2}$ orbital arises from the particle-hole transformation to the $3d$ shell filled with 9 electrons (with $3d_{x^2-y^2}$ orbital being half-filled).

[10] G. Martinez and P. Horsch, Phys. Rev. B **44**, 317 (1991).

[11] M. Troyer, H. Tsunetsugu, and T. M. Rice, Phys. Rev. B **53**, 251 (1996).

[12] S. R. White and D. J. Scalapino, Phys. Rev. B **55**, 6504 (1997).

[13] Note that the Luttinger liquid theory, which predicts spin-charge separation in the 1D case, does not apply to the t–J model on the ladder with gapped spin excitations [13].

[14] E. Dagotto and T. M. Rice, Science **271**, 618 (1996).

[15] G. Ghiringhelli, N. B. Brookes, E. Annese, H. Berger, C. Dallera, M. Grioni, L. Perfetti, A. Tagliaferri, and L. Braicovich, Phys. Rev. Lett. **92**, 117406 (2004).

[16] L. Braicovich, L. J. P. Ament, V. Bisogni, F. Forte, C. Aruta, G. Balestrino, N. B. Brookes, G. M. De Luca, P. G. Medaglia, F. M. Granozio, M. Radovic, M. Salluzzo, J. van den Brink, and G. Ghiringhelli, Physical Review Letters **102**, 167401 (2009).

[17] J. Schlappa, T. Schmitt, F. Vernay, V. N. Strocov, V. Ilakovac, B. Thielemann, H. M. Rønnow, S. Vanishri, A. Piazzalunga, X. Wang, L. Braicovich, G. Ghiringhelli, C. Marin, J. Mesot, B. Delley, and L. Patthey, Physical Review Letters **103**, 047401 (2009).

[18] L. J. P. Ament, M. van Veenendaal, T. P. Devereaux, J. P. Hill, and J. van den Brink, Review of Modern Physics **83**, 705 (2011).

[19] V. Bisogni, S. Kourtis, C. Monney, K. Zhou, R. Kraus, C. Sekar, V. Strocov, B. Buechner, J. v. d. Brink, L. Braicovich, T. Schmitt, M. Daghofer, and J. Geck, Phys. Rev. Letters **112**, 147401 (2014).

[20] See Supplementary Material [url], which includes Refs. [31-38].

[21] B. Lake, A. M. Tsvelik, S. Notbohm, D. Alan Tennant, T. G. Perring, M. Reehuis, C. Sekar, G. Krabbes, and B. Buchner, Nature Physics **6**, 50 (2010).

[22] E. Bordas, C. de Graaf, R. Caballol, and C. J. Calzado, Phys. Rev. B **71**, 045108 (2005).

[23] G. Ghiringhelli, A. Piazzalunga, C. Dallera, G. Trezzi, L. Braicovich, T. Schmitt, V. N. Strocov, R. Betemps, L. Patthey, X. Wang, and M. Grioni, Review of Scientific Instruments **77**, 113108 (2006).

[24] V. N. Strocov, T. Schmitt, U. Flechsig, T. Schmidt, A. Imhof, Q. Chen, J. Raabe, R. Betemps, D. Zimoch, J. Krempasky, A. Piazzalunga, X. Wang, M. Grioni, and L. Patthey, J. Synchrotron Rad. **17**, 631 (2010).

[25] H.-Y. Huang, N. A. Bogdanov, L. Siurakshina, P. Fulde, J. van den Brink, and L. Hozoi, Physical Review B **84**, 235125 (2011).

[26] M. Moretti Sala, V. Bisogni, C. Aruta, G. Balestrino, H. Berger, N. B. Brookes, G. M. De Luca, D. Di Castro, M. Grioni, M. Guarise, P. G. Medaglia, F. Miletto Granozio, M. Minola, P. Perna, M. Radovic, M. Salluzzo, T. Schmitt, K. J. Zhou, L. Braicovich, and G. Ghiringhelli, New Journal of Physics **13**, 043026 (2011).

[27] K. Koepernik and H. Eschrig, Phys. Rev. B **59**, 1743 (1999).

[28] K. Wohlfeld, S. Nishimoto, M. W. Haverkort, and J. van den Brink, Phys. Rev. B **88**, 195138 (2013).

[29] A. B. Harris, T. Yildirim, A. Aharony, O. Entin-Wohlman, and I. Y. Korenblit, Phys. Rev. Lett. **91**, 087206 (2003).
[30] K. Wohlfeld, M. Daghofer, A. M. Oleś, and P. Horsch, Phys. Rev. B **78**, 214423 (2008).
[31] C. Sekar, G. Krabbes, and A. Teresiak, Journal of Crystal Growth **273**, 403 (2005).
[32] R. Neudert, S.-L. Drechsler, J. Málek, H. Rosner, M. Kielwein, Z. Hu, M. Knupfer, M. S. Golden, J. Fink, N. Nücker, M. Merz, S. Schuppler, N. Motoyama, H. Eisaki, S. Uchida, M. Domke, and G. Kaindl, Phys. Rev. B **62**, 10752 (2000).
[33] L. Hozoi *et al. priv. comm.*
[34] A. C. Walters, T. G. Perring, J. Caux, A. T. Savici, G. D. Gu, C. Lee, W. Ku, and I. A. Zaliznyak, Nature Physics **5**, 867 (2009).
[35] T. K. Kim, H. Rosner, S.-L. Drechsler, Z. Hu, C. Sekar, G. Krabbes, J. Málek, M. Knupfer, J. Fink, and H. Eschrig, Physical Review B **67**, 024516 (2003).
[36] K. Wohlfeld, A. M. Oleś, and G. A. Sawatzky, Phys. Rev. B **81**, 214522 (2010).
[37] H. Fujisawa, T. Yokoya, T. Takahashi, S. Miyasaka, M. Kibune, and H. Takagi, Phys. Rev. B **59**, 7358 (1999).
[38] B. J. Kim, H. Koh, E. Rotenberg, S.-J. Oh, E. Eisaki, N. Motoyama, S. Uchida, T. Tohyama, S. Maekawa, and C. Shen, Z.-X.and Kim, Nature Physics **2**, 397 (2006).