Anelastic spectroscopy of the cluster spin-glass phase in La$_{2-x}$Sr$_x$CuO$_4$

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An increase of the acoustic absorption is found in La$_{2-x}$Sr$_x$CuO$_4$ ($x=0.019, 0.03$, and $0.06$) close to the temperatures at which freezing of the spin fluctuations in antiferromagnetic-correlated clusters is expected to occur. The acoustic absorption is attributed to changes of the sizes of the quasifrozen clusters induced by the vibration stress through magnetoelastic coupling.

I. INTRODUCTION

The low doping region of the phase diagram of La$_{2-x}$Sr$_x$CuO$_4$ is attracting considerable interest, due to the appearance of unconventional correlated spin dynamics and ordering processes (for a review see Ref. 1). In undoped La$_2$CuO$_4$ the Cu$^2+$ spins order into a 3D antiferromagnetic (AF) state with the staggered magnetization in the $ab$ plane. Doping by Sr rapidly destroys the long range AF order, with $T_N$ passing from 315 K to practically 0 K around $x_c \approx 0.02$. Above this critical value of the Sr content no long range AF order is experimentally observed at finite temperature. There are also indications that the holes are segregated into domain walls, sometimes identified as charge stripes, which separate hole-poor regions where the AF correlations build up. The holes should be mobile along these “charge rivers,” but at low $x$ they localize near the Sr atoms below $\sim 30$ K, causing a distortion of the spin texture of the surrounding Cu$^{2+}$ atoms. For $x < x_c$ the spin distortions around the localized holes are decoupled from the AF background, and freeze into a spin-glass (SG) state below $T_f(x) \approx (815 \text{ K})x$. For $x > x_c$, a cluster spin-glass (CSG) state is argued to freeze below $T_g(x) \approx 1/x$ and AF correlations develop within the domains defined by the charge walls, with the easy axes of the staggered magnetization uncorrelated between different clusters. The formation of the SG and CSG states are inferred from sharp maxima in the $^{139}$La NQR (Refs. 1, 3, 5) and $\mu$SR (Ref. 6) relaxation rates, which indicate the slowing of the AF fluctuations below the measuring frequency ($\sim 10^7 \sim 10^8$ Hz in those experiments) on cooling, and from the observation of irreversibility, remnant magnetization, and scaling behavior in magnetic susceptibility experiments.

Here we report the observation of a steplike increase of the low-frequency acoustic absorption close to the temperature at which the spin freezing process is detected in the NQR measurements. The absorption is ascribed to changes of the sizes of the frozen clusters induced by the vibration stress through magnetoelastic coupling, or equivalently to the motion of the walls between them.

II. EXPERIMENTAL AND RESULTS

The samples where prepared by standard solid state reaction as described in Ref. 10 and cut in bars approximately $40 \times 4 \times 0.6$ mm$^3$. The final Sr contents and homogeneities where checked from the temperature position and sharpness of the steps in the Young’s modulus and acoustic absorption due to the tetragonal (HTT) to orthorhombic (LTO) transition, which occurs at a temperature $T_t$ linearly decreasing with doping. The transitions appear narrower in temperature than the one of a Sr-free sample, indicating that the width was mostly intrinsic and not due to Sr inhomogeneity, except for the sample at the lowest Sr content. The Sr concentrations estimated in this way turned out $x=0.0185 \pm 0.0015$, $0.0315 \pm 0.0015$, and $0.0645 \pm 0.002$, in good agreement with the nominal compositions. In the following the samples will be referred as $x=0.019, 0.03$, and $0.06$.

The complex Young’s modulus $E$ was measured by electrostatically exciting either of the lowest three flexural modes and detecting the vibration amplitude by a frequency modulation technique. The elastic energy loss coefficient (or reciprocal of the mechanical $Q$) is related to the imaginary part $E''$ of $E$ by $Q^{-1} (\omega,T) = E''(\omega,T)/E'(\omega,T)$, and it was measured by the decay of the free oscillations or the width of the resonance peak.

In Fig. 1 the anelastic spectra of three samples with $x = 0.019, 0.03, 0.06$ below 16 K measured exciting the first flexural mode are reported. A steplike increase of the absorption is observed around or slightly below $T_g$ (Ref. 11) The gray arrows indicate the values of $T_g$ in the magnetic phase diagrams deduced from NQR (Ref. 3) (lower values) and $\mu$SR (Ref. 6) (higher values) experiments, which are in agreement with the data in Ref. 11 [for the sample with $x = 0.019$ the $T_g(x=0.02)$ values are indicated]. The black arrows indicate the temperature of the maximum of the $^{139}$La NQR relaxation rate measured on the same samples in a separate study, which indicate a freezing in the spin-glass phase, as discussed later. The coincidence of the temperatures of the absorption steps with those of freezing of the

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spin fluctuations suggest a correlation between the two phenomena.

The sample with \( x = 0.03 \) was outgassed from excess O by heating in vacuum up to \( 790 \) K, while the other two samples where in the as-prepared state, therefore containing some interstitial O. The concentration \( \delta \) of excess O is a decreasing function of \( x \) (Ref. 14) and should be negligible for \( x = 0.06 \) but not for \( x = 0.019 \). This fact allowed us to observe the absorption step singled out from the high-temperature tail of an intense peak that occurs at lower temperature (see the sharp rise of dissipation below \( 3 \) K for \( x = 0.019 \) in Fig. 1). Such a peak has been attributed to the tunneling-driven tilt motion of a fraction of the O octahedra.\(^{12,15} \) The LTO phase is inhomogeneous on a local scale,\(^{15-18} \) and a fraction of the octahedra would be unstable between different tilt orientations, forming tunneling systems which cause the anelastic relaxation process. The interstitial O atoms force the surrounding octahedra into a fixed tilt orientation, resulting in a decrease of the fraction of mobile octahedra and therefore in a depression of the absorption peak. In addition, doping shifts the peak to lower temperature at a very high rate, due to the coupling between the tilted octahedra and the hole excitations.\(^{12} \) Therefore, it is possible to reduce the weight of the low-temperature peak by introducing concentrations of interstitial O atoms that are so small that they do not change appreciably the doping level due to the Sr substitutions. Figure 2 compares the absorption curves of the \( x = 0.019 \) sample in the as-prepared state with a concentration \( \delta = 0.002 \) of excess O and after removing it in vacuum at high temperature. The initial concentration \( \delta \) has been estimated from the intensity of the anelastic relaxation process due to the hopping of interstitial O,\(^{18} \) whose maximum occurs slightly below room temperature at our measuring frequencies (not shown here). The presence of excess O indeed decreases and shifts to lower temperature the tail of the peak in Fig. 2, while the effect on the absorption step is negligible. This justifies the comparison of the sample with \( x = 0.019 \) and \( \delta > 0 \) together with the other samples with \( \delta = 0 \), and demonstrates that the nature of the low-temperature peak is different from that of the steplike absorption.

### III. DISCUSSION

The present data show the presence of a step in the acoustic absorption at the boundary of the spin-glass quasiordered state in the \( T \times \) magnetic phase diagram. The case of the \( x = 0.019 \) sample is less clear cut, since the step is rather smooth. Furthermore, the Sr content is within the range \( 0.018 < x < 0.02 \), at the boundary between the SG and the CSG phases, where the phase diagram is largely uncertain.\(^{15} \) The \( T_{g}(x) \) line ends at \( 15 \) K for \( x = 0.018 \), and the line \( T_{g}(x) \) starts from \( 10 - 12 \) K at \( x = 0.02 \) (Refs. 8, 11, 13). A larger spread of experimental data\(^{11} \) (from 7.8 to 12.5 K) is actually observed just at \( x = 0.02 \).

A mechanism which in principle produces acoustic absorption is the slowing down of the magnetic fluctuations toward the spin-glass freezing. When measuring the spectral density \( J_{\text{spin}}(\omega, T) \) of the spin fluctuations (the Fourier transform of the spin-spin correlation function), e.g., through the \( ^{139} \) La NQR relaxation rate, a peak in \( J_{\text{spin}} \) is found at the temperature at which the fluctuation rate \( \tau^{-1}(T) \) becomes equal to the measuring angular frequency \( \omega \). This explains why the maximum of the \( \mu \)SR relaxation rate is at higher temperature than that of NQR. Near the glass transition the magnetic fluctuation rate was found to approximately follow the law\(^{3} \) \( \tau^{-1} \propto (T - T_{g}(T_{g}^{0} - T_{g}, T_{g}^{0}) / T_{g}^{0})^{2} \), and the temperature at which the condition \( \omega \tau = 1 \) for the maximum of relaxation is satisfied for \( \omega / 2\pi = 12 - 19 \) MHz is close to \( T_{g} \). A similar peak would be observed in the spectral density of the lattice strain \( J_{\text{lat}}(\omega, T) \), if the spin fluctuations cause strain fluctuations through magnetoelastic coupling. The acoustic absorption is proportional to the spectral density of the strain and hence to \( J_{\text{lat}}^{n} \): \( Q^{-1} = \omega J_{\text{str}} / T \propto \omega J_{\text{lat}} / T \), and therefore at our frequencies (\( \omega \leq 50 \) kHz) we should observe a narrow peak at a temperature slightly lower than the ones detected by NQR relaxation. The absorption steps in Fig. 1 can hardly be identified in a strict way as due to the contribution from the
freezing magnetic fluctuations because they appear as steps instead of peaks. We propose that the main contribution comes from the stress-induced movement of the boundaries between the clusters of quasifrozen antiferromagnetically correlated spins. The mechanism is well known for ferromagnetic materials, but is possible also for an ordered AF state, if an anisotropic strain is coupled with the easy magnetization axis. In this case, the elastic energy of domains with different orientations of the easy axis would be differently affected by a shear stress, and the lower energy domains would grow at the expenses of the higher energy ones. The dynamics of the domain boundaries is different from that of the critical or freezing fluctuations and generally produces broad peaks in the susceptibilities. An example is the structural HTT-LTO transformation in the same samples, where the appearance of the orthorhombic domains is accompanied by a steplike increase of the acoustic absorption.\(^{20}\) We argue that the features in the anelastic spectra just below \(T_g\) are associated with the stress-induced motion of the walls enclosing the clusters of AF correlated spins. More properly, the anelastic relaxation is attributed to the stress-induced changes of the sizes of the different domains.

The \(x=0.019\) sample is at the border \(x_c\approx0.02\) between the SG and CSG states. The NQR measurements on the same sample\(^{13}\) indicate a spin-freezing temperature \(\sim9\) K, closer to the CSG \(T_f(x_c)\) rather than to the SG \(T_f(x_c)\), which is consistent with the presence of moving walls, otherwise absent in the SG state. Nonetheless, following the model proposed by Gooding et al.\(^{21}\) we do not expect a sharp transition between the SG and the CSG states. According to that model, at low temperature the holes localize near the Sr dopants, and in the ground state an isolated hole circulates clockwise or anticlockwise over the four Cu atoms neighbors to Sr. Such a state induces a distortion of the surrounding Cu spins, otherwise aligned according to the prevalent AF order parameter. The spin texture arising from the frustrated combination of the spin distortions from the various localized holes produces domains with differently oriented AF order parameters, which can be identified with the frozen AF spin clusters. The dissipative dynamics which we observe in the acoustic response should arise from the fact that the energy clusters. The dissipative dynamics which we observe in the acoustic response should arise from the fact that the energy absorption is not peaked at \(\omega\) cause to the low-temperature peak (Fig. 2) and to the rest of the anelastic spectrum.\(^{15,12}\) This is consistent with a dissipation mechanism of magnetic rather than of structural origin.

IV. CONCLUSION

The elastic energy loss coefficient of \(\text{La}_{2-x}\text{Sr}_{x}\text{CuO}_4\) (proportional to the imaginary part of the elastic susceptibility) measured around \(10^2\) Hz in samples with \(x=0.019, 0.032,\) and 0.064 shows a steplike rise below the temperature of the transition to a quasifrozen cluster spin-glass state. The origin of the acoustic absorption is thought to be magnetoelastic coupling, namely, anisotropic in-plane strain associated with the direction of the local staggered magnetization. The absorption is not peaked at \(T_g\) and therefore does not directly correspond to the peak in the dynamic spin susceptibility due to the spin freezing. Rather, it has been ascribed to the stress-induced changes of the sizes of the spin clusters, or equivalently to the motion of the walls. The phenomenology is qualitatively accounted for in the light of the model of Gooding et al.,\(^{21}\) of magnetic correlations of the \(\text{Cu}^{2+}\) spins induced by the holes localized near the Sr dopants.

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