Evidence for correlation between spin and charge dynamics in La$_2$Cu$_{1-x}$Li$_x$O$_4$

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From ac magnetic susceptibility measurements on Li-doped La$_2$CuO$_4$, with $H_{ac}$ parallel and perpendicular to the CuO plane, we find frequency-dependent behavior below a spin-glass temperature ($T_{SG}$). $T_{SG}$ obtained from magnetic susceptibility is higher than the charge-glass temperature $T_{CG}$ obtained from dielectric constant measurements, indicating that spin freezes first and drives charge freezing at a lower temperature. Similar frequency dependence of the two characteristic freezing temperatures underlines that charge and spin dynamics are strongly correlated in this cuprate compound. © 2009 American Institute of Physics. [DOI: 10.1063/1.3062830]

I. INTRODUCTION

When magnetic interactions among adjacent moments are frustrated, the magnetic system cannot establish long-range magnetic order, but rather goes through a transition to a spin-glass state where randomly oriented spins are frozen.$^1$ In the high-$T_c$ parent compound La$_2$CuO$_4$, a spin-glass behavior appears with Li doping, showing a dome of the spin-glass transition temperatures centered around $x_c$, where $T_N$ is completely suppressed.$^{2-5}$ Unlike most spin-glass systems, where the entire spins participate in the freezing process, however, the $S=1/2$ spin of the Cu$^{2+}$ ions in La$_2$Cu$_{1-x}$Li$_x$O$_4$ partially contribute to the spin-glass phase, while the remnant Cu spins contribute to a long-range magnetic order for $x<x_c$. Sr doping the parent compound, La$_2$Sr$_{2-x}$CuO$_4$, also suppresses $T_N$ and induces partial spin freezing for $x<x_c$.\(^6\)\(^-\)\(^1\)\(^1\)

In this case, the spin-glass state persists deep into the superconducting state up to $x=0.15$, indicating coexistence of spin-glass and superconducting phases.\(^8\)\(^\text{-}\)\(^1\)\(^2\)\(^1\)\(^3\)\(^1\)\(^4\)\(^1\)\(^5\)\(^1\)\(^6\)\(^1\)\(^7\)\(^1\)\(^8\)\(^1\)\(^9\)\(^1\)\(^0\)\(^1\)\(^1\)\(^2\)\(^1\)\(^3\)\(^1\)\(^4\)\(^1\)\(^5\)\(^1\)\(^6\)\(^1\)\(^7\)\(^1\)\(^8\)\(^1\)\(^9\)\(^1\)\(^0\)\(^1\)\(^1\)\(^2\)\(^1\)\(^3\)\(^1\)\(^4\)\(^1\)\(^5\)\(^1\)\(^6\)\(^1\)\(^7\)\(^1\)\(^8\)\(^1\)\(^9\)\(^1\)\(^0\)\(^1\)\(^1\)\(^2\) The ubiquitous presence of spin-glass behavior both in the long-range antiferromagnetic (AFM) and superconducting phase suggests that understanding nature of the glass dynamics is critical to properly guide a theory for the high-$T_c$ superconductivity.

Recently, neutron scattering measurements have suggested that the unconventional form of the spin-glass phase in the high-$T_c$ cuprates is a reflection of strong anisotropy between in-plane and interplanar antiferromagnetic correlations of the neighboring Cu spins.\(^2\) Charge dynamics of the hole-doped cuprate La$_2$Cu$_{1-x}$Li$_x$O$_4$ ($x=0.023$) has been explored via low-frequency dielectric response, where a large drop in the dielectric constant below a characteristic temperature $T_{CG}$ and a frequency dependence of $T_{CG}$ indicates the realization of a charge-glass state.\(^13\) Here, $T_{CG}$ is the charge-freezing temperature. At this Li-concentration ($x=0.023$), magnetization measurements have shown a signature of long-range AFM order at 100 K, indicating that $x=0.023$ is slightly lower than the critical concentration ($x_c$). Even though there are many experiments that show spin-glass behavior in the Li-doped cuprates, uncertainty in the doping concentration prevents a direct comparison between the spin and charge dynamics of the high-$T_c$ cuprate compounds. In this brief report, we show ac (alternating current) magnetic susceptibility measurements on La$_2$Cu$_{1-x}$Li$_x$O$_4$ ($x=0.023$), the same composition that was measured in the dielectric constant study.\(^13\) Observation of a frequency-dependent magnetic susceptibility, a hallmark of a spin-glass state, underlines that the spin and charge degrees of freedom are intricately connected.

II. EXPERIMENTAL DETAILS

Single crystals of the Li-doped lanthanum cuprate La$_2$Cu$_{1-x}$Li$_x$O$_4$ ($x=0.023$) were synthesized by a flux method.\(^3\) ac magnetic susceptibility was measured as a function of temperature and in the frequency range 100 Hz to 10 kHz with a quantum design ac measurement system. The measured single crystals were from the same batch that were reported by Ref. 13. In these measurements, a small ac driving field ($\approx$10 Oe) was applied parallel and perpendicular to the crystalline c-axis at zero dc magnetic field.

III. RESULTS AND DISCUSSION

Figure 1 shows the real part of magnetic susceptibility ($\chi'$) of La$_2$Cu$_{1-x}$Li$_x$O$_4$ ($x=0.023$) or Li-La$_2$14 for ac magnetic field applied perpendicular [Fig. 1(a)] and parallel [Fig. 1(b)] to the c-axis. $\chi'$ initially increases with decreasing temperature for both ac field directions and is independent of frequency for $T>30$ K. With further decreasing temperature, the ac susceptibility reveals a strong frequency dependence, a hallmark of spin-glass behavior. For $H_{ac} \perp$ c-axis, the susceptibility starts to decrease below $\approx$7 K at $f$ =100 Hz [squares in Fig. 1(a)], which we assign as a spin-freezing temperature $T_{SG}$. With increasing frequency, as expected for spin-glass systems, $T_{SG}$ increases.

In Fig. 1(b), $\chi'$ of Li-La$_2$14 is shown for 100 (squares), 1 K (circles), and 10 kHz (triangles), where $H_{ac}$ is applied parallel to the c-axis. At 100 Hz, deviation from a monotonic
increase in \( \chi' \) occurs with decreasing temperature at \( \approx 17 \) K (marked by an arrow). Similar to \( H_{ac} \perp c \)-axis [Fig. 1(a)], a local maximum, which we assign as a spin-freezing temperature for this ac field direction, increases with increasing frequency. Below the \( T_{SG} \), \( \chi' \) decreases initially with decreasing temperature before increasing again, which is completely different from that of \( \chi' \) for \( H_{ac} \perp c \)-axis. The increase in the susceptibility at low temperatures has been observed often in a system with magnetic impurities. The lack of an increase in \( H_{ac} \perp c \)-axis, however, rules out an impurity scenario. The disparate temperature dependences of the magnetic susceptibility on ac magnetic field direction may be relevant to the anisotropic magnetic interactions in Li-La214, where antiferromagnetic correlation among Cu\(^{2+} \) spins within the CuO plane is very strong, while the interlayer coupling is weak.

Recent neutron scattering measurements on La\(_2\)Cu\(_{0.92}\)Li\(_{0.08}\)O\(_4\) have shown that only a fraction of spins become spin glass, while the rest become spin liquids because the in-plane antiferromagnetic exchange coupling dominates the interplanar one.\(^2\)

Are spin and charge-glass states correlated? If so, which glass state appears first and drives the other? In order to provide a clue to these questions, we plot both spin- and charge-freezing temperatures of La\(_2\)Cu\(_{1-x}\)Li\(_x\)O\(_4\) in Fig. 2. The charge-freezing temperature \( T_{CG} \) for \( E \parallel [001] \) is obtained from Ref. 13, where a large dielectric constant at high temperature drops by a factor of 100 below \( T_{CG} \) (diamonds). \( T_{SG} \) for \( H_{ac} \parallel [001] \) (squares) closely follows \( T_{CG} \) while \( T_{SG} \) for \( H_{ac} \perp [001] \) (circles) is distinctly different from \( T_{CG} \). In order to estimate the spin-glass transition temperature \( T_{SG} \) in the zero-frequency limit \( (T_0) \), we use a power-law form of \( \tau(T)=a(T-T_0)^{-n} \), which describes critical slowing down of the relaxation rate \( \tau \) in a spin glass.\(^{14}\) The frequency dependence of \( T_{CG} \) is best described by this model with \( T_0=5 \) K and \( n=3.2 \) (see the dashed line). The other dashed lines through the squares and the circles represent a least-squares fit to the \( T_{SG} \)'s, where the spin-glass transition temperature \( T_0 \) is 5.5 and 13.6 K for \( H_{ac} \) perpendicular and parallel to the \( c \)-axis, respectively. In the fitting, \( n \) was fixed to 3.2, the same exponent that describes \( T_{CG} \) Even though only three data points are available for the spin-glass fit, the good agreement between the evolution of \( T_{SG} \) and the power-law form with fixed \( n \) suggests that similar critical slowing down occurs both in the spin- and charge-relaxation rates. We note a large difference between spin- and charge-glass transition temperatures for magnetic (or electric) field direction along the \( c \)-axis: \( T_{SG}=13.6 \) K and \( T_{CG}=5 \) K in the zero-frequency limit, indicating that spins are frozen first and doped holes that separate the frozen spins freeze at a lower temperature. In the Sr-doped La\(_{1.94}\)Sr\(_{0.06}\)CuO\(_4\), where superconductivity and spin-glass phases coexist below 5 K, NMR measurements indicate that doped holes are frozen first and the hole-poor antiferromagnetic (AF) clusters frozen at a much lower temperature.\(^8\) The difference in the charge-spin dynamics between the Li-doped and Sr-doped compounds suggests that the origin of the spin (or charge) glass may differ in the two doped materials.
IV. CONCLUSION

The ac magnetic susceptibility of La$_2$Cu$_{1-x}$Li$_x$O$_4$ with $x=0.023$ has been measured for $H_{ac}$ parallel and perpendicular to the $c$-axis at zero dc magnetic field. The susceptibility shows a frequency-dependent behavior below $T_{SG}$, a hallmark of a spin-glass behavior for both field orientations. The low-temperature behavior of $\chi'$, however, reveals a qualitatively different temperature dependence between the two-field directions, reflecting a strong anisotropy between the in-plane and interplane exchange magnetic correlations. For magnetic (or electric) field direction along [001], a form of spins freezes first, then charge freezing follows at lower temperatures. Both spin- and charge-freezing temperatures follow the same form of critical slowing down of the relaxations rates, indicating a strong correlation between inhomogeneous spin and charge dynamics.

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