Evidence for correlation between spin and charge dynamics in $La_2Cu_{1-x}Li_xO_4$

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From ac magnetic susceptibility measurements on Li-doped La_2CuO_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_2CuO_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 $La_2Cu_{1-x}Li_xO_4$ partially contribute to the spin-glass phase, while the remanent Cu spins contribute to a long-range magnetic order for $x<x_c$. Sr doping the parent compound, $La_{2-x}Sr_xCuO_4$, also suppresses T_N and induces partial spin freezing for $x<x_c$.[6–11] 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.[8,10,12] 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 $La_2Cu_{1-x}Li_xO_4$ ($x=0.023$) 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 $La_2Cu_{1-x}Li_xO_4$ ($x=0.023$), 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.

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II. EXPERIMENTAL DETAILS

Single crystals of the Li-doped lanthanum cuprate $La_2Cu_{1-x}Li_xO_4$ ($x=0.023$) were synthesized by a flux method.[5] 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 (=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 (χ') of $La_2Cu_{1-x}Li_xO_4$ ($x=0.023$) or Li-La214 for ac magnetic field applied perpendicular [Fig. 1(a)] and parallel [Fig. 1(b)] to the *c*-axis. χ' 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 ≈ 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), χ' of Li-La214 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

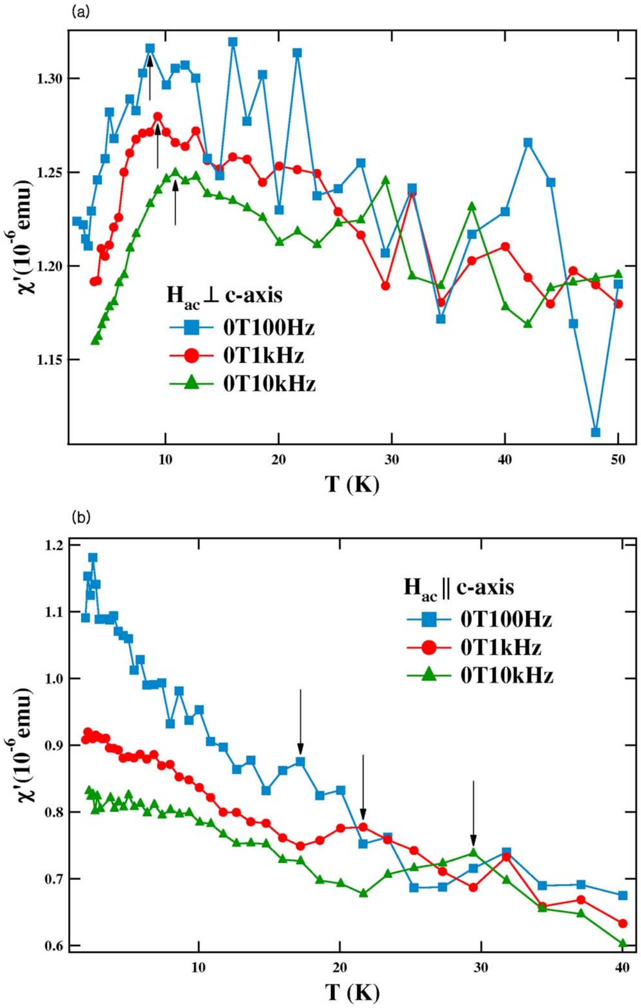

FIG. 1. (Color online) ac magnetic susceptibility of $La_2Cu_{1-x}Li_xO_4$ ($x=0.023$) as a function of temperature at zero dc magnetic field (a) for $H_{ac} \perp c$-axis and (b) for $H_{ac} \parallel c$-axis at 100, 1 K, and 10 kHz. Here H_{ac} is the amplitude of an oscillation magnetic field. The arrows indicate deviation temperatures from a monotonic temperature dependence of the susceptibility, which are assigned as spin-freezing temperatures.

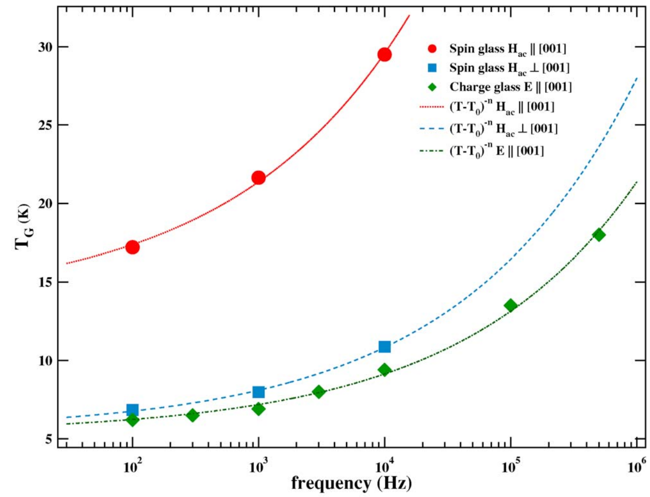

FIG. 2. (Color online) Frequency dependence of the charge-(T_{CG}) and spin-(T_{SG}) freezing temperatures of $La_2Cu_{1-x}Li_xO_4$ ($x=0.023$). Diamonds describe T_{CG}, squares represent T_{SG} for $H_{ac} \perp c$-axis, and circles depict T_{SG} for $H_{ac} \parallel c$-axis. Dashed lines are a least-squares fits to $\tau = a(T-T_0)^{-n}$ (see the text).

increase in χ' occurs with decreasing temperature at ≈ 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}, χ' decreases initially with decreasing temperature before increasing again, which is completely different from that of χ' 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 $La_2Cu_{0.94}Li_{0.06}O_4$ 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 $La_2Cu_{1-x}Li_xO_4$ 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 $H_{ac} \perp [001]$ (squares) closely follows T_{CG}, while T_{SG} for $H_{ac} \parallel [001]$ (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 τ 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 $La_2Cu_{1-x}Li_xO_4$ 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 χ', 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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