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 \((\mathrm{Ca}_{x}\mathrm{La}_{1-x})(\mathrm{Ba}_{1.75-x}\mathrm{La}_{0.25+x})\mathrm{Cu}_2\mathrm{O}_y\), where the maximum \( T_c \) (\( T_c^{\mathrm{max}} \)) 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 \( T_N^{\mathrm{max}} \) 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 \( J_a \) and \( J_b \), 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 \( J_a \) and \( J_b \).

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 \((\mathrm{Ca}_{x}\mathrm{La}_{1-x})(\mathrm{Ba}_{1.75-x}\mathrm{La}_{0.25+x})\mathrm{Cu}_2\mathrm{O}_y\) (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 \( \mathrm{CuO}_2 \) planes does not depend on \( x \).\(^7\)^\(^8\) The difference in the unit-cell parameters \( a \) and \( c/3 \) between the two extreme families \((x=0.1 \text{ and } 0.4)\) is \( 1\% \).\(^4\) Thus, variations in \( T_c^{\mathrm{max}} \) 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.\(^8\)^\(^\text{Ref.} \)\(^5\)\(^\text{Ref.} \)\(^6\) In fact, the only strong variation between families noticed at present is the in-plane oxygen buckling angle.\(^1\)\(^1\) This property can modify the intraplane near- and next-near-neighbor hopping, or interplane hopping, which controls the magnetic interaction strengths \( J \) and \( J_b \).\(^1\)\(^2\) 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 \((\mathrm{Ca}_{x}\mathrm{La}_{1-x})(\mathrm{Ba}_{1.75-x}\mathrm{La}_{0.25+x})\mathrm{Cu}_2\mathrm{O}_y\). (b) The critical temperatures are normalized by the maximum critical temperature \( T_c^{\mathrm{max}} \) of each family \((x)\), and \( y \) is replaced by mobile hole density variation \( \Delta p_m \) (see text). (c) The same as (b) but the normalization is by one number per family, referred to as \( T_N(x,y) \), which makes all \( T_N(x,y) \) curves collapse to one.

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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 $\chi$ minimum is no longer detectable. At dopings lower than $y \sim 6.85$ the $\chi$ 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 $\chi_{300}$. 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. 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. Therefore, it is conceivable that the division of $\chi$ 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 $\theta$ and $T'$. We are mostly interested in these two parameters and the evolution of $\chi_{300}$ with doping.

In Fig. 3(a) we plot $\chi_{300}$ for the different families. It is clearly increasing as a function of $y$. The expected contribution from core electrons, taken from the standard tables, 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. Therefore, the increasing of $\chi_{300}$ with doping must result from either an increasing density of states at the Fermi level or decreasing correlation length $\xi$. At very low doping, near the AFM phase, there are some differences between the families; the $\chi_{300}$ is higher for the $x=0.4$ family. However, at the doping level in which superconductivity appears, $\chi_{300}$ 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. The correlation length scenario is not consistent with our previous claims that $J(x)$ varies by 30% between families since $\xi$ has exponential $J$ dependence.

Since CLBLCO obeys the Uemura relation $T_c \sim n_x$, in the entire doping range, where $n_x$ 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 $T_c \approx J(x)n_x$, and that in CLBLCO not all the holes condense to superfluid.
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. 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 $T_c^\text{max}$ or the maximum $T_N$ ($T_N^\text{max}$). The $x=0.4$ family, which has the highest $T_c^\text{max}$ and $T_N^\text{max}$, 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 $T^*/T_N^\text{max}$ or $T^*/T_N^\text{max}$. In Fig. 1(b) we present all critical temperatures, normalized by $T_c^\text{max}$, as a function of $\Delta \rho_m$. 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 $T^*/T_c^\text{max}$, as a function of $\Delta \rho_m$. The same problem is observed here as well. Next, we normalize all critical temperatures by $T_N^\text{max}$ as shown in Fig. 1(c). The values of $T_N^\text{max}$ are chosen so that the $T_N(\Delta \rho_m)/T_N^\text{max}$ curves collapse onto each other, and are 379, 391.5, 410, and 423 K for the $x=0.1...0.4$ families, respectively. Therefore, $T_N^\text{max}$ should be interpreted as the extrapolation of $T_N$ to the lowest $\Delta \rho_m$ in Fig. 1(c). In this case, the $T_g$ curves of all family also collapse, but the $T_c$ dome does not. In Fig. 4(b), we zoom-in on the $T^*/T_N^\text{max}$ as a function of $\Delta \rho_m$. Now all the normalized $T^*$ curves overlap. Thus $T^*$ of each family scales better with $T_N^\text{max}$ than with $T_c^\text{max}$. This is our main finding.

As for the CW parameter $\theta$, 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 $\theta \approx 0$. As we go to higher doping levels, this magnetic energy scale increases. This trend was previously observed by Bobroff et al. It is also clear that there are systematic variations of $\theta$ between the families. The $x=0.4...
shown in Fig. 4 suggests that we are taking into account the coupling between the planes $T_c$ in the planes was only a result of magnetic interaction between the spins hinted above. When we normalize $J$ in-plane energy scale of each family $\Delta p_{\text{in-plane}}$, contradicts this possibility. When we normalize $T^*$ by $T^*_{\text{max}}$, we are taking into account the coupling between the planes $J_\perp$ (3D) as well. The success of the normalization by $T^*_{\text{max}}$, 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. Finally, it is consistent with Nakano et al. where by comparing LSCO to B1$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ a proportionality between $T^*$ and an unspecified magnetic energy scale is found.

To summarize, after converting oxygen level $y$ to mobile holes variations $\Delta p_{\text{in-plane}}$, we find that the crossover temperature $T^*$ measured by susceptibility in the CLBLCO system is proportional to $T^*_{\text{max}}$. $T_N$ 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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