Model for an extra phonon mode in La_2CuO_4 and La_2NiO_4

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Phonon dispersion curves for two-dimensional analogs of La_2CuO_4 and La_2NiO_4 are modeled by adding a scalar degree of freedom to mimic a low-lying charge-fluctuation mode. The resulting model bears a close resemblance to the "extra phonon mode" seen in neutron scattering by Pintschovius *et al.* In particular, the mode is invisible to neutrons at $\mathbf{Q}=\mathbf{0}$ because scalar symmetry prevents admixture with vibrational modes at $\mathbf{Q}=\mathbf{0}$. Possible interpretations of this new mode are suggested.

Pintschovius, Reichardt, and co-workers[1-4] have measured phonon dispersion curves by inelastic neutron scattering on single crystals of the semiconductors La_2CuO_4 and La_2NiO_4. Among many anomalies, the most interesting was the appearance in La_2CuO_4 of an "extra mode" with $\hbar\omega\simeq 60$ meV, seen when the wave vector $\mathbf{Q}$ is not too close to $\mathbf{Q}=\mathbf{0}$. Very recently, in a single crystal of superconducting $La_{1.9}Sr_{0.1}CuO_4$, the extra mode has again been found,[5,6] with similar properties, but reduced frequency $\hbar\omega\approx 40$ meV. This dramatic softening contrasts with the majority of the modes which are much less strongly altered by doping. It is natural to speculate that this mysterious extra phonon is closely connected with the mystery of why T_c is high. If this mode is electronic in origin (as proposed by the authors of the experimental papers[2,5,6]), then it can be seen by neutrons only when it mixes with nuclear displacement. (The direct coupling of the neutron to the electron spin is significantly smaller than the coupling to nuclear density.) I present a model which has this property and show that it behaves qualitatively like experimental observations. This model invokes a single extra scalar degree of freedom in each unit cell, which couples to local compressions of the planar oxygen atoms. Speculations are given about the possible interpretation of this scalar variable. The model is also applied to La_2NiO_4 and suggests that an extra mode might occur here also, at higher ω, offering a unification of the "breathing-mode" anomalies seen in these materials.

Rather than cloud matters by modeling all 21 branches of phonons in an admittedly oversimplified way, I prefer to attempt a model of minimum complexity which contains the essential idea. Therefore, consider a two-dimensional (2D) plane of CuO_2, with three atoms per unit cell as shown in Fig. 1(a). Consider only displacements in the plane, denoted by vectors $\mathbf{u}(\mathbf{1})$ for the Cu atom at $\mathbf{1}=ma\hat{\mathbf{x}}+na\hat{\mathbf{y}}$ and $\mathbf{v}(\mathbf{1}\pm\frac{1}{2}a\hat{\mathbf{x}})$ or $\mathbf{v}(\mathbf{1}\pm\frac{1}{2}a\hat{\mathbf{y}})$ for the bridging oxygen atoms. As in Fig. 1(a), two springs are used, with constants K_1 for the Cu—O stretch and K_2 for the O—O stretch (or O—Cu—O bond bend). The Hamiltonian is

$$H_0=\tfrac{1}{2}\sum[M\dot{\mathbf{u}}(\mathbf{1})^2+m\dot{\mathbf{v}}(\mathbf{1}+\tfrac{1}{2}a\hat{\mathbf{x}})^2+m\dot{\mathbf{v}}(\mathbf{1}+\tfrac{1}{2}a\hat{\mathbf{y}})^2]+\tfrac{1}{2}\sum\{K_1[u_x(\mathbf{1})-v_x(\mathbf{1}+\tfrac{1}{2}a\hat{\mathbf{x}}]^2+3\text{ others}\}$$
$$+\tfrac{1}{4}\sum\{K_2[v_x(\mathbf{1}+\tfrac{1}{2}a\hat{\mathbf{x}})-v_y(\mathbf{1}+\tfrac{1}{2}a\hat{\mathbf{x}})-v_x(\mathbf{1}+\tfrac{1}{2}a\hat{\mathbf{y}})+v_y(\mathbf{1}+\tfrac{1}{2}a\hat{\mathbf{y}}]^2+3\text{ others}\}\ . \quad (1)$$

The masses of Cu and O atoms are denoted by M and m, respectively. This model has six degrees of freedom per unit cell and, therefore, has two acoustic and four optic branches, obtained by solving a 6×6 matrix. If frequencies are measured in dimensionless units $\omega(2K_1/m)^{-1/2}$ and using the experimental mass ratio $m/M=0.252$, the dispersion curves have a single free parameter K_2/K_1. In Fig. 1(c) the curves are plotted for $K_2/K_1=0.2$. Note that a single zero eigenfrequency occurs at $\mathbf{Q}=(\pi/a)(1,1)$. This "soft mode" corresponds to rigid twists of square CuO_4 units [with M_4 symmetry, shown in Fig. 1(d)]. The model of Fig. 1(a) clearly has no springs to inhibit such a twist. In the perovskite-structure material $SrTiO_3$, exactly this twisting instability is found to condense to a static distortion at $T=110$ K. La_2CuO_4 and La_2NiO_4 have a closely related instability involving twists of CuO_6 octahedra around [110] rather than [001] axes (the tetragonal to orthorhombic transition). Thus the soft twisting mode should not be regarded as an unrealistic feature of the model. The highest-energy phonon of the model is the breathing mode of oxygen atoms at $\mathbf{Q}=(\pi/a)(1,1)$ [with M_1 symmetry, shown in Fig. 1(e)]. This mode seems[2-6] to couple to the extra mode, and it is expected to couple to charge fluctuations on the CuO_4 complexes.

To keep the model as simple as possible, it will remain ordered and harmonic. Then an extra mode can only enter if a new degree of freedom is added. Consider a scalar degree of freedom $\rho(\mathbf{1})$ as described by

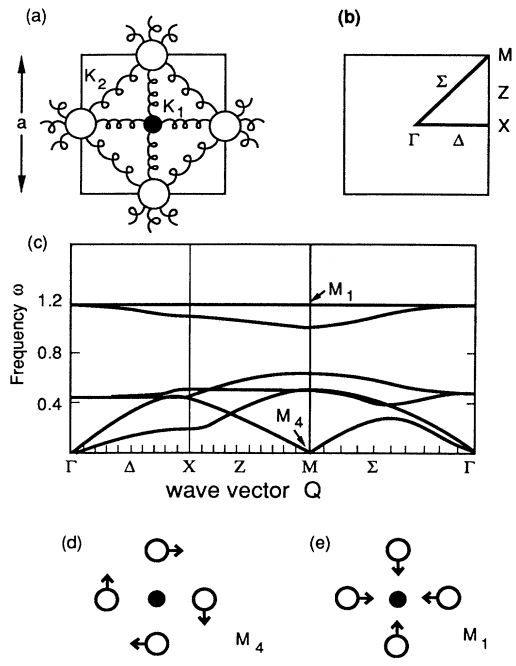

FIG. 1. Unit cell of planar CuO_2 in (a) direct and (b) reciprocal space. Open circles are oxygens. The dispersion curves corresponding to $K_2/K_1=0.2$ are plotted in (c). The "twist" (M_4) and "breathing" (M_1) modes are marked and sketched in (d) and (e).

$$H'=\tfrac{1}{2}\sum[\mu\dot{\rho}(\mathbf{1})^2+C\rho(\mathbf{1})^2]$$
$$+\sum\{R\rho(\mathbf{1})[v_x(\mathbf{1}-\tfrac{1}{2}a\hat{\mathbf{x}})$$
$$-v_x(\mathbf{1}+\tfrac{1}{2}a\hat{\mathbf{x}})+v_y(\mathbf{1}-\tfrac{1}{2}a\hat{\mathbf{y}})$$
$$-v_y(\mathbf{1}+\tfrac{1}{2}a\hat{\mathbf{y}})]\}\ . \qquad (2)$$

Normally, when extra degrees of freedom are added in lattice dynamics (as in a shell model), no mass is assigned, because they are adiabatically eliminated by a condition like $-\partial H/\partial\rho(\mathbf{1})=0$. Here we replace zero on the right-hand side by classical dynamics, $\mu\ddot{\rho}(\mathbf{1})$. Doubtless any actual extra degrees of freedom would have more complicated quantum dynamics, but I postulate that the low-energy collective modes of these quantum degrees of freedom can be described by harmonic oscillators of effective mass μ and self-restoring force C. The coupling of $\rho(\mathbf{1})$ to lattice degrees of freedom $\mathbf{u}(\mathbf{1}),\mathbf{v}(\mathbf{1})$ is fixed to within an adjustable coupling constant R by the assumption that $\rho(\mathbf{1})$ has scalar symmetry. Similar scalar (but adiabatic) models have been proposed for metals like Nb,[7,8] and rules for working out coupling terms were given in Ref. 8. There are now seven degrees of freedom and seven eigenfrequencies. Two new dimensionless parameters $r_1=Cm/K_1\mu$ and $r_2=Rm^{1/2}K_1\mu^{1/2}$ govern the modified dispersion curves. Figure 2 shows these curves for two special choices of (r_1,r_2).

At small Q the symmetric compression scales as $|\mathbf{Q}|$, and at $\mathbf{Q}=\mathbf{0}$ the extra mode in completely decoupled from the lattice, with eigenfrequency $(C/\mu)^{1/2}$. At $\mathbf{Q}=(\pi/a)(1,1)$, the extra mode couples only to the M_1 breathing mode, while at general $\mathbf{Q}$ points the extra mode is coupled to all six phonons. Experiments[1-4] show that in La_2NiO_4 the breathing mode is softened at the $(\pi/a)(1,1)$ point, as expected because of coupling to high-energy charge fluctuations, but in La_2CuO_4 the breathing mode occurs at a surpisingly high frequency, as if pushed up in energy by a lower-lying extra mode. In Fig. 2 parameters (r_1,r_2) are chosen to mimic these behaviors. The extra mode in Fig. 2(a) lies below the breathing mode as seen in La_2CuO_4, while in Fig. 2(b) it lies above the breathing mode, causing the softening seen in La_2NiO_4. Unlike the case for La_2CuO_4, the experiment in La_2NiO_4 did not find a clear extra mode [except near $Q=(\pi/a)(0.6,0.6)$, where the breathing mode had a split peak line shape]. Therefore, I propose that it would be worth extending the neutron measurements to higher energies in La_2NiO_4 to look for signs of an extra mode lying above the breathing mode. Of course, the model cannot be taken literally, and it is impossible to predict exactly the energy or the width of the charge fluctuation band causing the softening, but the analogy with the case of La_2CuO_4 is striking and worth pursuing. In fact, possible evidence for an extra mode in La_2NiO_4 already exists in Fig. 6 of Ref. 1, where direct measurement of the phonon density of states has an extra bump at high ω relative to the spectrum derived from the measured dispersion curves.

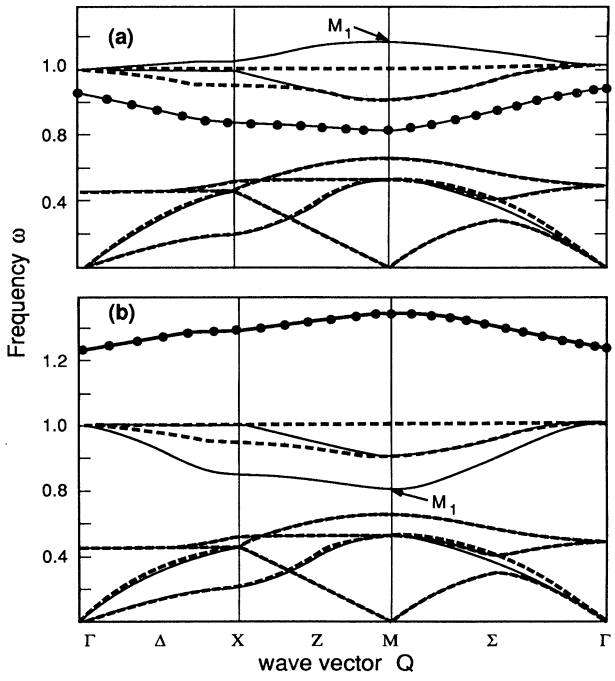

FIG. 2. Dispersion curves after adding the scalar degree of freedom. The new mode is denoted by the line through solid circles. Previous results (without scalar, $K_2/K_1=0.2$) are denoted by dashed lines. Case (a) (La_2CuO_4) has $r_1=1.1$, $r_2=0.4$. Case (b) (La_2NiO_4) has $r_1=2.8$, $r_2=0.9$.

To confirm the model of Fig. 2(a) as a reasonable caricature of La_2CuO_4, it is necessary to consider the neutron line shape. The neutron cross section is taken as the usual Van Hove correlation function $S(\mathbf{q},\omega)$ or nuclear density-density correlation function. The coherent one-photon part has been calculated and is shown in Fig. 3. The extra mode is silent at $\mathbf{Q}=\mathbf{0}$ because it has no nuclear component, whereas at $\mathbf{Q}=(\pi/a)(1,1)$ it mixes strongly with oxygen breathing and has as much intensity as the higher-energy branch. This behavior mimics closely what was seen in the experiment.[2,4,5]

Density functional (local density approximation) band theory has had excellent success at understanding structural energies, although vibrational frequencies (by the "frozen phonon" method) have only been calculated[9] at $\mathbf{Q}=\mathbf{0}$ and $\mathbf{Q}=(\pi/a)(1,1,0)=X$. For La_2CuO_4, Ref. 9 obtained a value $\hbar\omega_{\text{theor}}=91$ meV for the X-point breathing mode, remarkably close to $\hbar\omega_{\text{expt}}=88$ meV. The next lowest mode of X_{1g} symmetry was predicted to lie at $\hbar\omega_{\text{theor}}=59$ meV, which agrees well with the extra mode. The eigenvector was found to be dominated by the transverse oxygen "scissors" motion, which can also be viewed a breathing around the empty sites at the corners of Fig. 1(a). In Ref. 9 the mode is referred to as "quadrupolar," but the authors have assured me that they do indeed refer to the scissors distortion of X_{1g} symmetry. The longitudinal quadrupolar distortion where O_x atoms breathe toward Cu while O_y breathe away, belongs to a different representation X_{2g} and was not calculated in Ref. 9. The present model also associates the 60-meV extra mode with X_{1g} symmetry, but proposes that the component of the eigenvector which is nuclear rather than electronic should closely resemble the X_{1g} breathing mode. These divergent theoretical suggestions can be tested by an experimental determination of the eigenvector.

Now I speculate on the possible origins of this extra mode, looking first at nonelectronic interpretations. Perhaps, the most trivial possibility is an origin in crystalline disorder. A good reference experiment in this respect is the study by Tsunoda *et al.*[10] of phonons in $Ni_{1-x}Pt_x$ alloys. For $x=0.05$ the heavy Pt impurities have quasilocalized vibrations ("resonant modes") degenerate with the continuum. These modes are not seen directly, but induce "breaks" in the dispersion curves at the resonant frequency with two-peaked line shapes in restricted portions of Q space, similar to what was seen in the breathing mode of La_2NiO_4.[1] For $x=0.30$ the resonant mode is seen over much of the Brillouin zone and appears as a splitting of the TA branch into two components, slightly reminiscent of La_2CuO_4. Difficulties with this explanation are (1) lack of a plausible dense defect to interact with oxygen breathing at 60 meV, (2) failure to observe evidence for defect modes at lower energies, and (3) the narrow width seen experimentally for the extra mode.

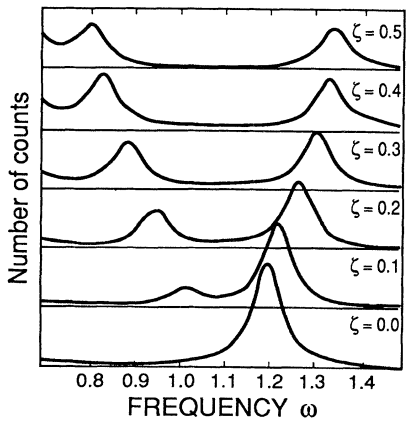

FIG. 3. $S(\mathbf{Q},\omega)$ vs ω for six values of $\mathbf{Q}=(2\pi/a)(3-\zeta,3-\zeta)$, for comparison with Fig. 3 of Ref. 2 or Fig. 5 of Ref. 3. The vertical scale is the same for all Q's.

Anharmonicity could conceivably cause an extra mode, and high-T_c oxides are doubtless quite anharmonic, but objections (2) and (3) above would still apply. As an example, CuCl is a very anharmonic material for which Raman spectroscopy[11] has seen an extra or, more properly, "split" $\mathbf{Q}=\mathbf{0}$ optic mode. However, neutron scattering[12] away from $\mathbf{Q}=\mathbf{0}$ sees only the normal number of broad phonons, with broadening becoming so pronounced that well-defined quasiparticle peaks mostly disappear by 300 K. La_2CuO_4 is by comparison relatively harmonic.

Magnetic collective modes also exist in these materials. However, their dispersion is believed to be understood[13] and bears little relation to the "extra phonon" in La_2CuO_4. Furthermore, the magnetic form factor falls rapidly with $|\mathbf{q}|=|\mathbf{Q}+\mathbf{G}|$, leaving little oscillator strength near the $\mathbf{G}=(2\pi/a)(3,3,0)$ point where the extra mode was seen in Refs. 2–6.

If we reject defects, anharmonicity, and magnetism, we are left with electronic mechanisms. The extra mode bears little resemblance to any of the conventional electronic collective excitations: plasmons, interband excitations, excitons (bound interband excitations), Cooperons (fluctuating Cooper pairs), or bipolarons (strongly bound Cooperons). However, many electron spectroscopies [infrared, Raman, and electron energy loss[14–17] (EELS)] have reported unconventional spectra which also do not seem to fall into the usual categories. If the extra mode is electronic, it must be linked to some or all of these anomalies.

The most conventional possibility is plasmons. The crystals used for neutron scattering were not perfectly semiconducting, but were partially metallic owing to structural imperfections. Gervais and co-workers[18,19] did infrared measurements on exactly the same crystals. The spectra with $\mathbf{E}\perp\mathbf{c}$ contained, in addition to phonons, a broad Drude response and a "midinfrared" band. The Drude plasma frequencies ω_p fitted to ir data were 70 meV for La_2CuO_4 and 180 meV for La_2NiO_4, remarkably close to the energies of the extra oscillators used in Fig. 2. However, the widths of the Drude peaks were twice the plasma frequencies, instead of ~ 1 meV wide (i.e., limited by resolution[2–5]). Conventional low-energy plasmons, found in doped semiconductors[20] like PbTe, interact strongly with small Q LO phonons, but disappear at

$Q \sim \omega_p / v_F \ll \pi / a$. Thus, in spite of the astonishing coincidence of the values of ω_P, the extra mode looks nothing like a conventional plasmon. Furthermore, in the doped materials $La_{1.9}Sr_{0.1}CuO_4$, the extra mode is pushed to lower to ω rather than tracking with $\hbar\omega_P$.[5,6]

A midinfrared band seems to occur in all high-T_c superconductors (as well as some related nonsuperconducting materials). This band is often strongest near $\hbar\omega \approx 0.5$ eV, but may extend below 0.1 eV. However, we need a peak in the charge susceptibility $\mathrm{Im}\chi$ or $\mathrm{Im}(\epsilon^{-1})$, not a peak in $\mathrm{Im}(\epsilon)$, to couple to $S(\mathbf{Q},\omega)$ and phonons. Raman and EELS measure $\mathrm{Im}(1/\epsilon)$. An anomalous electronic background has been reported in Raman spectrum of $YBa_3Cu_3O_7$ (but not La_2CuO_4) especially by Klein *et al.*[21] The conventional electronic background falls off for $\hbar\omega > Qv_F$, where Q is very small in a Raman experiment, but several high-T_c materials show broad electronic scattering for $\hbar\omega \gtrsim 0.1$ eV. The mechanism is unknown, as is the $\mathbf{Q}$ dependence.

Mihailović *et al.*[16] have observed photoinduced absorption peaks in insulating La_2CuO_4 at 650 and 480 cm^{-1}. The latter frequency agrees well with the extra mode, whereas the former lies slightly below the highest oxygen bond-stretching vibrations. These authors have noted the correspondence with the extra mode. They interpret their data as arising from a localized vibration in the neighborhood of a self-trapped polaronic electronic excitation created by the pump photons. Since neutron scattering is done under conditions where the polaron levels would be thermally dilute, it is not easy to see how this interpretation can be extended to explain the extra mode. Nevertheless, the photoinduced peak at 480 cm^{-1} may be closely connected with the extra mode.

A recent EELS experiment by Demuth *et al.*[17] reported a feature which behaves somewhat like a superconducting gap $2\Delta \approx 50-70$ meV ($\sim 8k_B T_c$ in $YBa_2Cu_3O_7$ and $Bi_2Sr_2CaCu_2O_8$). As T increases through T_c, the feature becomes harder to see and shifts somewhat down in energy, but may remain at ~ 45 meV even up to 298 K in $YBa_2Cu_3O_7$. It is tentatively identified as "superconducting fluctuations" (Cooperons) at $T \approx T_c$ and as an "acoustic plasmon" at 298 K. It is quite surprising that Cooper pair excitations can be seen at all in EELS since they are expected to provide only a weak peak in $\mathrm{Im}(\epsilon^{-1})$. Nevertheless, I feel that this feature, whether it is Cooperon or heavy plasmon or, more likely, something else not yet understood, is the best candidate so far for the electronic mode corresponding to the extra phonon in La_2CuO_4. Furthermore, if this new pole in $1/\epsilon$ is a real effect, it is hard to escape the idea that it is the extra boson needed to supplement conventional phonon coupling to give high T_c.

Finally, there are connections between the charge-fluctuation model proposed here and various other theories; I shall mention only those most familiar to me. $BaBiO_3$ is a case where charge fluctuations and breathing-mode phonon instabilities are known to be closely related. In the insulating state, breathing distortions are frozen,[22] and an associated charge fluctuation or disproportionation occurs. Rice and Sneddon[23] have discussed a bipolaronic interpretations of this transition, and generalizations to other dimensions have been discussed by Baeriswyl and Bishop.[24] Inelastic neutron scattering has measured phonon densities of states.[25,26] Dispersion curves[26] are known for most phonons in $BaPb_{0.75}Bi_{0.25}O_3$. In the energy region $\hbar\omega \sim 35-50$ meV, no sharp phonons are seen, but broad features reminiscent of "lattice defects of a very high defect density"[26] are seen, and the density of states contains significant weight and strong features in this region. It is possible that these complexities are related to the extra mode.

Both Rice and Wang[27] and Timusk and Tanner[28] have given models for coupled phonon and electronic excitations which can model features of the "mid-infrared" band. Enhancement of Raman strengths in $YBa_2Cu_3O_7$ by coupling Raman-active phonons to intra-planar charge fluctuations was modeled by Barišić, Kupčić, and Batistić,[29] while enhancement of infrared strengths in $YBa_2Cu_3O_7$ by coupling of infrared-active phonons to axial charge fluctuations was discussed by Batistić *et al.*[30] However, a low-lying narrow electronic mode does not immediately arise in any of these models.

In summary, the extra phonon mode seen in Refs. 2–6 seems likely to be related to an electronic collective excitation. However, it does not carry the signature of any familiar electronic mode. A simple classical model fits the data qualitatively, but the interpretation is mysterious.

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