Tunneling-driven tilt modes of the O octahedra in La$_{2-x}$Sr$_x$CuO$_4$: Strong dependence on doping

F. Cordero

Istituto di Acustica "O.M. Corbino," CNR, Area della Ricerca di Roma–Tor Vergata, Via del Fosso del Cavaliere 100, I-00133 Roma, Italy
and INFM, Italy

R. Cantelli

Dipartimento di Fisica, Università di Roma "La Sapienza," Piazzale Aldo Moro 2, I-00185 Roma, Italy
and INFM, Italy

M. Ferretti

Dipartimento di Chimica e Chimica Fisica, Università di Genova, Via Dodecanneso 31, I-16146 Genova, Italy
and INFM, Italy

The anelastic spectrum of La$_{2-x}$Sr$_x$CuO$_4$ ($x=0$, 0.008, 0.019, 0.032) has been measured down to 1.5 K, in order to see the effect of doping on the intrinsic lattice fluctuations already found in stoichiometric La$_2$CuO$_4$, and identified with tunneling driven tilt modes of the O octahedra. Slight doping with Sr causes a drastic increase of the transition rates and relaxation strength of the tunneling systems. The influence of doping on the relaxation rate is interpreted in terms of direct coupling between the tilts of the octahedra and the hole excitations. However, the observed fast dependence of the rate on temperature cannot be explained in terms of the usual models of coupling between a tunneling system and the conduction electrons.

I. INTRODUCTION

Among the several noticeable properties of the superconducting cuprates, the interplay between the lattice and the electrons is particularly intriguing and gives rise to a variety of phenomena which are difficult to be systematized. On one side, there are several indications of separation of the charge carriers into nanodomains which are superconducting and antiferromagnetic,[1] and the charge phase separation is thought to be an essential ingredient of the mechanisms leading to high-T_c superconductivity.[2–4] There is also growing evidence that the superconducting and antiferromagnetic domains consist of stripes with a periodicity which is incommensurate with the lattice and dependent on doping, suggesting that these phenomena exclusively arise from interactions between the charge carriers.[2] Indeed, although a lattice modulation should correspond to the charge modulation, superlattice Bragg peaks are hardly observed in the superconducting cuprates, unless the modulation is pinned at a commensurate wave vector by sufficient disorder of the ion sizes in the La sublattice.[5] The lack of definite superlattice peaks is attributed to the fluctuating nature of these stripes and their short coherence length.

On the other side, there is growing evidence that the lattice of perovskite-type materials, including the layered superconducting cuprates, locally has a lower symmetry than that evinced from diffraction experiments. Such anomalous lattice fluctuations can be reproduced in models of highly anharmonic lattice dynamics, e.g., due to the high polarizability of the O ions,[6] without explicit introduction of mobile charge carriers. Local lattice fluctuations, charge separation or charge density waves instabilities and the mechanisms of electron-phonon interaction may well be connected to each

other, but no clear relationship between these phenomena has been established yet.

Previous anelastic spectroscopy[7] and joint anelastic and NQR measurements[8] showed that the tilt modes of the O octahedra in undoped La$_2$CuO$_4$ present pseudodiffusive dynamics, namely collective thermally activated hopping between the minima of a multiwell potential. In what follows we are concerned with the faster motion whose effects in the anelastic spectra are observed at liquid He temperature, and imply a tunneling driven motion[7] of more local character. The present results show a drastic increase of the characteristic frequencies of these pseudodiffusive modes in La$_{2-x}$Sr$_x$CuO$_4$ with slight doping, indicating that these tilt modes are directly coupled with the electronic excitations.

II. EXPERIMENTAL

We measured the complex Young's modulus of La$_{2-x}$Sr$_x$CuO$_{4+\delta}$ at small doping down to 1.2 K. The samples, with nominal compositions $x=0$, 0.0075, 0.015, and 0.03, were prepared by standard solid-state reaction from powders first treated for 18 h in air at 1050 °C, checked by x-ray diffraction, pressed, sintered for 18 h in air at 1050 °C as described in Ref. 9 and cut as bars approximately $40\times4\times0.6$ mm^3. The Sr content and homogeneity of the samples where checked from the transition between the tetragonal (HTT) and the orthorhombic (LTO) structure, which occurs at a temperature T_t linearly decreasing with doping[10] as $T_t(x)=(535-2180\,x)$ K. The effect of the transition on the complex Young's modulus was measured after outgassing from interstitial O, which also lowers the transformation temperature. The transition from HTT to LTO produces a huge softening of the in-plane shear modulus, whose temperature dependence in measurements on single crystals
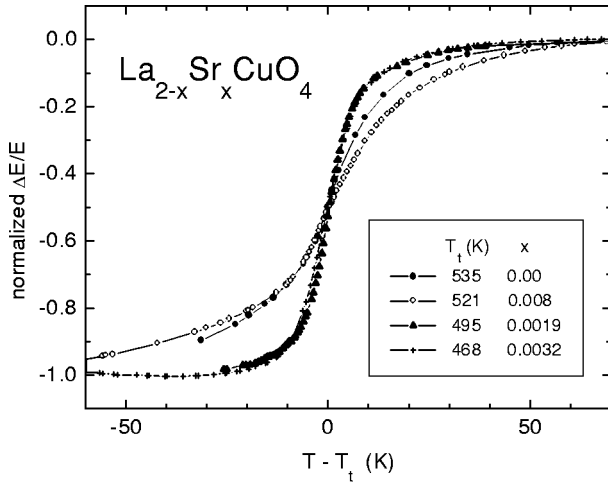
FIG. 1. Normalized variation of the Young's modulus of four samples with nominal $x=0$, 0.0075, 0.015, and 0.030 at the HTT/LTO transition. The normalization is both in the amplitude and temperature position of the step. The samples are labeled with the values of x obtained from the combined analysis of the steps in the modulus and in the absorption.

FIG. 2. Anelastic spectrum of $La_{2-x}Sr_xCuO_4$ ($x=0.019$) after O reduction in vacuum, measured exciting three flexural modes.

could be fitted within the HTT phase by using an appropriate Landau free energy.[11] However, unlike a usual displacive transformation, the softening proceeds also in the LTO phase,[12] instead of recovering. For this reason, the step in the Young's modulus is rather broad and it is not straightforward to determine an exact transition temperature T_t. Concomitantly with the modulus drop, the absorption increases, so that it is possible to determine the occurrence of the transition also from the absorption step. We assumed that in the Sr-free sample it is $T_t=535$ K (Ref. 10), which falls exactly at half of the modulus step, and determined T_t for $x>0$ by shifting the temperature scale in order to overlap the steps in the modulus (as shown in Fig. 1) or in the absorption. In this way, for the three Sr-doped samples we find $T_t=(518\pm3)$ K, (493.5 ± 1.5) K, and (464 ± 4) K, corresponding to $x=0.008\pm0.0015$, 0.019 ± 0.001, and 0.032 ± 0.002, respectively.

From Fig. 1 it appears that, except for the sample with $x=0.008$, the transition of the Sr-doped samples is sharper than that of the Sr-free sample, ensuring that the broadening is intrinsic and Sr is homogeneously distributed. The broader transition of the sample with $x=0.008$ may indicate some inhomogeneity in the Sr distribution at such very low doping level.

The imaginary part (proportional to the acoustic absorption, or reciprocal of the mechanical Q) and real part (Young's modulus E) of the dynamic modulus where measured by electrostatically exciting the flexural vibrations of the bars suspended on thin wires in correspondence with the nodal lines. The resulting anelastic spectra provide information on the low frequency dynamical processes, mainly of relaxational or diffusive character, i.e., described by a correlation function of the form $\exp(-t/\tau)$, where τ is the relaxation (correlation) time. In the ideal case of independent units described by the same τ, one has pure Debye relaxation:[13]
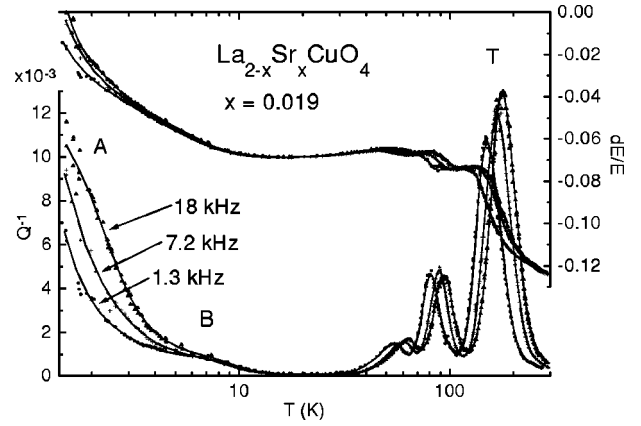
$$Q^{-1}=\Delta\frac{\omega\tau}{1+(\omega\tau)^2}, \quad \frac{dE}{E}=-\Delta\frac{1}{1+(\omega\tau)^2}, \qquad (1)$$

which are a peak in the absorption and a step in the modulus dispersion centered at the temperature for which $\omega\tau=1$, where $f=\omega/2\pi$ is the sample vibration frequency. The relaxation rate τ^{-1} is generally an increasing function of temperature, and therefore a temperature scan of the complex modulus presents the absorption peak and modulus step at a temperature which decreases with decreasing the measuring frequency, or with increasing relaxation rate. The relaxation strength Δ is proportional to the fraction of the relaxing units, which are the dynamically tilting octahedra, for the processes which we are concerned with. The measurements were made on cooling, but the whole relaxation spectrum of the Sr-free samples is reproducible below room temperature also on heating (unpublished results); since Sr-free samples exhibit the same absorption peaks of Fig. 1, although with different intensities or temperatures, we expect stability against thermal cycling below room temperature also in the present case.

In the as-prepared condition, the presence of interstitial O was clearly detected in the anelastic spectra of all the samples. The content of excess O was reduced by heating in high vacuum up to 790 K.

III. RESULTS

Figure 2 presents the anelastic spectrum of the sample with $x=0.019$ after O reduction, measured during the same cooling run at three excitation frequencies corresponding to the 1st, 3rd, and 5th flexural modes. The whole spectrum shifts to higher temperature for higher measurement frequency, indicating that all the processes below 300 K are thermally activated with relaxation rates $\tau^{-1}(T)$ which are increasing functions of temperature. Since the resonant frequency for flexural vibrations is proportional to $\sqrt{E}$, the relative change of Young's modulus dE/E is given by $[f(T)/f_0]^2-1$; for each mode, the reference frequency f_0 has been chosen in order to let dE/E to coincide for all the modes near 30 K, where the relaxation processes are negligible.

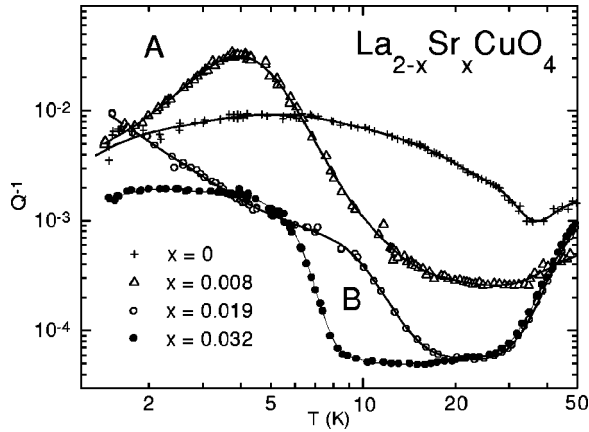The peak around 160 K, labeled T, is due to the coopera-

FIG. 3. Anelastic spectra of reduced $La_{2-x}Sr_xCuO_4$ with $x=0$ (6.2 kHz), $x=0.008$ (6.8 kHz), $x=0.019$ (7.2 kHz), and $x=0.032$ (9.2 kHz).
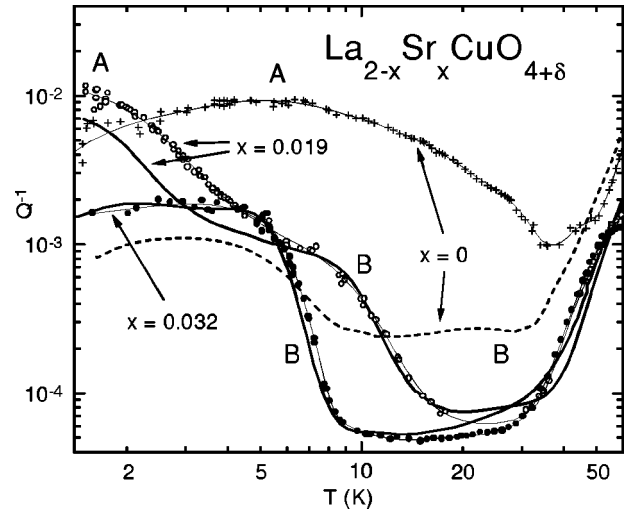

FIG. 4. Effect of the interstitial O present in the as-prepared state on processes A and B. All the data refer to the fifth vibration modes (6.2 kHz for $x=0$, 18 kHz for $x=0.019$, 22 kHz for $x=0.032$). The same symbols of Fig. 2 are used for the reduced state, while the thick lines are for the as-prepared state containing excess O.

tive tilt motion of the octahedra; such a process has been measured in La_2CuO_4 also as a maximum in the [139]La NQR relaxation rate, and has been discussed in detail in Ref. 8. It has been interpreted in terms of propagating tilt waves, with an effective activation energy (2800 K for $x=0$) which is higher than the barrier separating different minima of the multiwell local potential felt by each octahedron, due to the cooperative character of the motion of the octahedra (the calculated barriers are of the order of few hundreds K; see Refs. 14 and 15). Peak T in Fig. 1 is similar to that measured in undoped La_2CuO_4, but with a smaller amplitude and a slightly smaller activation energy (2600 K).

One of the two minor peaks between 60 and 100 K may correspond to a similar anelastic peak which has been observed at higher Sr and Ba doping, and assigned to an electronic relaxation;[16] this process will be dealt with in a separate work.[17]

We will focus on the relaxation which appears below 10 K and is labeled A in Fig. 2, again due to intrinsic lattice fluctuations,[7,18] presumably in correspondence to the tilt waves, whose collective motion is frozen at these temperatures. The central result, shown in Fig. 3, is the dependence of peak A on doping with Sr, after reducing the content of excess O. The peak measured exciting the samples at 6–9 kHz is centered at 5 K for $x=0$, but already at $x=0.008$ the maximum has shifted to 4 K and increased in intensity by 3.5 times;[19] at $x=0.019$ the maximum is shifted below 1.4 K, and at $x=0.03$ the peak is no more visible within the experimental temperature window. It cannot be said whether the relaxation magnitude continues increasing for $x>0.008$, since only the tail of the peak is observable at higher doping levels.

We can state that *doping has a very strong effect on the low temperature relaxation:* it *increases its intensity* and shifts it to lower temperature, which implies through Eq. (1) an *acceleration of the pseudodiffusive dynamics of the octahedra.* A progressive shift of peak A to lower temperature on doping with excess O had already been observed,[7] but small and obscured by a concomitant decrease of the peak intensity due to the blocking effect of interstitial O (see later). The present measurements show that the enhancement of the relaxation rate with doping is huge and also the relaxation

magnitude increases, when the complications from blocking effects are reduced.

The minor step visible below 7 K at $x=0.032$ in Fig. 3, labeled B, is distinct from the main relaxation process A, since it is present also at $x=0.019$, and has a different behavior on O doping. This is shown in Fig. 4, where the absorption is reported both in the outgassed state (same symbols of Fig. 3) and in the as-prepared state (thick lines without experimental points). Interstitial O is present in the samples prepared in air, and its concentration is estimated as $\delta \simeq 0.005$ in our Sr-free samples.[20] We estimate $\delta > 0.002$ and 0.001 for $x=0.019$ and 0.032, respectively, based on the intensity of the relaxation process[21] due to the diffusion of excess O (not shown here).

The above O concentrations are considerably smaller than those of the Sr dopant, but have a profound effect on the relaxation spectrum, since each interstitial O atom blocks several surrounding octahedra into fixed orientations, inhibiting their pseudodiffusive motion within the multiwell potential. Indeed, peak T is completely suppressed in all the samples in the as-prepared state (not shown for $x \neq 0$; see Ref. 7 for $x=0$). The effect of excess O on peak A is less drastic, indicating that the latter mechanism involves a smaller number of octahedra than peak T; it consists of a depression of the intensity and a shift to lower temperature. The peak shift to lower temperature is due to the holes doped by excess O,[7] as discussed later. This is clearly seen in Fig. 4 for $x=0$, and to a smaller extent for $x=0.019$ below 7 K; instead, the step labeled B remains unaffected by the presence of excess O for both $x=0.019$ and 0.032, indicating that its nature is different from that of the major peak. In the following we will discard the small steplike feature B with respect to the main peak A.

IV. DISCUSSION

A. Collective tilt modes of the octahedra

The low temperature relaxational response of the tilt modes of the octahedra may be due to the formation of fluc-

tuating low-temperature tetragonal (LTT) domains in the LTO structure[7] (the LTT phase is actually observed by diffraction experiments only around $x = \frac{1}{8}$ under particular doping conditions[22]). Such domains may correspond to the locally correlated atomic displacements found in a model anharmonic lattice of perovskites,[6] but the case of $La_{2-x}Sr_xCuO_4$ and possibly of other layered cuprates may be different, since they consist of O polyhedra which are connected in two dimensions instead of three. It is even possible that the bidimensional array of connected octahedra is describable as a one-dimensional system.[23] Although recent extended x-ray absorption fine structure[24] (EXAFS) and atomic pair distribution function[25] (PDF) measurements of $La_{2-x}Sr_xCuO_4$ exclude a prevalent LTT local tilt at small x, we think that the model by Markiewicz[23] of a LTO structure arising from a LTT ground state (dynamic Jahn-Teller phase) provides a good framework for analyzing the dynamics of the connected octahedra without charge doping. In the LTT pattern the octahedra are tilted about axes passing through the in-plane Cu-O bonds, so that the tilt of an octahedron determines the staggered tilts of all the octahedra in the same row perpendicular to the rotation axis; instead, the neighboring rows are weakly coupled, since they share O atoms which remain in the Cu plane. In this manner, the system becomes a one-dimensional array of rows of octahedra, with each row described by a single angle of staggered tilts. The one-dimensional nonlinear equation of motion of the tilt angles admits solitonic solutions corresponding to propagating LTO walls which separate LTT domains. The density, thickness and speed of the walls depend on the lattice potential, and an array of closely spaced walls produces an average LTO lattice.[23] Although the mobile charges do not play any role, this picture closely corresponds to the phase of superconducting LTO-like and antiferromagnetic LTT-like stripes that is proposed to be common to all the superconducting cuprates;[26] these tilt waves could well provide a preferred locus for the charge stripes, at least at lower doping.

The combined anelastic and NQR relaxation measurements of the peak T have been interpreted[8] in terms of the propagation of LTO and LTT solitonic tilt waves, similar to those proposed by Markiewicz. Solitonic solutions are generally found in one-dimensional models of anharmonic lattices;[27–29] their spectral density contains a component of pseudodiffusive motion,[27] which produces a central peak observable in NQR experiments[30] and anelastic relaxation[8] of the form of Eq. (1). These solitonic waves are walls between different domains,[27–29] and in the present case of very low doping, where both diffraction and local probes[24,25] indicate a prevalence of LTO tilts, the tilt waves should consist of LTT walls separating LTO domains. They should differ, however, from the usual twin walls between LTO domains, likely responsible for the increased dissipation below the HTT to LTO transformation.[7,12] The possibility should be explored that tilt waves exist, not necessarily separating different LTO domains related to each other by a rotation of 90°, but that can form within a same LTO domain; they would be relatively stable, since the LTT pattern is a local minimum of the potential energy.[14,31] Another difference between the tilt waves responsible for peak T and the twin walls can be that they have little or no correlation along the c axis (making difficult their observation by diffraction).

B. Peak A and the tunneling-driven tilting of the octahedra

The mechanism responsible for peak A should involve the combined motion of few octahedra or even few O atoms, as indicated by the faster dynamics and the reduced blocking effect of O with respect to peak T (involving whole rows of octahedra and completely frozen at the temperature of peak A).

As mentioned above, the relaxing units could consist of fluctuating LTT domains in the LTO structure,[7] corresponding to the locally correlated atomic displacements proposed for the anharmonic lattice of perovskites.[6] On the other hand, it is possible that the more unstable configurations of the octahedra correspond to the frozen tilt waves. As a possible mechanism for peak A, we suggest the propagation of kinks formed on the tilt waves. The formation and propagation of kink pairs could be the elementary steps for bending a straight tilt wave or wall, analogously to the case of dislocations.[32] The formation of a kink pair corresponds to the shift of a wall segment by a lattice unit perpendicularly to the wall itself; it would require a relatively high energy and could contribute to the slower relaxation peak T. Once formed, the kinks could easily propagate or oscillate along the wall, involving the switching of only few O octahedra or atoms.

Another simple mechanism which could explain the low temperature relaxation is the tunneling of single O atoms. Indeed, EXAFS experiments[33] suggest that the apical O atoms close to Sr dopants feel a double-well potential in the c direction. Peak A, however, is rather explainable in terms of tunneling between minima that correspond to tilting of the octahedra. In fact, it is present also in Sr-free samples and its suppression by interstitial O and accelerated dynamics upon doping are easily interpreted in terms of tilt modes.

Let us now consider the issue whether peak A is describable in terms of Eq. (1) with appropriate expressions and distributions for the relaxation strength Δ and rate τ; this is usually assumed in many cases of atomic tunneling, like light or off-center atoms in crystals or the two-level systems (TLS) in glasses.[34] A check which does not require the knowledge of the forms of Δ and τ is to verify that the high temperature side of the absorption peak divided by the measurement frequency $f = \omega/2\pi$ is independent of f. In fact, if Eq. (1) is true for each independent relaxation unit, with τ a decreasing function of temperature, then at temperatures above the maximum relaxation $\omega\tau$ becomes smaller than unity and can be discarded in the denominator; therefore, for the high temperature side of the peak one has $Q^{-1}/\omega \simeq \Delta(T)\tau(T)$, independent of frequency. This is true for any form of $\Delta(T)$ and $\tau(T)$ and even if integrated over distributions of τ and Δ, until the condition $\omega\tau \ll 1$ is satisfied for all the elementary relaxations. Figure 5(b) reports the function $J = Q^{-1}T/f$ (which is proportional to the spectral density of strain and therefore of the atomic motions, see later) for peak A at $x = 0.008$, after subtraction of a constant background shown in Fig. 5(a). The spectral densities measured at the three different frequencies merge on the same curve at high temperature, indicating that Eq. (1), integrated over distributions of τ and Δ, is indeed appropriate for describing peak A, at least for $x = 0.008$.
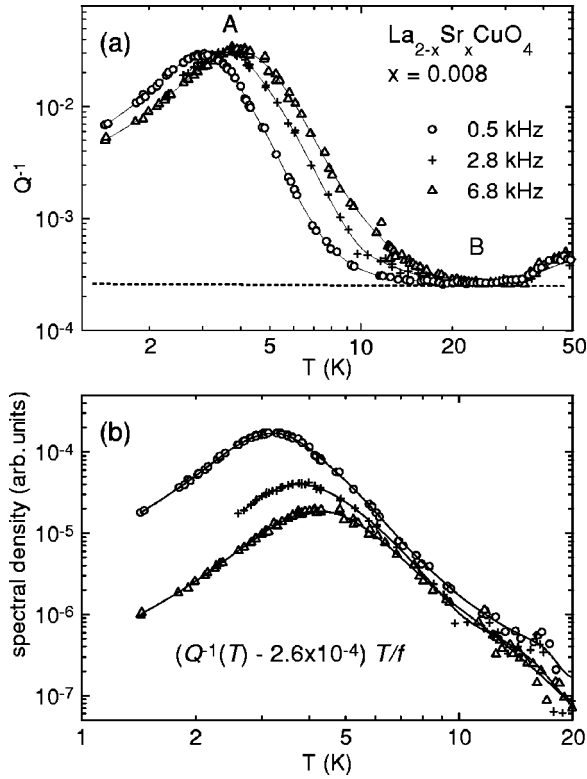
FIG. 5. Peak A for $x=0.008$ measured at three vibration frequencies. The dashed line in (a) is the background which was subtracted in order to derive the spectral density $J(\omega,T)=Q^{-1}T/\omega$ of process A in (b).

The $J(\omega,T)$ obtained for $x=0$ does not satisfy the same condition, because it is described by a broad distribution of $\tau(T)$ which vary with temperature at a slower rate than in the case $x>0$, so that the condition $\omega\tau\ll1$ is not satisfied in the high temperature side of the peak for all the elementary relaxations. Therefore, from the temperature position and shape of peak A we deduce that doping changes the mean relaxation rate τ^{-1} from a slowly varying function of temperature in the undoped case to a function which increases faster with temperature. The high temperature side of the spectral density in Fig. 5(b) indicates that, for $x=0.008$, the rate τ^{-1} increases faster than T^4 above 6 K.

The function $J(\omega,T)$ is proportional to the spectral density of the strain ε, namely the Fourier transform of the correlation function $\langle\varepsilon(t)\varepsilon(0)\rangle$, which in turn is directly related to the correlation function of the displacements of the O atoms, or the tilts of the octahedra. In fact, the absorption may be written as $Q^{-1}=S''/S'$, where $S^*=S'-iS''$ is the complex compliance, with S' the reciprocal of the Young's modulus in the present case, and thanks to the fluctuation-dissipation theorem[35] one has

$$S''=(\omega V/2k_BT)\int dt\, e^{-i\omega t}\langle\varepsilon(t)\varepsilon(0)\rangle, \qquad (2)$$

where V is the sample volume,[36] or

$$J(\omega,T)=\frac{T}{\omega}Q^{-1}(\omega,T)\propto\int dt\, e^{i\omega t}\langle\varepsilon(t)\varepsilon(0)\rangle. \qquad (3)$$

The fact that Eq. (1) describes peak A implies $\langle\varepsilon(t)\varepsilon(0)\rangle=\langle\varepsilon_0^2\rangle e^{-t/\tau}$, which is due to atomic displacements changing at an average rate τ^{-1}. In the tunneling model, such displacements are associated with the transitions of the tunnel system (TS) between its eigenstates. Such transitions are promoted by the interaction between the TS and the various excitations of the solid, generally consisting of emission and absorption of phonons and scattering of the conduction electrons.[34] Instead, the resonant motion of the atoms of the TS between the potential minima while remaining in the same eigenstate produce a peak in the spectral density centered at the tunneling frequency, which is different from Eq. (1) and would be observable only at much higher frequencies.

An important difference between the relaxation process A and those due to the TLS in glasses is that the latter are characterized by an extremely broad distribution of parameters, mainly the tunneling energy t and the asymmetry between the minima of the double-well potential; instead, in $La_{2-x}Sr_xCuO_4$ the geometry of the tunnel systems is much better defined, being some particularly unstable configurations of the octahedra. As a consequence, the TLS relaxation in glasses produces a plateau in the acoustic absorption and a linear term in the specific heat as a function of temperature, whereas in $La_{2-x}Sr_xCuO_4$ we observe a well-defined peak in the absorption, and no linear contribution to the specific heat has been reported.

C. Interaction between the tilts of the octahedra and the hole excitations

We now argue that the marked acceleration of the local fluctuations with doping (narrowing and shift to lower temperature of peak A) is the manifestation of a direct coupling between the tilts of the octahedra and the holes, similarly to the TS in metals, whose dynamics is dominated by the interaction with the conduction electrons.[34] The transition rate of a TS is generally of the form

$$\tau^{-1}(T)\propto t^2 f(T) \qquad (4)$$

$$f(T)=f_{ph}(T)+f_{el}(T), \qquad (5)$$

where t is the tunneling matrix element and $f_{ph}(T)$ and $f_{el}(T)$ contain the interaction between the TS and the phonons and electrons, whose excitation spectra depend on temperature and hole density. Doping is expected to reduce the potential barriers between the different tilts of the octahedra,[7] and therefore to increase t. This is explained in terms of a reduction of the lattice mismatch between the CuO_2 planes and the La/Sr-O layers,[37] which is thought to be the driving force for the octahedra tilting; the more evident manifestation is the decrease with doping of the HTT/LTO transition temperature and of the average tilt angle in the LTO phase.[38,39] Therefore, the introduction of few percent of holes may increase the tunneling frequency $t/\hbar$, and possibly slightly modifies the phonon spectrum, affecting also $f_{ph}(T)$; however, these are expected to be minor changes, certainly not enough to cause the qualitative change of the relaxation rate from the undoped to the $x=0.008$ case. The greater part of the enhancement of the relaxation rate has to be attributed to the increased interaction with the doped holes, corre-

sponding to the term $f_{el}(T)$. Similar effects are observed on the TS's in metals, whose dynamics is dominated by the interaction with the conduction electrons.[34] For example, the relaxation rate of interstitial H tunneling near an O atom in Nb sharply drops below the critical temperature,[40,41] when the opening of the superconducting gap reduces the possibility of scattering of the electrons from the tunneling particle. Models for the interaction between tunneling systems and electrons have been developed and successfully adopted, but the electronic excitation spectrum of the cuprate superconductors is certainly different from that of metals, and new models of the TS-electron interaction are needed. Indeed, the asymptotic behavior of $J(\omega, T)$ at high and low temperature for $x = 0.008$ indicate that $f_{el}(T) \sim T^n$ with $n \sim 3 - 5$; this differs from the temperature dependence of $f_{el}(T)$ for tunnel systems in standard metals, which is less than linear.[34]

It is even possible that the pseudodiffusive lattice modes modify the electron-phonon coupling. In fact, the characteristic frequencies of these modes are far too slow for having any influence on the electron dynamics for $x \leq 0.02$, but increase dramatically with doping. In terms of the multiwell lattice potential, it is possible that at higher doping the barriers between the minima become small enough to give rise to an enhancement of the electron-phonon coupling predicted by some models with anharmonic potentials.[42] Such double-well potentials have been searched for a long time, especially for the apical O atoms in $YBa_2Cu_3O_{6+x}$ (Ref. 43) and $La_{2-x}Sr_xCuO_{4+\delta}$ (Ref. 33) and are thought to play an important role in determining the dominant mechanism of electron-phonon interaction.[42]

D. Static and dynamic tilt disorder

The observation of an increased intensity of peak A at $x > 0$ with respect to $x = 0$ (Fig. 3) is important, since it provides an explanation for the apparent discrepancy between the previous anelastic experiments on $La_2CuO_{4+\delta}$ (Ref. 7) on one side and EXAFS (Ref. 24) and atomic PDF (Ref. 25) measurements on $La_{2-x}Sr_xCuO_4$ on the other side. The acoustic technique sees a dynamic tilt disorder whose effect increases with reducing δ (and therefore doping), while the latter techniques see the opposite effect of an increasing tilt disorder with increasing x. Actually, the EXAFS spectra and atomic PDF are sensitive to both static and dynamic disorder; at $x = 0$ the instantaneous fraction of octahedra swept by the tilt waves is relatively small (few percents according to a crude estimate[8] of the NQR relaxation intensity of peak T) and its effect on the PDF or EXAFS spectra is undetectable. At higher x, the instantaneous tilt disorder will increase due to both the disorder in the La/Sr sublattice and the lattice fluctuations. Instead, the anelastic spectroscopy is sensitive only to the dynamic disorder with characteristic frequency comparable to the measurement frequency; the introduction of interstitial O certainly increases the static tilt disorder but also inhibits the dynamic one, resulting in the depression of peaks A and T observed in Ref. 7. By introducing substitutional Sr, which disturbs the lattice much less than interstitial O, it is possible to observe that doping actually increases the fraction of fluctuating octahedra, and not only the static disorder.

V. CONCLUSIONS

It has been shown that in $La_{2-x}Sr_xCuO_4$ there are pseudodiffusive tilt modes of the O octahedra which give rise to tunneling systems. Such tunneling modes are already present in the undoped state but their relaxation rate and intensity strongly increase with doping. The enhancement of the rate of the local fluctuations is ascribable to the direct coupling between the tilts of the octahedra and the electronic excitations. The form of this interaction is different from the known cases of coupling between tunneling systems and the conduction electrons.

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M. Visscher, S. E. Brown, Z. Fisk, S.-W. Cheong, B. Alten, E. T. Ahrens, K. A. Kubat-Martin, J. D. Maynard, Y. Huang, D. R. Kirk, K. A. Gillis, H. K. Kim, and M. H. W. Chan, Phys. Rev. B **41**, 2098 (1990)] and interpreted as domain wall relaxation or an anharmonic phonon mode.

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