

Self-generated electronic heterogeneity and quantum glassiness in the high-temperature superconductors

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We present a systematic study of the spin and charge dynamics of copper oxide superconductors as a function of carrier concentration x . Our results portray a coherent physical picture, which reveals a quantum critical point at optimum doping ($x=x_{opt}$), and the formation of a glassy state at $x<x_{opt}$. This mechanism is argued to arise as an intrinsic property of doped Mott insulators, and therefore to be largely independent of material quality and level of disorder.

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I. INTRODUCTION

Many interesting materials ranging from magnetorestrictive manganite films¹ and field-effect transistors,^{2,3} to unconventional low-dimensional superconductors,⁴ find themselves close to the metal-insulator transition. In this regime, competition between several distinct ground states^{4,5} produces unusual behavior, displaying striking similarities in a number of different systems. Electronic heterogeneity^{1,6} emerges, giving rise to “mesoscopic” coexistence of different ordered phases. Typically, a large number of possible configurations of these local regions have comparable energies, resulting in slow relaxation, aging, and other signatures of glassy systems. Because the stability of such ordering is controlled by doping-dependent quantum fluctuations^{7,8} introduced by itinerant carriers, these systems can be regarded as prototypical quantum glasses—a paradigm of strongly correlated matter.

Here we report the emergence, with the first added carriers, and consequent evolution of dynamical heterogeneity and glassy behavior across the phase diagram of the high-transition-temperature (T_c) superconductors (HTS). Based on data of the spin and charge dynamics, we draw a phase diagram (Fig. 1) and propose that self-generated glassiness both in the spin and charge sector^{5,6} may be a key feature necessary to understand many of the unconventional properties of both the superconducting and the normal state.

II. GLASSINESS IN THE PSEUDOGAP PHASE

In the archetypal HTS, $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (LSCO), the parent two-dimensional (2D) antiferromagnetic insulator (AFI) La_2CuO_4 displays a sharp peak in the magnetic susceptibility at the Néel temperature $T_N=300$ K. T_N decreases with hole doping and the transition width broadens (Fig. 2, upper panel). Concurrently, we find that with the first added holes ($x=0.01$) a second freezing transition (T_F) emerges at lower temperatures (Fig. 2, lower panel), as if the added holes freeze separately. Experimental evidence from various techniques, and on several HTS, show a systematic evolution of T_F with doping ($x>0.01$), as shown in Fig. 3 for results

obtained by μSR (more data with closer spaced dopings and from different experimental techniques can be found in the literature^{9–14}). Although the present experimental resolution throughout the literature leaves some doubts as to whether T_F increases strongly with carrier concentration or is weakly doping dependent, it is certain that it emerges immediately with the first added carriers. The peak in the spin-lattice relaxation rate (Fig. 2, lower panel), for example, is one of the indications for the short-range nature of this second freezing.

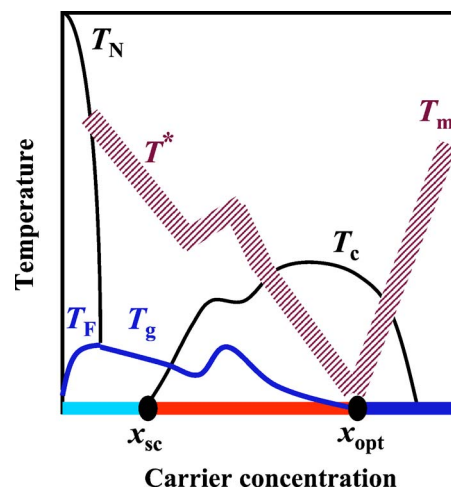


FIG. 1. (Color online) Schematic plot indicating the three major ground-state regimes in the phase diagram of the archetypal HTS. T_N is the Néel temperature, T_F and T_g the onset of short-range freezing to an electronic glass, and T_c the superconducting transition temperature. At $x<x_{sc}$ the material is a glassy insulator. At $x_{sc}<x<x_{opt}$ a microscopically inhomogeneous conducting glassy state emerges, with intercalated superconducting and magnetic regions. At $x=x_{opt}$ the system experiences a quantum glass transition and at $x>x_{opt}$ the material transforms into a homogeneous metal with BCS-like superconducting properties. The superfluid density is maximum at $x=x_{opt}$. The crossover scales T^* and T_m characterizing normal-state transport (see text for details), vanish at the quantum glass transition.

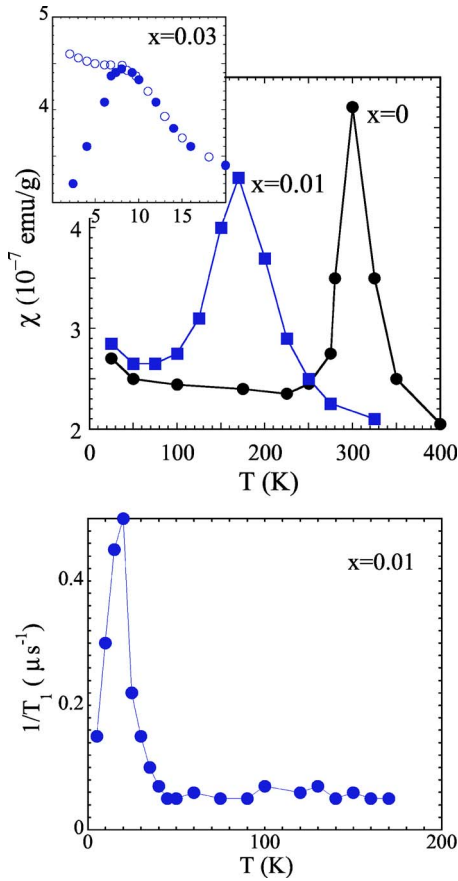


FIG. 2. (Color online) The upper panel depicts the temperature dependence of the susceptibility for LSCO ($x=0$ and 0.01) single crystals with $H \parallel c$. The associated inset is data for $x=0.03$ showing the transformation of the material to a glass. The lower panel depicts a peak in the spin-lattice relaxation $1/T_1$ data obtained by zero field μ SR on LSCO ($x=0.01$).

In the high- T_c cuprates we have the flexibility of studying the physical characteristics of this short-range freezing by adding just 1 percent more carriers to the system. As shown in Fig. 2 (inset, upper panel) and Fig. 3 (upper panel), with $x=0.03$ the short-range order has been fully exposed, and we can now perform thermodynamic studies to gain further insight into its nature. Figure 2 (inset, upper panel) indicates that at $x > 0.02$, $T_N=0$ but spin freezing persists below a sharply defined temperature T_g .¹⁵ Here, the low-field susceptibility displays a well-defined cusp, and a thermal hysteresis is observed at $T < T_g$ (Fig. 2, upper panel), where the material displays scaling, memory effects like “traditional” glasses, and is described by an Edwards-Anderson order parameter.¹⁶ Although the signal of this glassy order is somewhat rounded at T_g , and the field-cooled curve shows a small but distinct increase with decreasing temperature (both suggestive of a cluster glass), as compared to other traditional spin-glass systems such as Au-Fe or Cu-Mn, the basic characteristics of the glass order agree very well. The short-range freezing may also be responsible for some of the puzzling properties of these compounds, in particular in the region below optimal doping. Notably, it is in this doping range that a pseudogap phase develops,^{4,17} although its behavior at $x < 0.03$ remains to be clarified.

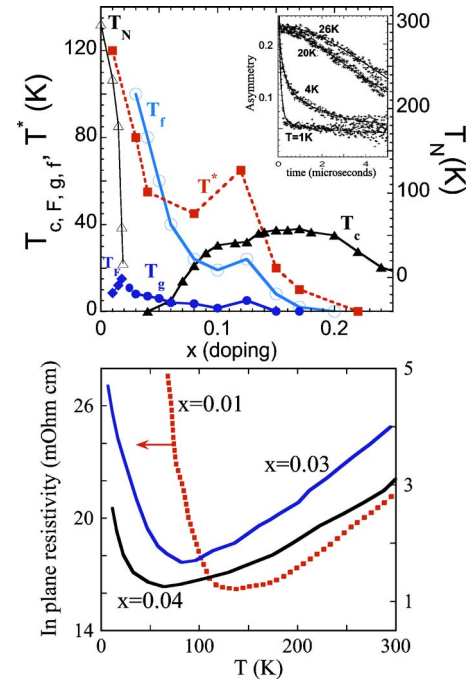


FIG. 3. (Color online) The upper panel depicts the doping dependence of the Néel temperature T_N , the second freezing T_F , and glass temperature T_g (Refs. 10 and 15). The latter two scales are determined by a peak in the spin-lattice relaxation $1/T_1$, and in the case of the nonsuperconducting samples also by uniform static magnetic susceptibility. The data point for $x=0.025$ is taken from Niedermayer *et al.* (Ref. 9). Also shown is the superconducting transition temperature T_c . The inset depicts μ SR spectra for $x=0.08$ at different temperatures, showing a deviation from a Gaussian signaling the entrance of fluctuating electronic moments into the μ SR frequency window at approximately 25 K ($=T_f$) and their eventual freezing below 4 K ($=T_g$) (Refs. 10 and 15). The doping dependence of T_f is also included. T^* is a crossover from metallic-like to insulatinglike resistivity determined from the in-plane resistivity measurements shown in the lower panel of this figure and in Ref. 24.

The question arising, of course, is to what extent electronic heterogeneity may be a generic state of correlated electron matter, and how, if at all, the charge is affected by the aforementioned freezing. Note that the emergence of electronic heterogeneity with charge doping is not unique to the HTS. It has been demonstrated in semiconductors,³ ruthenates,¹⁸ nickelates,¹⁹ other copper oxides,^{19,20} and manganites,²¹ where transport experiments indicate that at least some glassy features originate from slow-charge dynamics. Further evidence supporting that glassiness in the charge and the spin channels is generic among a wide range of materials and that the two emerge hand-in-hand was recently provided by measurements of the dielectric constant on $\text{La}_2\text{Cu}_{1-x}\text{Li}_x\text{O}_4$ and $\text{La}_{2-x}\text{Sr}_x\text{NiO}_4$.¹⁹ These materials are not superconducting, but their spin response is almost identical to that of cuprate superconductors, while the dielectric response is remarkably similar to conventional (structural) glasses.

Interestingly, the immediate emergence of glassy characteristics observed in the electronic moments detected by μ SR

and the uniform magnetic-spin-susceptibility experiments discussed above suggest analogous effects in the charge response, which we have examined here by measuring the in-plane electrical resistivity, ρ_{ab} , of LSCO single crystals (Fig. 3, lower panel). The data for $x=0.01-0.04$ show a crossover (resistivity minima) in $\rho_{ab}(T)$ from metallic to insulatinglike at a characteristic temperature which we denote as T^* (Fig. 3, upper panel). Although the crossover takes place over a wide temperature region, it clearly occurs at $T \ll T_N$ for $x < 0.02$ and has a doping dependence somewhat similar to T_g —it decreases with increasing doping. This similarity indicates the association between short-range freezing order and charge retardation. The precision and detailed doping dependence of the resistivity data when compared with the freezing temperatures obtained by μ SR and susceptibility (Fig. 3) across the boundary ($x \leq 0.02$ and $x > 0.02$) demonstrate the correlation between the insulatinglike behavior in resistivity (T^*) with the onset of the freezing of electronic moments to a glass. Moreover, the emergence of a glass order with the first added carriers speaks against impurity effects, but instead for an intrinsic property, i.e., one arising due to the added carriers rather than due to some undetected extrinsic impurity.

III. COEXISTENCE OF GLASSINESS AND SUPERCONDUCTIVITY

In light of many unconventional properties of HTS below optimum doping,⁴ it is important to probe the possible correlation between the identified dynamical heterogeneities and superconductivity (present for $x > x_{sc} = 0.05$ for LSCO). Muon spin relaxation (μ SR) has been successful in identifying the freezing of electronic moments under the superconducting dome of various HTS.^{9,10,12,15,22,23} Figure 3 (inset, upper panel) shows a typical example [LSCO, $x=0.08$ ($T_c = 21$ K)] of spectra with a glass transition at low temperatures displaying an initial rapid relaxation.¹⁵ Similar examples can be found in any of the aforementioned μ SR references. The amplitude of the muon spin polarization reveals that all muons inside the sample experience a nonzero local field, indicating that the magnetism persists throughout the entire volume of the sample. However, the absence of a dip in the polarization function at the lowest times indicates the presence of a large number of low field sites—superconducting and magnetic regions intercalated on a microscopic (≤ 2 nm) scale.^{9,10,15,22} These observations suggest the presence of magnetic stripes or droplets, in agreement with independent indications^{4,9-12,14,15,22} from transport, spectroscopic studies, and the susceptibility data shown above for the nonsuperconducting dopings. From these studies we conclude that superconductivity coexists with glassiness on a microscopic scale throughout the bulk of the material. This behavior is not limited to LSCO, although the latter has been investigated most extensively. Similar results have been observed in the “stripe-compound” Nd-LSCO, pure and Y-doped Bi-Sr-Ca-Cu-O, pure or Ca-doped Y-Ba-Cu-O, and more recently Ca-Na-Cu-O-Cl.^{9-12,14,15,22,23} The common observations in this wide range of materials (with varying number of copper oxygen planes per unit cell,

crystal structures, superconducting transition temperatures, etc.) adds credence to the case that emergent glassiness is driven purely by the added carriers to the Mott insulator. It is also possible that at a much lower temperature the regions giving rise to glassy characteristics at the specific measured temperature or frequency may form a periodic structure. In fact μ SR studies have consistently provided evidence for such ordering for at least $x < 0.10$.^{9,10,12,15,22} We note that in the case of LSCO one might need to consider the possibility that the disorder introduced by Sr might be significant. However, the emergence of a short-range freezing with the first added holes seen both in La-Sr-Cu-O and Y-Ba-Cu-O indicates that glassiness must be intrinsic to the system’s evolution from an insulator towards a superconductor. It tells us that glassiness is dictated not by the type of cation substitution, but by the charge doping itself.

IV. SPIN-CHARGE CORRELATIONS AROUND THE SUPERCONDUCTING DOME

Encouraged by the resistivity results at low dopings (Fig. 3) for a correlation between glass and charge retardation, we proceeded to examine the region where $T_c > 0$ by reanalyzing early measurements in high magnetic fields.²⁴ Here bulk superconductivity is suppressed, revealing information about low- T charge transport in the normal phase. The combination of our data and those by Boebinger *et al.*²⁴ reveal the presence of a striking similarity between the doping dependence of the spin freezing and the resistivity minima all the way to the overdoped region (Fig. 3). We find T_g , T_f , and T^* decrease upon doping, except for an increase around $x = \frac{1}{8}$ (Fig. 3), which is thought to reflect stripe pinning,⁴ or some other form of commensurate charge ordering.²⁵ Furthermore, $T^*(x)$ closely tracks the onset of the slowing down of spin fluctuations ($T = T_f$) before they freeze into a glassy state ($T = T_g$). Also, the resistivity data when put together with results for the freezing of electronic moments obtained by μ SR and susceptibility reveal the excellent correlation between spin and charge retardation across the phase diagram of HTS (Fig. 3). This observation indicates the central role of spin freezing to the charge mobility. Moreover, it emphasizes the role of intrinsic disorder to the pure metal-insulator transition previously suggested to exist in these materials.²⁴ We find that the metal-to-insulator transition in the HTS is governed by the glassy characteristics. The change of the ground state from a pure metal to insulator transition near $x = 0.15$ suggested earlier²⁴ is not precisely correct. In fact (Fig. 3, upper panel), the system changes to metallic at the point where glassiness vanishes, and this occurs nearer $x = 0.20$ —a result indicating the “smearing” of the metal to insulator transition, caused by the intervening intrinsic electronic disorder. This result is consistent with theoretical predictions⁷ that the effective disorder

$$W_{eff} = (W^2 + V^2 q_{EA})^{1/2},$$

seen by the charge carriers can be strongly enhanced by static and dynamic fluctuations associated with glassy ordering. Here, W is the energy scale of the impurity potential, V measures the electron-electron interactions, and q_{EA} is the

frozen-order parameter fluctuation in the glassy phase.⁷ Assuming that the self-generated randomness dominates the impurity potential (i.e., $W^2 \ll V^2 q_{EA}$), this mechanism also explains the correlations between the doping dependence of T_g and T^* . This is true since one expects $T_g(x) \sim [q_{EA}(x)]^{1/2}$, and the resistivity crossover scale T^* should be set by the effective disorder W_{eff} .

V. INTERMEDIATE CONDUCTING GLASS PHASE

The correlation between conductivity and glassiness indicates that for $x < x_{sc}$ ($=0.05$) we are dealing with a strongly localized insulator displaying hopping transport at $T < T^*$. Here, the number of free carriers can be expected to vanish at $T=0$, in agreement with recent Hall-effect measurements.²⁶ On the other hand, for $0.05 < x < 0.20$ the number of carriers is found to be finite,²⁶ suggesting an itinerant system even in the normal phase. In this regime, DC transport has a much weaker²⁴ (although still insulatinglike) temperature dependence. However, the observed logarithm T resistivity upturn in this region has been shown²⁴ to be inconsistent with conventional localization (interaction) corrections, which could indicate an insulating ground state. Instead, estimates²⁷ reveal this behavior to be consistent with that expected for metallic droplet-charging (tunneling) processes, as seen in quantum dots and granular metals.²⁷ These results suggest that in this regime HTS are inhomogeneous metals, where conducting droplets connect throughout the sample, and a metal-insulator transition in the normal phase happens *exactly* at $x=x_{sc}$. On the other hand, at lower densities the conducting droplets remain isolated, and the material may be viewed as an insulating cluster or stripe glass. Now, as carrier concentration increases, they connect and the carriers are free to move throughout the sample, forming filaments or “rivers.” This is, in fact, the point where free carriers emerge in Hall-effect data²⁶ and phase-coherent bulk superconductivity arises at $x > x_{sc}$. This observation indicates that it is the inhomogeneous nature of the underdoped glassy region which controls and limits the extent of the superconducting phase at low doping. It also tells us that the details of the transport and thermodynamic properties are also strongly influenced by the electronically disordered ground state.

Therefore, based on evidence for charge retardation, freezing, and uniformly distributed electronic heterogeneity in the form of glassy stripes or droplets, we propose that in the interval $x_{sc} < x < x_{opt}$ an intermediate phase arises in the form of a bad metal. The emergence of such an intermediate conducting glass phase separating a conventional metal and a glassy insulator has, in fact, been predicted in recent theoretical work.⁸ The present work provides systematic experimental evidence in support of this scenario, which may in fact be a generic feature of doped Mott insulators.

VI. QUANTUM GLASS TRANSITION

We now ask whether a true quantum critical point (QCP) separates the glassy non-Fermi liquid and the metallic-Fermi liquidlike regimes. We need an experiment where one may gradually increase the amount of disorder, enhance short-

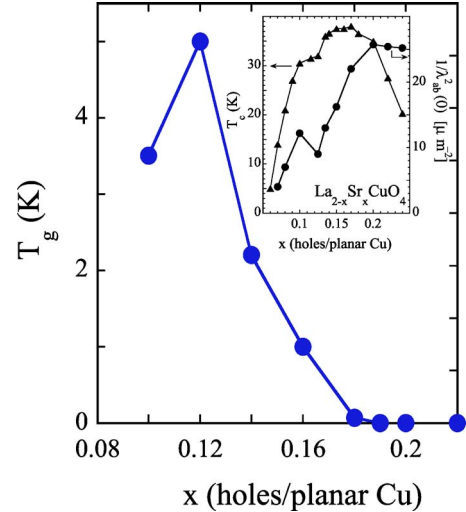


FIG. 4. (Color online) Doping dependence of the glass transition temperature T_g for LSCO doped with 5% Zn (Ref. 28). The inset shows data for the superconducting transition temperature T_c [15] and superfluid density $n_s(0) \sim 1/\lambda_{ab}^2(0)$ (Refs. 10 and 15) for the pure LSCO system, indicating the transition in the superconducting ground state precisely at the concentration where glassiness vanishes.

range correlations, suppress superconductivity, and fully expose the glassy ground state. As discussed in detail in earlier works, these conditions are met by Zn^{2+} doping.^{10,15,28} Figure 4 depicts characteristic data for $La_{2-x}Sr_xCu_{0.95}Zn_{0.05}O_4$ with the normal state exposed ($T_c=0$) across the T - x phase diagram. The similar doping dependence of T_g for pure (Fig. 3) and up to 5% Zn-doped (Fig. 4) samples^{10,15,28} indicates that regardless of a sample being pure, Zn doped, superconducting or not, we obtain universal behavior: A set of glassy phase transitions, enhanced near $x=\frac{1}{8}$, and ending at a specific doping $x=x_{opt}$ at $T=0$, supporting the presence of a quantum glass transition. The fact that even with a large amount of disorder that is enough to even suppress superconductivity across the whole phase diagram of LSCO, the doping dependence of T_g remains essentially the same as in the pure system, clearly tells us that glassiness is predominantly self-generated. That is, it emerges by doping the parent Mott insulator with carriers, consistent with those theoretical scenarios that predict phase separation^{4,5} at low doping. Coulomb interactions, however, enforce charge neutrality and prevent^{4,5} global phase separation; instead, the carriers are expected⁶ to segregate into nanoscale domains to form a stripe (cluster) glass.⁶ As quantum fluctuations increase upon doping,^{7,8} such a glassy phase should be eventually suppressed at a quantum critical point, which in LSCO emerges around $x=x_{opt} \approx 0.2$.

Remarkable independent evidence that a QCP is found precisely at $x=x_{opt}$ is provided by the observation of a sharp change in the superfluid density $n_s(0) \sim 1/\lambda_{ab}^2(0)$ [where $\lambda_{ab}(0)$ is the absolute value of the in-plane penetration depth]. At $x > x_{opt}$, $n_s(0)$ is mainly doping independent (Fig. 4, inset), while the T dependence is in good agreement with the BCS weak-coupling d -wave prediction.¹⁰ At dopings below the quantum glass transition, $n_s(0)$ is rapidly suppressed

(note the enhanced depletion near $x=\frac{1}{8}$ precisely where T_g and T^* are enhanced, which may be taken as indicative of a competition between glass order and superconductivity) and there is a marked departure of $n_s(T)$ from the canonical weak coupling curve.¹⁰ Similar behavior has been observed in other HTS and in the c -axis component.¹⁰ Therefore, the penetration depth data show that the onset of quasi-static magnetic and charge correlations coincides with an abrupt change in the superconducting ground state.

In addition, a crossover temperature T_m at $x > x_{opt}$ separating marginal Fermi liquid transport at $T > T_m$ from more conventional metallic behavior at $T < T_m$ also seems to drop²⁹ to very small values around optimum doping (see, e.g., the schematic plot in Fig. 1). At $x > x_{opt}$ the ground state becomes metallic and homogeneous, with no evidence for glassiness or other form of nanoscale heterogeneity.^{10,15,24,26,28}

These results provide strong evidence of a sharp change in ground-state properties at $x=x_{opt}$, and the emergence of vanishing temperature scales as this point is approached—just as one expects at a QCP. Moreover, it is in this doping region where the Fermi surface topology of LSCO changes from holelike to electronlike.³⁰ Note that the extent of the region between the doping where antiferromagnetism (AF) vanishes and x_{sc} , and the extent of the region between the latter and x_{opt} , are material dependent and expected to vary across the HTS families. As a matter of fact, measurements of the in-plane resistivity on underdoped ($T_c=60$ K) ultra-clean YBCO single crystals with superconductivity suppressed by pulsed magnetic fields found that both the temperature dependence and magnitude of the measured resistivity are in excellent agreement with those for LSCO ($x=0.17$).³¹ This result on YBCO indicates that the evolution of the ground-state transport in these two very different HTS families (YBCO, LSCO) is essentially the same, but the characteristic changes in the ground state are shifted in doping. Furthermore, it is for the 60 K YBCO that μ SR¹² shows a decline in T_g . These results suggest that as in LSCO ($x=0.17$), adding a few percent more carriers to the 60 K

YBCO will eventually lead to a metallic resistivity all the way to the lowest temperatures (just as it happens for $x=0.20$ in LSCO). This is likely to also coincide with the absence of a glass transition and optimal superfluid density at these higher dopings in YBCO. In light of these results and correlations, further studies would be extremely useful. Furthermore, for a better understanding of the magnetic ground state in these ultraclean YBCO crystals, and for a closer comparison to LSCO, it would be interesting to actually measure the magnetic susceptibility to the lowest possible temperatures in the superconducting-antiferromagnetic doping region of YBCO.

VII. SUMMARY

In summary, we have identified three distinct doping regimes: (1) $x < x_{sc}$; (2) $x_{sc} < x < x_{opt}$; and (3) $x > x_{opt}$, separated by two critical points: a quantum glass transition at $x=x_{opt}$ and a normal state metal-insulator transition at $x=x_{sc}$ within the glassy phase. We propose HTS could bear resemblance to other materials close to disorder-driven metal-insulator transitions, where electronic heterogeneity and self-generated glassiness arise with the first added charge carriers—a mechanism that may potentially explain many puzzling features of cuprate superconductors. Our evidence for an intercepted QCP by self-generated glass order indicates glassiness could be a convenient and useful “tool” to study putative QCP’s in strongly correlated electron systems.

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