Experimental investigation of the origin of the crossover temperature in cuprate superconductors via dc magnetic susceptibility

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We investigate the crossover temperature T^* as a function of doping in $(\mathrm{Ca}_x\mathrm{La}_{1-x})(\mathrm{Ba}_{1.75-x}\mathrm{La}_{0.25+x})\mathrm{Cu}_3\mathrm{O}_y$, where the maximum T_c ($T_c^{\max}$) varies continuously by 30% between families (x) with minimal structural changes. T^* is determined by dc-susceptibility measurements. We find that T^* scales with the maximum Néel temperature $T_N^{\max}$ of each family. This result strongly supports a magnetic origin of T^* and indicates that three dimensional interactions play a role in its magnitude.

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Free electrons do not have high-temperature crossovers such as a pseudogap (PG), spin gap (SG), or development of antiferromagnetic (AFM) correlations. In the cuprates, all of these exist, yet the interactions that lead to them are not completely clear. Nevertheless, the crossovers occur at a temperature T^* which is much higher than T_c, and closer to the three-dimensional (3D) ordering temperature of the parent compound in the AFM state. Therefore, it is speculated that T^* emerges from AFM fluctuations, and that the crossovers are intimately linked, namely, the interaction responsible for one might be responsible for all.[1–3] Therefore, it is crucial to test the possibility of correlations between T^* of a particular system and its magnetic properties, such as the Néel temperature T_N of the parent compound, or its constituents, the in- and out-of-plane Heisenberg coupling constant J and $J_\perp$, respectively. This is the motivation of the work presented here. We provide experimental evidence that strongly supports a magnetic origin for T^*. Moreover, we show that T^* stems from 3D interactions, similar to the Néel order, involving both J and $J_\perp$.

We investigate the origin of the T^* by studying its variations as a function of the compound's magnetic properties, where small chemical changes are an implicit parameter. The variations in the magnetic properties are achieved by using four different families of the $(\mathrm{Ca}_x\mathrm{La}_{1-x})(\mathrm{Ba}_{1.75-x}\mathrm{La}_{0.25+x})\mathrm{Cu}_3\mathrm{O}_y$ (CLBLCO) system, having the $\mathrm{YBa}_2\mathrm{Cu}_3\mathrm{O}_y$ (YBCO) structure, with $x=0.1,\ldots,0.4$. The phase diagram of the CLBLCO families is shown in Fig. 1(a). T_c was measured by resistivity[4] and the spin-glass temperature T_g (Ref. 5) and T_N (Ref. 6) by muon spin relaxation. Despite the rich phase diagram, the different CLBLCO families have negligible structural differences. All compounds are tetragonal, and there is no oxygen chain ordering as in YBCO.[4] The hole concentration in the CuO_2 planes does not depend on x.[7,8] The difference in the unit-cell parameters a and $c/3$ between the two extreme families ($x=0.1$ and 0.4) is 1%.[4] Thus, variations in $T_c^{\max}$ due to variations in ionic radii are not relevant.[9] The level of disorder, as detected by Cu and Ca nuclear magnetic resonance, is also identical for the different families.[8,10] In fact, the only strong variation between families noticed at present is the in-plane oxygen buckling angle.[11] This property can modify the intraplane near- and next-near-neighbor hopping, or interplane hopping, which controls the magnetic interaction strengths J and $J_\perp$.[12] The strong magnetic and superconducting variations of the CLBLCO system, accompanied by minimal structural changes, make it ideal for a correlation study between T^* and magnetism.

In this project we determine T^* using temperature-dependent magnetization measurements. In Fig. 2(a) we present raw data from four samples of the $x=0.2$ family with different doping levels. At first glance the data contain only

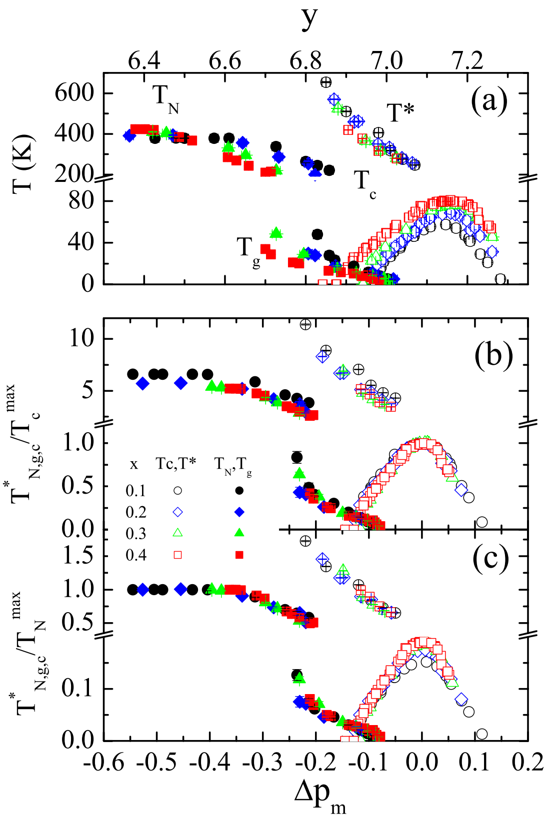

FIG. 1. (Color online) (a) The phase diagram of $(\mathrm{Ca}_x\mathrm{La}_{1-x})(\mathrm{Ba}_{1.75-x}\mathrm{La}_{0.25+x})\mathrm{Cu}_3\mathrm{O}_y$. (b) The critical temperatures are normalized by the maximum critical temperature $T_c^{\max}$ of each family (x), and y is replaced by mobile hole density variation Δp_m (see text). (c) The same as (b) but the normalization is by one number per family, referred to as $T_N^{\max}$, which makes all $T_N(x,y)$ curves collapse to one.

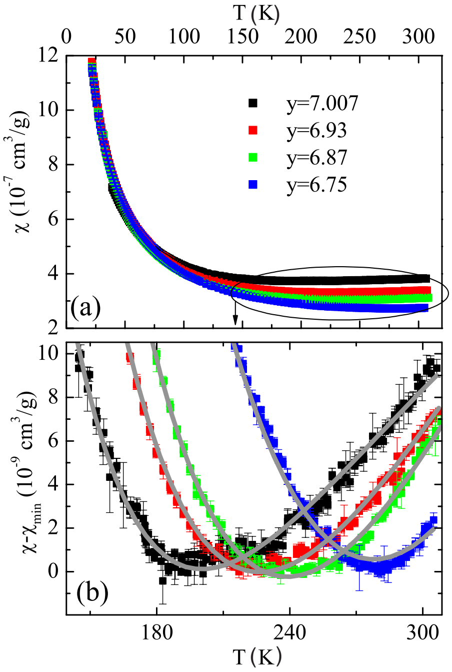

FIG. 2. (Color online) (a) Raw data from four samples of the $x=0.2$ family with a different doping levels. (b) Zoom-in on the data in the ellipse of panel (a) after the minimum value of χ is subtracted. The solid lines are fits to Eq. (1).

two features: A Curie-Weiss (CW) type increase of χ at low temperatures, and a nonzero baseline at high-temperature χ_{300}. This baseline increases with increasing y. The CW term could be a result of isolated spins, impurities, or spins on the chain layer. However, as we will show shortly, there is much more to it. The baseline shift could be a consequence of variations in the core and Van Vleck (CVV) electron contribution or an increasing density of states at the Fermi level.

A zoom-in on the high-temperature region, marked by the ellipse, reveals a third feature in the data: a minimum point of χ. To present this minimum clearly, we subtracted from the raw data the minimal value of the susceptibility $\chi_{\min}$ for each sample, and plotted the result on a tighter scale in Fig. 2(b). The χ minimum is a result of decreasing susceptibility upon cooling from room temperature, followed by an increase in the susceptibility due to the CW term at low T. This phenomenon was previously noticed by Johnston *et al.* in YBCO,[13] and Johnston and Nakano *et al.* in $La_{2-x}Sr_xCuO_4$ (LSCO).[14,15] The minimum point moves to higher temperatures with decreasing oxygen level as expected from T^*. There are three possible reasons for this decreasing susceptibility: (I) increasing AFM correlations upon cooling,[14] (II) opening of a SG where excitations move from $q=0$ to the AFM wave vector,[16] or (III) disappearing density of states at the Fermi level as parts of the Fermi arc are being gapped out when the PG opens as T/T^* decreases.[17]

In order to determine the T^* we fit the data to a three component function

$$\chi = \frac{C_1}{T+\theta} + \frac{C_2}{\cosh\left(\frac{T^*}{T}\right)} + C_3. \tag{1}$$

The data are fitted without any restriction on the parameters. The quality of the fit is demonstrated in Fig. 2(b) by the solid lines; it captures the data precisely with barely observable deviations at very low doping, where T^* is at the edge of our measurement window. Of course on the broader scale of Fig. 2(a) there are no differences between the fit function (not shown) and data. At dopings higher than $y \sim 7.1$ the CW term overwhelms the crossover term due to its low T^*, and the χ minimum is no longer detectable. At dopings lower than $y \sim 6.85$ the χ minimum is out of the measurement window. These samples are not analyzed. The C's determined by the fits (not shown) are found to behave smoothly and monotonically as a function of doping and family. C_2 and C_3 have the same order of magnitude as χ_{300}. The function $\cosh^{-1}(T^*/T)$ was chosen only because it fits the data best. However, we will analyze only the scaling properties of T^* for which the absolute value is not relevant.

As for the amplitudes, it is most natural to relate C_1 to the weight of an impurity related CW contribution, C_2 to the crossover electrons, and C_3 to free electrons and CVV susceptibility. This division is based on the 2D Heisenberg model that predicts a decreasing susceptibility with decreasing temperature.[18] However, the situation at hand is closer to the *t-J* model for which the susceptibility is calculated by high-temperature series expansion, and its behavior at $T \to 0$ is not known.[19] Therefore, it is conceivable that the division of χ into impurities, crossover, and free electron and CVV contribution is artificial, that there is no impurities contribution, and that the susceptibility simply has two energy scales θ and T^*. We are mostly interested in these two parameters and the evolution of χ_{300} with doping.

In Fig. 3(a) we plot χ_{300} for the different families. It is clearly increasing as a function of y. The expected contribution from core electrons, taken from the standard tables,[20] is also expected to increase, but with a variation that is smaller in an order of magnitude. The Van Vleck contribution is also taken as a constant.[14] Therefore, the increasing of χ_{300} with doping must result from either an increasing density of states at the Fermi level or decreasing correlation length ξ. At very low doping, near the AFM phase, there are some differences between the families; the χ_{300} is higher for the $x=0.4$ family. However, at the doping level in which superconductivity appears, χ_{300} is similar to all families. The density-of-states scenario is consistent with previous claims that the doping level in CLBLCO is x independent, at least in the superconducting region.[4,8] The correlation length scenario is not consistent with our previous claims that $J(x)$ varies by 30% between families[6] since ξ has exponential J dependence.[18]

Since CLBLCO obeys the Uemura relation $T_c \propto n_s$ in the entire doping range,[21] where n_s is the superconducting carrier density, we conclude that the proportionality constant varies between families, or that not all holes turn superconducting. This conclusion reinforces our previous claims that $T_c \simeq J(x) n_s$,[6] and that in CLBLCO not all the holes condense to superfluid.[8]

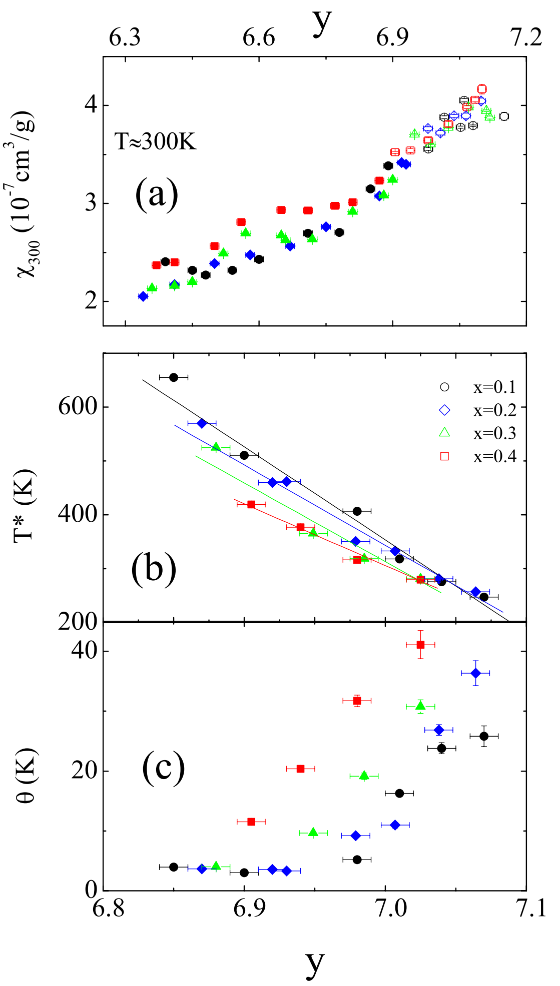

FIG. 3. (Color online) (a) The susceptibility of all the samples at $T=300$ K. (b) T^* as a function of doping and families. The solid lines are guides to the eye. (c) The Curie-Weiss temperature θ as a function of doping and families.

The T^* parameter obtained from the fits is depicted in Fig. 3(b) on a tight scale (the solid lines are a guide to the eye), and as part of the full phase diagram in Fig. 1(a). It behaves like the well-known PG or SG T^* measured by other techniques on a variety of superconductor samples.[1] At the same time, a decrease of T^* with doping is consistent with the AFM correlation picture as a progressive departure from the Mott insulator. More importantly, a small but clear family dependence of T^* is seen. At first glance it appears that T^* has anticorrelation with $T_c^{\max}$ or the maximum T_N ($T_N^{\max}$). The $x=0.4$ family, which has the highest $T_c^{\max}$ and $T_N^{\max}$, has the lowest T^*, and vice versa for the $x=0.1$ family.

However, this conclusion is reversed if instead of plotting the T^* as a function of oxygen level, it is properly normalized and plotted as a function of mobile hole variation Δp_m. By mobile holes we mean holes that *do* turn superconducting as discussed above. Δp_m is defined in two steps: (I) The chemical doping measured from optimum, $\Delta y=y-y_0$, is obtained for each compound (y_0 is the oxygen level at $T_c^{\max}$). (II) Δy is multiplied by a different constant per family $K(x)$, namely, $\Delta p_m=K(x)\Delta y$.[6] The K's are chosen so that the superconductor domes, normalized by $T_c^{\max}$ of each family, collapse onto each other; $K=0.76,0.67,0.54,0.47$ with 5% accuracy for $x=0.1\ldots0.4$, see Fig. 1(b).

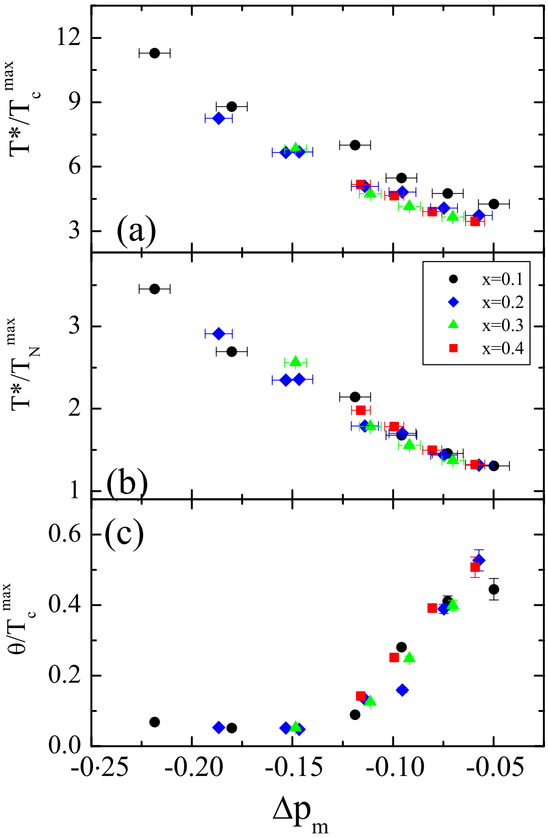

FIG. 4. (Color online) (a) $T^*/T_c^{\max}$, (b) $T^*/T_N^{\max}$, and (c) the Curie-Weiss temperature θ, as a function of mobile hole variation Δp_m (see text).

We examine two possible normalizations of the critical temperatures: by $T_c^{\max}$ or $T_N^{\max}$. In Fig. 1(b) we present *all* critical temperatures, normalized by $T_c^{\max}$, as a function of Δp_m. As expected, all domes scale onto each other. So do the glass temperatures T_g. T_N for $x=0.2$, 0.3, and 0.4 families also collapse nicely. However, T_N for the $x=0.1$ family does not. In Fig. 4(a) we zoom in on the $T^*/T_c^{\max}$, as a function of Δp_m. The same problem is observed here as well. Next, we normalize all critical temperatures by $T_N^{\max}$ as shown in Fig. 1(c). The values of $T_N^{\max}$ are chosen so that the $T_N(\Delta p_m)/T_N^{\max}$ curves collapse onto each other, and are 379, 391.5, 410, and 423 K for the $x=0.1\ldots0.4$ families, respectively. Therefore, $T_N^{\max}$ should be interpreted as the extrapolation of T_N to the lowest Δp_m in Fig. 1(c). In this case, the T_g curves of all family also collapse, but the T_c domes do not. In Fig. 4(b), we zoom-in on the $T^*/T_N^{\max}$ as a function of Δp_m. Now all the normalized T^* curves overlap. Thus T^* of each family scales better with $T_N^{\max}$ than with $T_c^{\max}$. This is our main finding.

As for the CW parameter θ, although we did not expect any correlations between this parameter and x or y, we found an interesting trend shown in Fig. 3(c). In the antiferromagnetic region $\theta\sim0$. As we go to higher doping levels, this magnetic energy scale increases. This trend was previously observed by Bobroff *et al.*[22] It is also clear that there are systematic variations of θ between the families. The $x=0.4$

has the strongest θ, and the $x=0.1$ the weakest. In fact, in Fig. 4(c) we plot $\theta/T_c^{\max}$ as a function of Δp_m. All data points collapse to a single curve. Once again we find that the proper doping parameter is Δp_m, and not oxygen level y, and the same energy scales control both θ and $T_c^{\max}$. These trends suggest that θ has nothing to do with impurities, as already hinted above.

Our results bare important information on T^*. When we normalize T^* by $T_c^{\max}$, we are actually normalizing by the in-plane energy scale of each family $J(x)$.[6] If the crossover was only a result of magnetic interaction between the spins in the planes (2D), T^* should have scaled with $T_c^{\max}$. The imperfect normalization by $T_c^{\max}$, demonstrated in Fig. 4(a), contradicts this possibility. When we normalize T^* by $T_N^{\max}$, we are taking into account the coupling between the planes $J_\perp$ (3D) as well. The success of the normalization by $T_N^{\max}$, shown in Fig. 4(b), implies that T^* is governed by 3D magnetic interaction.

The importance of $J_\perp$ was previously discussed in Ref. 3. Our finding is also consistent with the concept of a 3D to 2D crossover above T_c in which planes decouple from each other.[23] Finally, it is consistent with Nakano *et al.* where by comparing LSCO to $Bi_2Sr_2CaCu_2O_{8+\delta}$ a proportionality between T^* and an unspecified magnetic energy scale is found.[24]

To summarize, after converting oxygen level y to mobile holes variations Δp_m, we find that the crossover temperature T^* measured by susceptibility in the CLBLCO system is proportional to $T_N^{\max}$. T_N is set by both in- and out-of-plane coupling constants that are determined by in- and out-of-plane hoppings. This result suggests a 3D magnetic origin for T^*. In addition, the CW-like term of the susceptibility is not a result of impurities. It might be an intrinsic property of doped CuO_2 planes at low temperatures.

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[1] T. Timusk and B. Statt, Rep. Prog. Phys. **62**, 61 (1999); M. R. Norman, D. Pines, and C. Kallin, Adv. Phys. **54**, 715 (2005); P. A. Lee, N. Nagaosa, and X.-G. Wen, Rev. Mod. Phys. **78**, 17 (2006).

[2] N. Harrison, R. D. McDonald, and J. Singleton, Phys. Rev. Lett. **99**, 206406 (2007).

[3] A. J. Millis and H. Monien, Phys. Rev. Lett. **70**, 2810 (1993); M. U. Ubbens and P. A. Lee, Phys. Rev. B **50**, 438 (1994); L. B. Ioffe, A. I. Larkin, A. J. Millis, and B. L. Altshuler, JETP Lett. **59**, 65 (1994).

[4] D. Goldschmidt, G. M. Reisner, Y. Direktovitch, A. Knizhnik, E. Gartstein, G. Kimmel, and Y. Eckstein, Phys. Rev. B **48**, 532 (1993).

[5] A. Kanigel, A. Keren, Y. Eckstein, A. Knizhnik, J. S. Lord, and A. Amato, Phys. Rev. Lett. **88**, 137003 (2002).

[6] R. Ofer, G. Bazalitsky, A. Kanigel, A. Keren, A. Auerbach, J. S. Lord, and A. Amato, Phys. Rev. B **74**, 220508(R) (2006).

[7] O. Chmaissem, Y. Eckstein, and C. G. Kuper, Phys. Rev. B **63**, 174510 (2001).

[8] A. Keren, A. Kanigel, and G. Bazalitsky, Phys. Rev. B **74**, 172506 (2006).

[9] X.-J. Chen and H. Su, Phys. Rev. B **71**, 094512 (2005).

[10] Sebastien Marchand, Ph.D. thesis, Universite Paris 6.

[11] R. Ofer, A. Keren, O. Chmaissem, and A. Amato, arXiv:0806.4736 (unpublished).

[12] J. Zaanen and G. A. Sawatzky, Can. J. Phys. **65**, 1262 (1987); E. Pavarini, I. Dasgupta, T. Saha-Dasgupta, O. Jepsen, and O. K. Andersen, Phys. Rev. Lett. **87**, 047003 (2001); X. J. Chen and H. Q. Lin, Phys. Rev. B **69**, 104518 (2004).

[13] D. C. Johnston, A. J. Jacobson, J. M. Newsam, J. T. Lewandowski, D. P. Goshorn, D. Xie, and B. Yelon, *Chemistry of High-Temperature Superconductors* (American Chemical Society, Washington, DC, 1987), p. 149.

[14] D. C. Johnston, Phys. Rev. Lett. **62**, 957 (1989).

[15] T. Nakano, M. Oda, C. Manabe, N. Momono, Y. Miura, and M. Ido, Phys. Rev. B **49**, 16000 (1994).

[16] J. Rossat-Mignod, L. P. Regnault, C. Vettier, P. Bourges, P. Burlet, J. Bossy, J. Y. Henry, and G. Lapertot, Physica B (Amsterdam) **180-181**, 383 (1992); L. P. Regnault, P. Bourges, P. Burlet, J. Y. Henry, J. Rossat-Mignod, Y. Sidis, and C. Vettier, Physica C, **235-240**, 59 (1994); M.-H. Julien, P. Carretta, M. Horvatić, C. Berthier, Y. Berthier, P. Ségransan, A. Carrington, and D. Colson, Phys. Rev. Lett. **76**, 4238 (1996).

[17] A. Kanigel, M. R. Norman, M. Randeria, U. Chatterjee, S. Souma, A. Kaminski, H. M. Fretwell, S. Rosenkranz, M. Shi, T. Sato, T. Takahashi, Z. Z. Li, H. Raffy, K. Kadowaki, D. Hinks, L. Ozyuzer, and J. C. Campuzano, Nat. Phys. **2**, 447 (2006).

[18] A. Auerbach and D. P. Arovas, Phys. Rev. Lett. **61**, 617 (1988).

[19] R. R. P. Singh and R. L. Glenister, Phys. Rev. B **46**, 11871 (1992).

[20] P. W. Selwood, *Magnetochemistry*, 2nd ed. (Interscience, New York, 1956), p. 78.

[21] A. Keren, A. Kanigel, J. S. Lord, and A. Amato, Solid State Commun. **126**, 39 (2003); A. Kanigel, A. Keren, A. Knizhnik, and O. Shafir, Phys. Rev. B **71**, 224511 (2005).

[22] J. Bobroff, W. A. MacFarlane, H. Alloul, P. Mendels, N. Blanchard, G. Collin, and J. F. Marucco, Phys. Rev. Lett. **83**, 4381 (1999).

[23] E. Berg, E. Fradkin, E. A. Kim, S. A. Kivelson, V. Oganesyan, J. M. Tranquada, and S. C. Zhang, Phys. Rev. Lett. **99**, 127003 (2007); Q. Li, M. Hücker, G. D. Gu, A. M. Tsvelik, and J. M. Tranquada, *ibid.* **99**, 067001 (2007).

[24] T. Nakano, N. Momono, M. Oda, and M. Ido, J. Phys. Soc. Jpn. **67**, 2622 (1998).