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Photoinduced expansion of cuprate superconductors: Evidence of strong electron-lattice coupling

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Intense light pulses cause "colossal" (larger than thermal) expansion in cuprate films. We show that a simple ionic model accounts for both the static expansion caused by chemical doping and the photoinduced dynamic expansion. Because of the (static or intermittent) reduction of ionic charges, the ions feel lesser electrostatic attraction and move apart. The displacements can be large since the Madelung energy can change by more than 1 eV/atom. From this ionic picture, one would expect significant structural reconstruction at heterointerfaces and photoinduced shifts in Raman frequencies.

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Whether coupling of charge excitations to the crystal lattice in cuprates and other strongly correlated oxides is strong and crucial[1] or weak and essentially irrelevant[2] for the mechanism of high-temperature superconductivity (HTS) has been a matter of avid debate. We believe that a recent experimental report[3] of "colossal" photoinduced expansion in $La_2CuO_{4+\delta}$ (LCO) allows an unambiguous answer at least to the question whether this coupling is strong or not. In the experiment of Gedik *et al.*,[3] a thin LCO film was exposed to intense femtosecond light pulses and the evolution of crystallographic structure was monitored in real time by pulsed electron diffraction. The c-axis lattice constant was observed to intermittently expand by as much as 0.3 Å. On the length scale characteristic of amplitudes of thermal lattice vibrations, such lattice distortion is *colossal*—the crystal should have melted. Large (static) expansion also occurs in cuprates upon chemical doping[4,5] and, as we will show below, it cannot be simply ascribed to the difference between the ionic radii of host and dopant ions.

Model. Generally, lattice expansion accompanies a reduction in crystal cohesion energy. In cuprates (and other related oxides), the cohesion is primarily of ionic origin and the crystal energy can be estimated as follows:

$$U = (e^2/2)\Sigma q_i q_j/|\mathbf{r}_i - \mathbf{r}_j| + (1/2)\Sigma A_{ij}\exp(-B_{ij}|\mathbf{r}_i - \mathbf{r}_j|). \quad (1)$$

The first term is the Madelung energy (U_M); $\mathbf{r}_i$, $\mathbf{r}_j$ denote the positions of the ions and q_i, q_j are their charges (in units of e). The second term is the core repulsion energy (U_c) modeled as the sum of nearest-neighbor repulsion terms in the standard Born-Mayer form. The sums are over all i,j with $\mathbf{r}_i \neq \mathbf{r}_j$. This conceptually simple framework accounts well for a range of material properties—the cohesion energy, the charge-transfer gap, the crystal structure, the compressibility, and even the phonon frequencies and the critical pressure for the orthorhombic-to-tetragonal phase transition.[6] We fix the parameters A_{ij} and B_{ij} in such a way that the minimum of total energy U coincides with the experimentally determined structure.[4] We do not explicitly treat covalency of the in-plane Cu-O bonds and van der Waals interactions; these are included implicitly through the choice of A_{ij} and B_{ij} and are very small ($<1\%$) terms anyway.[6,7] This is adequate in the present context since we are only interested in small relative changes of structure upon doping.

In Fig. 1(a), we show a simplified model of the crystallographic structure of LCO. The drawing shows only one-half of the crystallographic unit cell; the actual cell contains 14 atoms (2 f.u.) and is twice taller. Note that the La-O layers are not flat (as they are usually depicted) but very strongly corrugated because of electrostatic forces—the distance between the La plane and the nearest O plane is $c_3 = 0.59$ Å. If the cations were La^{2+} and Cu^{4+}, the LaO and CuO_2 layers would have been neutral. However, the strong chemical force (i.e., the large energy difference between the La^{2+} and Cu^{4+} levels) strongly favors La^{3+} and Cu^{2+}. This makes the CuO_2 layer polar; the areal charge density is huge, $-2e$ per 3.8

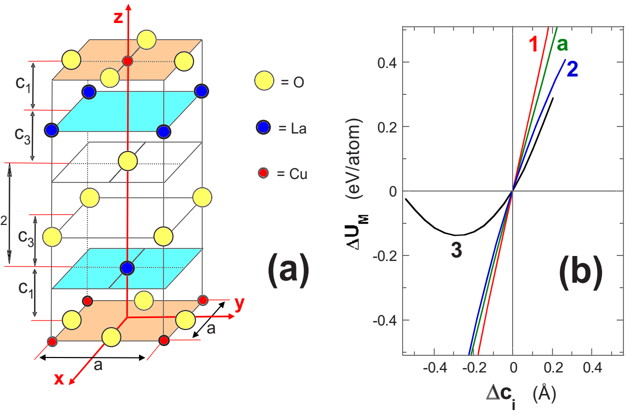

FIG. 1. (Color online) (a) A simplified structural model (one-half the crystallographic unit cell) of La_2CuO_4. At room temperature, the structure is tetragonal, as shown, and the space group is *I*4/*mmm*. The distance from the CuO_2 plane to the La atom is $c_1 = 1.82$ Å, from La to the O atom right above it $c_2 = 2.33$ Å, and from the La plane to the nearest O plane $c_3 = 0.59$ Å. Altogether, $c = 4c_1 + 2c_2 + 2c_3 = 13.12$ Å, while $a = 3.80$ Å (Ref. 4). Note that the La-O layers are not flat (as they are usually depicted) but very strongly corrugated; in this sketch, the La-O corrugation length c_3 is exaggerated for clarity. (b) The dependence of the calculated Madelung energy per atom (U_M) on the displacements of atoms from their equilibrium positions, Δc_i ($i = 1, 2, 3$) and Δa, where all other coordinates are kept fixed. Notice the anomalous dependence of U_M on c_3.

$\times 3.8$ Å^2 plaquette, which equals -2 C/m^2, much higher than in any existing ferroelectric material. This generates enormous internal electric fields ($\sim 10^9$ V/cm) between the layers; as a consequence, the lattice distorts—the negatively charged $(CuO_2)^{2-}$ plane repels O^{2-}, which moves away while La^{3+} shifts toward the $(CuO_2)^{2-}$ plane. As a result, in LCO, there is no "hard contact" between Cu and the apical oxygen (O2). The Cu-O2 distance (2.41 Å) is a good deal larger than the sum of the respective ionic radii, $R(Cu^{2+})+R(O^{2-}) \approx 1.9$ Å. The very concept of "Cu–apical oxygen bond" is questionable—if a CuO molecule were stretched to 2.41 Å, it would dissociate. More surprisingly, the dependence of U_M on c_3 (while keeping a, c_1, and c_2 fixed at their equilibrium values) shows a minimum at $c_3=0.32$ Å [see Fig. 1(b)]. This is quite unusual—U_M is a monotonous function (with a negative curvature) of all other lattice parameters and it generally diverges as $r \to 0$. The explanation of this "paradox" is that the LCO structure can be perceived as a set of rigid comblike layers that "levitate" in the electrostatic Madelung potential.

To calculate U from Eq. (1), we have used the following Born-Mayer parameters: $A_{\text{Cu-O}}=0.30$ keV, $A_{\text{La-O}}=1.18$ keV, $A_{\text{O-O}}=0.29$ keV, and $B_{\text{Cu-O}}=B_{\text{La-O}}=B_{\text{O-O}}=2.66$ Å^{-1}. With this choice, we reproduce accurately the experimental crystal structure of undoped La_2CuO_4,[4] as well as its bulk modulus $B=113$ GPa.[8] In what follows, we have kept these values constant—there were no open fitting parameters in the calculations presented here. However, our findings and conclusions are actually remarkably robust with respect to the choice of parameters since to the first order they are determined by the change in the Madelung energy, which is an order of magnitude larger than the core repulsion ($U_M=-390$ eV and $U_c=59$ eV per unit cell).

Chemical doping. We first discuss the effects of Sr→La doping in $La_{2-x}Sr_xCuO_{4-\delta}$ (LSCO). This is a stringent test of our model because the dependence of all atomic positions on the doping level x is known in detail from comprehensive neutron diffraction data.[4] Since the LSCO samples in Ref. 4 had some oxygen deficiency δ, we assume the actual number of holes (per f.u.) to be $h \approx x-2\delta$. We model (roughly) this situation by putting $q(\text{La})=3-0.5h$; since the holes are known to distribute between the copper and in-plane oxygen (O1) atoms with roughly 20% weight on Cu, we let $q(\text{Cu})=2+0.2h$ and $q(\text{O1})=-2+0.4h$. Then, we compute $U(h;a,c_1,c_2,c_3)$ and find the position of its minimum as a function of the doping level h. In this way, we obtain, without any fitting, the new atomic coordinates $c_i(h)$ as well as the new lattice constants $a(h)$ and $c(h)$, which in Fig. 2 we compare to the experiment. The agreement is good; it is even better for Ba→La doping[5] since Ba is closer than Sr to La. Note that for simplicity and clarity, we kept the Born-Mayer parameters fixed; thus, we in effect ignore the differences between the ionic radii[9] of Sr^{2+}, Ba^{2+}, and La^{3+}. As it should be clear from Fig. 2, this does not affect the conclusions qualitatively; the first-order effect is the change in Madelung energy. We have not captured accurately the observed small contraction of the a lattice constant because we have not included a small negative contribution to U that comes from (attractive) metallic bonding that arises when insulating LCO is made metallic by doping. However, this effect can be safely ignored in what follows since the photoinduced experiments were made on thin LCO films epitaxially anchored to the substrate.

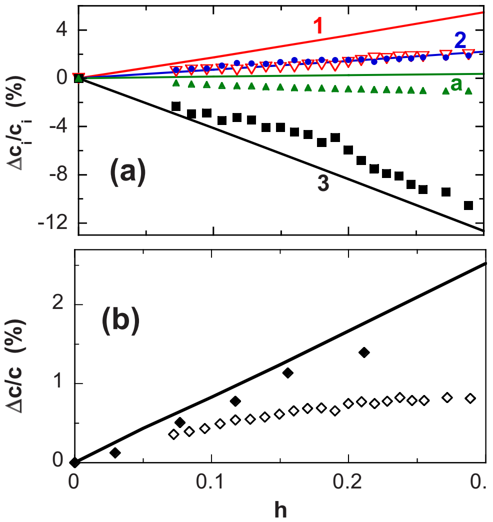

FIG. 2. (Color online) Lattice distortions due to chemical doping. (a) $\Delta c_i/c_i$ ($i=1,2,3$) and $\Delta a/a$ as functions of the hole concentration $h=x-2\delta$: the present calculations (solid lines) vs the experiment on $La_{2-x}Sr_xCuO_{4-\delta}$ (Ref. 4); ▽ stands for $\Delta c_1/c_1$, ● for $\Delta c_2/c_2$, ■ for $\Delta c_3/c_3$, and ▲ for $\Delta a/a$. (b) The same for $\Delta c/c$: the present calculations (solid line) vs the experiment on (◇) $La_{2-x}Sr_xCuO_{4-\delta}$ (Ref. 4) and (◆) $La_{2-x}Ba_xCuO_{4-\delta}$ (Ref. 5).

Since the lattice expands overall, it may appear surprising that dc_3/dh is not only huge in magnitude but *negative*. The explanation is that La moves away from the CuO_2 plane four times more than O2 and, consequently, the La-O2 corrugation decreases. Note also that both the Madelung and the cohesive energy change a lot: upon doping from $x=0$ to $x=0.4$, U changes by 2 eV/atom and U_M by 2.5 eV/atom. This has some significant implications. First, one must question whether it is possible, in principle, to quantitatively explain the entire (x,T) phase diagram just by varying the chemical potential in some fixed effective low-energy Hamiltonian. Next, the dramatic doping dependence of cohesive energy should be considered when interpreting the results of transport measurements in LSCO-based heterostructures.[10] In particular, one should expect structural (atomic) reconstruction near the heterointerfaces, e.g., between LSCO and $LaSrAlO_4$ (LSAO); otherwise, a disruption in the pattern of alternation of charged layers would cause the "polarization catastrophe."[11] Indeed, high-resolution cross-section transmission electron microscopy has already detected a sequence of several strange atomic monolayers (an "interface compound") between the LCO film and the LSAO substrate.[12] We believe that such interface reconstruction in oxide heterostructures could be understood and predicted by the use of ionic models akin to the present one.

Photodoping. Absorption of visible light polarized parallel to the CuO_2 planes causes charge-transfer excitation between O1 and Cu atoms.[13] We model this by assuming that the charge transfer (Δq per CuO_2 unit) is uniformly averaged over the entire CuO_2 plane, i.e., we let $q(\text{Cu})=2-\Delta q$, $q(\text{O1})=-2+\Delta q/2$, and determine the crystal configuration at which the new potential energy surface $U(\Delta q;a,c_1,c_2,c_3)$

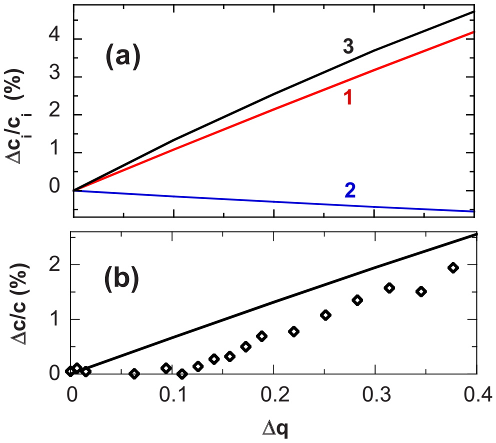

FIG. 3. (Color online) Lattice distortions due to photodoping. (a) $\Delta c_i/c_i$ as functions of Δq, the photoinduced charge transfer from O1 to Cu. (b) The concomitant lattice expansion $\Delta c/c$: the present calculations (solid line) vs the experiment (◇) (Ref. 3). The Born-Mayer parameters are the same as in Figs. 1(b) and 2. In the experiment, the large shifts in the equilibrium ion positions are only intermittent and they are related to large-amplitude coherent acoustic vibrations of the lattice.

has the minimum. In Fig. 3, we show the dependence of calculated relative displacements $\Delta c_i/c_i$ on Δq. Since in the experiment of Gedik *et al.* the LCO films were grown epitaxially on $LaSrAlO_4$ substrates,[10] here we keep the in-plane lattice constants fixed. The calculated expansion $\Delta c/c$ is in good agreement with the experiment [see Fig. 3(b)]. The model accounts for the most striking aspect of the data—colossal expansion. To be more precise, the static model only provides the maximum displacement that one could reach under dynamic excitation.[14] Interestingly, for high fluence, the experimental data come close to this limit. The actual dynamics is complex and involves large-amplitude *coherent* vibrations of the lattice; its quantitative description is beyond the scope of the present study.

So far, in femtosecond photodoping experiments, only the overall expansion of the unit cell has been measured. We predict significant intracell displacements of La and O2 ions as well. In particular, the corrugation c_3 should increase, in contrast to what happens in the case of chemical doping. The reason is that chemical doping changes the overall charge of CuO_2 and La layers, while light only causes intermittent charge transfer from O1 to Cu.

The fact that photodoping induces colossal expansion implies very strong coupling of charge-transfer excitations to the lattice. We see no alternative explanation—lattice expansion does not come from magnons, plasmons, excitons, etc. More specifically, expansion along the *c* axis can only come from *c-axis longitudinal acoustic phonons* (cLAPs). This strong coupling to cLAPs is unusual and characteristic of charged-layer superlattice structures; yet, it has been overlooked so far.

On the other hand, according to the present calculations, the coupling to out-of-plane optic vibrations is also strong. This can be shown by expanding the calculated atomic displacement pattern in terms of normal modes of the system, $\mathbf{Q}=\Sigma\alpha_i\mathbf{Q}_i$. It turns out that, apart from the *c*-axis expansion, there are only two contributors: (i) $\mathbf{Q}_1$, the $k=0$ vibration of the La dumbbell, a Raman active mode of A_{1g} symmetry at about 230 cm^{-1} (29 meV); (ii) $\mathbf{Q}_2$, the $k=0$ vibration of the O2 dumbbell, also A_{1g} and Raman active, at 440 cm^{-1} (55 meV).[15] A strong response from high-frequency optic vibrations clearly indicates that the effect cannot be of thermal origin. Colossal photoinduced expansion comes from large-amplitude coherent excitation of lattice vibrations.[14] This explains how the melting limit (the so-called Lindeman criterion) can be exceeded—it only applies to *incoherent* thermal vibrations that destroy the long-range crystalline order.

The large change in the Madelung energy upon chemical doping is reflected also in significant shifts of phonon frequencies observed, e.g., in Raman spectroscopy. For example, the $\mathbf{Q}_1$ mode frequency at room temperature shifts from 227 cm^{-1} in La_2CuO_4 to 215 cm^{-1} in $La_{1.7}Sr_{0.3}CuO_4$, i.e., by about -6%.[15] The intracell distortion induced by photodoping, as predicted in Fig. 3(a), should cause similar shifts; this should be observable either by picosecond-resolved frequency-domain Raman spectroscopy or even better by time-domain spectroscopy of coherent optic phonons.[14]

Our simple, homogeneous model depicts a spatially and temporally averaged situation. Any electronic inhomogeneity would bring about even *larger* local lattice distortion and *stronger* coupling to the lattice. Indeed, several local experimental probes have detected intracell ion displacements as large as 0.2 Å, much larger than in our model—as if the extra charges were rather localized.[16] Next, we have not examined here the electron conduction but from the above analysis, it is clear that any redistribution of charge in the CuO_2 plane will cause a strong response of nearby La and O2 ions—in other words, the coupling of moving electrons to the out-of-plane modes ought to be strong.

Naturally, one wonders about the bearing of these findings on the key question—the mechanism of HTS. Our model calculations—and the experiment of Gedik *et al.*—show that electron-lattice coupling in cuprates is strong, as it was postulated earlier.[1] However, we expect the same effect to occur in other structurally and chemically similar ionic oxides that are not superconducting at all, so clearly this cannot be the whole story. Nevertheless, the fact that in-plane charge excitations are strongly coupled to *c*-axis lattice vibrations—that *these excitations cannot be separated*—should not be ignored since they are ultimately responsible for both the normal state conductivity and HTS. It is a challenge to the theory to incorporate this together with other aspects such as strong electronic correlations.[2] For example, the large change with chemical doping of the Madelung potential at Cu and O1 positions must affect the hopping integral t and hence the superexchange interaction $J=t^2/U$, where U is the on-site Coulomb repulsion.

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