Bond-stretching phonon anomalies and charge fluctuations in copper oxide superconductors

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On the basis of a semiphenomenological model, it is argued that the anomalous properties of the bond-stretching (BS) in-plane phonon modes in cuprate superconductors and some other metallic perovskite oxides are due to coupling to the mid-infrared structure in the electronic charge fluctuation spectrum. The energy scale of this charge mode is several times larger than the phonon frequency, i.e., within 0.2–0.6 eV, and its doping and momentum dependence is discussed in relation to experimentally observed behavior of the BS phonons. Experiments on the effect of oxygen isotope substitution on the phonon softening and linewidth are suggested as a source of important information on the charge excitations, and equations describing isotope effects are derived. The set of equations is overcomplete and can therefore serve as a consistency test for the model. We discuss how the midinfrared scenario could offer an explanation of anomalous BS phonon observed in different perovskites in a unified framework.

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I. INTRODUCTION

Anomalous softening of oxygen bond-stretching phonon modes is observed in many metallic perovskite oxides like bismuthates, manganites, nickelates,[1–3] and in a growing number of experiments on cuprates, both hole and electron doped.[4–8] Doping with charge carriers causes a strong renormalization of longitudinal high-frequency Cu-O bond-stretching (BS) phonon modes (typically around 70–90 meV) that reaches 20% and more compared to the insulating compound. At the same time, the transverse optical phonon branch is almost unaffected. The phonon linewidth also shows unusually large values, up to 10 meV, and significant concentration and momentum dependence.[6–8] The doping dependence of the BS phonon softening from zone center to zone boundary is much stronger in the (100) [and (010)] directions than in the (110) direction. In the latter case a cosinusoidal dispersion is observed.[5] Furthermore, the doping dependence of the phonon dispersion proceeds continuously from small to overdoped regime (e.g., in hole-doped compounds $\delta \sim 0.3$), almost saturating close to optimal doping ($\delta \sim 0.15$).[8] Experiments on a number of materials indicate the existence of a dispersion minimum developing on cooling from room temperature to low temperatures (also below T_c) at some intermediate wave vector, e.g., $q \sim 0.3$ reciprocal lattice units towards Brillouin zone boundary in the (100) direction.[1,5,6,9,10]

Although such phonon anomalies might not be directly related to superconductivity, understanding the anomalous behavior of the in-plane phonons can be important for clarifying their role in superconducting materials (for a review and related discussions see, e.g., Refs. 11–13). For instance, it has been claimed that the magnitude of softening shows a temperature dependence, which parallels that of superconductive order parameter,[7,14] or a noticeable increase in the phonon density of states of some modes upon the superconducting transition.[15] The phonon mode in cuprates that is usually discussed in relation to superconductivity is the buckling B_{1g} mode with $\omega \sim 40$ meV. The behavior of this latter differs from that of BS phonon since its coupling to electrons is peaked at low momenta and its dispersion is essentially independent on hole concentration.[16]

The present paper is mainly concerned with understanding the origin of the BS phonon anomalous behavior observed in different perovskites, trying to give a unified picture of the experimental findings. This mode is of a special interest due to its direct coupling to momentum-resolved charge fluctuations and it could provide important information on charge dynamics that otherwise is difficult to extract from experiments. There exist different theoretical proposals and experimental data (sometimes controversial) addressing the properties of the BS modes. In particular, one can classify these proposals according to the energy scale of the charge mode that couples to the phonon: vicinity to charge-density-wave instability or dynamical stripes,[2,9,17] when the energy is lower than that of phonon; different resonance scenarios, when the energies are close to the phonon energy;[8,18] and infrared scenarios, when the energy scale is, e.g., that of the optical plasmon with energy about 1 eV.[19] Initially, to describe the phonon, softening *ab initio* LDA models have been developed (see review).[20] However, one of the shortcomings of the LDA theory is the prediction of an order-of-magnitude smaller linewidths compared to experiments.[7] Another approach based on an electronic (vibronic) mechanism of softening was proposed in Ref. 21. It assumes the existence of overscreening effect in cuprates, i.e., negative dielectric function $\varepsilon(q,0) < 0$ for some nonzero momenta. More recently, phonon anomalies observed in various cuprates are explained as a direct manifestation of the existence of the low-lying collective charge excitations associated with stripes.[22] The domain-wall motion, or the charge collective oscillation, is strongly coupled with breathing or bond-stretching modes and produces a drop or a gap in the phonon dispersion at a wave vector twice that of the spin modulation

$2Q$. This wave vector varies systematically with doping and in LSCO seems to saturate close to optimal doping.[23] Nevertheless, although the phenomenon of stripes has been confirmed in a few perovskite materials, the anomalous softening of the BS phonon mode seems to be a more universal feature of these materials.[1] For instance, in the $La_{1.69}Sr_{0.31}NiO_4$ the phonon anomaly in (100) is observed while the stripe order is along the (110) direction.[2] Another possibility can be related to the vicinity of incommensurate charge-density wave formation or the existence of acoustic plasmons (see e.g., Ref. 24). The latter are expected to exist in layered structures.[25,26] That is, besides of gapped optical-plasmon mode, the spectrum should also contain plasmons with dispersion of the form $\sqrt{aq+bq^2}$, where a and b are some parameters. Finally, anomalous phonon softening has been discussed in the context of strongly correlated models. An important feature of these models is the suppression of charge response at small carrier densities and the lowering of energy of the collective charge response that becomes transparent from the sum rules.[27–29] In particular, it has been found that at low energies the charge response is enhanced for long wavelengths compared to short wavelengths.[11,30] Using the slave-boson approach within the t-J model, it has been shown[28,30] that the key property of the charge response is the existence of a low-energy peak that is close to the energy scale of the phonon. With hole doping the peak increases in energy and its crossing with the phonon dispersion results in a doping-dependent softening that agrees with that observed in experiments.[8] The analysis of the charge susceptibility of the t-J model along the symmetry directions of the Brillouin zone[30] also shows that its dispersion along the (110) direction is much larger than in the (100) direction. Thus the charge-response anisotropy explains why the full-breathing phonon mode does not show anomalous behavior despite being very close in energy to the half-breathing mode. Recently it has been found that this property of the charge susceptibility is not changed by the effect of the long-range Coulomb interaction.[31] It has also been shown that coupling to the charge response due to the spinon particle-hole continuum centered around 1 eV can account for BS phonon softening in general agreement with experiment. Finally, concerning infrared scenarios, the optical plasmon at 1–2 eV[19,32] has been proposed to model screening effect and the phonon shifts that are visible in optical reflectance experiments.[33] Properties of such plasmon have been successfully described within the t-J model with the inclusion of the long-range part of the Coulomb interaction.[34]

In the present paper an attempt is made to extract some characteristics of the charge mode from the observed behavior of the BS phonons on the basis of a semiphenomenological model describing their effective coupling. In particular, our analysis indicates that the main contribution to the observed phonon renormalization comes from the midinfrared part of the charge spectrum in the vicinity of 0.3–0.6 eV. It has been known for a long time from optical absorption, inelastic electron scattering, Raman-scattering experiments, etc. that the charge dynamics in the normal state of cuprates is anomalous and not consistent with the Drude-type relaxation.[35] One can approximately describe the spectrum below the charge-transfer gap as consisting of a Drude-type peak and a broad hump, a part of which evolves into a plasmon of energy ~1 eV or more in the overdoped regime.[36] At not very large doping the spectra show a build up of a broad structure in the midinfrared region that is observed in various perovskites. The main weight of this structure is located at energies several times that of the BS phonons, the highest in energy among the phonon branches. If the coupling to this midinfrared (MIR) charge mode is assumed and if some general properties of models with strong-electron correlations are taken into account, then the anomalies of the BS phonons can be explained for different perovskites in a unified framework. We also motivate the necessity of new experiments to probe some specific properties of the BS phonon spectrum following from our analysis. Specifically, we discuss the effect of oxygen substitution on the phonon spectrum and linewidth.

The organization of the paper is the following. In Sec. II we introduce the semiphenomenological model describing the coupling of the BS phonon to a damped charge mode. On the basis of experimental estimation of linewidth and doping dependence we identify the charge mode with the midinfrared structure observed in the in-plane optical conductivity experiments. In Sec. III we discuss the characteristics of the charge mode, doping and momentum dependence, in relation to recent experiments of BS phonon softening in various perovskites. In Sec. IV we discuss oxygen isotope substitution on the phonon softening and linewidth that could provide important information on the charge mode. Equations describing the isotope effect are derived and quantitative estimates are given. Finally, in Sec. V we draw our conclusion and argue how the midinfrared scenario could offer an explanation of BS phonon softening in different perovskites.

II. DETERMINATION OF CHARGE MODE COUPLED TO BS PHONON

We follow the idea that the anomalous behavior of BS phonons is related to coupling to charge fluctuations of strongly interacting electrons. The phonon Green functions $D(q,\omega)$ renormalized by interactions with electrons is obtained by the Dyson equation

$$D^{-1}(q,\omega) = D_0^{-1}(q,\omega) - \Pi(q,\omega),$$

where $D_0(q,\omega)$ is the bare-phonon Green's function. The phonon self-energy $\Pi(q,\omega)$ is given by the product of charge susceptibility $P(q,\omega)$ and the form factor of the BS phonons[37]

$$|g(q)|^2 = \sum_{\mu=x,y} \frac{2g^2}{MN\omega_0} \sin^2\left(\frac{q_\mu}{2}\right),$$

where we have neglected the dispersion of the bare phonon ω_0. Then the renormalized phonon Green function along the (100) direction takes the form

$$D(q,\omega) = \frac{\omega_0}{\omega^2 - \omega_0^2[1 + \alpha \sin^2(q_x/2)P(q,\omega)]}, \qquad (1)$$

where the coupling constant $\alpha = \frac{2g^2}{MN\omega_0^2}$ has been estimated in Refs. 27 and 38. In terms of the usual electron-phonon cou-

pling λ, it falls in the intermediate regime, $\lambda \sim 1$. If the momentum q is in the (110) direction, then the contribution of the structure factor is doubled. We assume that charge fluctuations important for the renormalization of the BS phonons can be described by a single oscillator, e.g., a damped charge-fluctuation mode with strength η_q and dispersion Ω_q,

$$P(q,\omega) = \frac{\eta_q}{\omega^2 - \Omega_q^2 + i\Gamma_q\omega}. \tag{2}$$

Such a form is commonly used in analyzing experiments on optical absorption, dielectric function, etc. and complies with the general criteria for the response functions, e.g., it obeys the Kramers-Kronig relations. The ansatz (2) can be considered as a natural starting point for the analysis of the charge response from the phonon data. However, despite its apparent simplicity, the existing data are not sufficient for a full determination of all its parameters. Therefore in the last section we describe a procedure, which allows one to derive additional relations between parameters and measurable quantities. These new constraints, not only permit to determine the parameters, but serve also to verify the consistency of the model. In particular, if necessary, one can include additional oscillators into the ansatz.

In the following, based on the observed behavior of the BS phonons, i.e., phonon dispersion ω_q and linewidth γ_q, we try to estimate the charge-mode parameters. After the renormalized phonon frequency is found, the phonon damping can be related directly to that of the charge mode Γ_q by the relation

$$\gamma_q = \Gamma_q \frac{\omega_0^2 - \omega_q^2}{\Omega_q^2 - \omega_q^2}, \tag{3}$$

where the phonon dispersion ω_q is found as the zero of the real part of the denominator in Eq. (1) and the phonon damping γ_q from the imaginary part by

$$\omega_q^2 - \omega_0^2[1 + \alpha \sin^2(q_x/2)\mathrm{Re}\, P(q,\omega_q)] = 0, \tag{4}$$

$$\gamma_q\omega_q = -\omega_0^2\alpha \sin^2\left(\frac{q^x}{2}\right)\mathrm{Im}\, P(q,\omega_q). \tag{5}$$

After defining the effective coupling parameter $\beta_q \equiv \alpha\eta_q$, and using Eq. (2) one easily finds the relation

$$\frac{\omega_0^2 - \omega_q^2}{\Omega_q^2 - \omega_q^2} = \frac{\beta_q \sin^2(q_x/2)\omega_0^2}{(\Omega_q^2 - \omega_q^2)^2 + (\Gamma_q\omega_q)^2}, \tag{6}$$

that gives Eq. (3) after substitution into Eq. (4). The semiphenomenological model introduces three parameters for the charge-excitation mode (dispersion Ω_q, damping Γ_q, and weight η_q or β_q). Knowledge of the phonon dispersion ω_q and linewidth γ_q from neutron or inelastic x-ray-scattering experiments allows one to reduce the number of undetermined parameters by two. This can be achieved by using, e.g., Eqs. (3) and (6). Thus, if one finds a third relation to experimentally measurable quantities of the phonon spectrum, it becomes possible to determine all the characteristics of the charge mode. We will return to this issue at the end of the paper. Equation (3) has the important property of connecting experimentally measurable dispersions and linewidths of the phonon to the charge mode, thus allowing one to locate the charge excitation responsible for phonon anomalies. For instance, assuming that the charge excitation is the optical plasmon with $\Omega \simeq (1-2)$ eV $\sim (12-24)\omega_0$, and knowing that the phonon linewidth γ_q reaches around 10% of the phonon's frequency, $0.1\omega_0$, while its softening is about 20%, we can estimate Γ_q

$$\Gamma_q \sim 0.1\omega_0 \frac{(12)^2}{0.4} \gtrsim 40\omega_0 \sim 3 \text{ eV}. \tag{7}$$

Such a linewidth is difficult to reconcile with experiments.[19,32] On the other side, if we assume a mode in the midinfrared with $\Omega \sim (0.5-0.6)$ eV $\simeq (6-7)\omega_0$, then

$$\Gamma_q \sim 0.1\omega_0 \frac{(6)^2}{0.4} = 9\omega_0 = 0.72 \text{ eV}. \tag{8}$$

As observed experimentally,[39] as a function of doping, the midinfrared absorption structure moves to lower energies[35] reaching the minimal energy $0.2-0.3$ eV $\simeq (3-4)\omega_0$ and for this range of energy the estimate of Γ_q is

$$\Gamma_q \sim 0.1\omega_0 \frac{(4)^2}{0.4} = 4.0\omega_0 \simeq 0.3 \text{ eV}. \tag{9}$$

The values of the charge-mode linewidths (8) and (9) are in agreement with typical experimental data (see Table I of Ref. 39 summarizing the results on $La_{2-x}Sr_xCuO_{4-\delta}$, $Bi_2Sr_2CaCu_2O_8$, $YBa_2Cu_3O_{6.9}$) and also with the fitting analysis of *a-b* plane optical-conductivity data in the Drude-Lorentz (or multicomponent) models.[15,40–43] A quantitative comparison of our estimates to these results suggests that the midinfrared structure can be responsible for the anomalies of the BS phonon modes. We note that the anomalous BS phonon softening and the charge excitation in the midinfrared part of the spectrum are both present in a broad class of perovskite materials, however, the possibility of their relation has not been discussed before. Our conclusion is also supported by experiments on other perovskites. For instance, existence of such midinfrared excitation at 0.4–0.6 eV has been predicted theoretically in Ref. 44, and experimentally observed in Refs. 45–49. Another argument in favor of MIR charge mode comes from the fact that if one considers the coupling of BS phonon to charge mode with an energy lower than that of the phonon, a hardening of the phonon dispersion occurs instead of a softening, as can be seen from Eqs. (2) and (4). The hardening is caused by a change in sign of the phonon renormalization when the charge mode crosses the phonon energy. Such behaviour has been, for example, discussed in the context of phonon-plasmon resonance in semiconductor superlattices.[51]

We would like to stress that the above estimates of the charge response have been done by testing the known components, i.e., checking if its energy and width are compatible with Eq. (3). Although it does not reduce to merely fitting the parameters, this analysis requires validation of the semiphenomenological model. Such a possibility will be discussed further in Sec. IV.

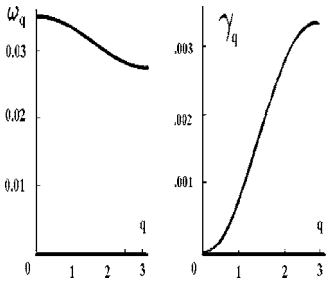

FIG. 1. Softening and linewidth of the bond-stretching phonon as given by the solution of Eq. (4) with $\omega_0=0.035$, $\Omega_q=c+v\sin(q/2)$, $c=0.15$, $v=0.01$, $\beta=0.01$, $\Gamma=0.17$.

III. ANALYSIS OF BS PHONON SOFTENING

From the above analysis we have concluded that the BS phonon anomalies can be related to the MIR charge mode, but a more detailed description is not possible without a specific dispersion for the charge mode or its coupling to phonons. Unfortunately, experimental data on the dispersion of the MIR structure are not available, therefore the following analysis is only qualitative and attempts to show some general consequences of the specific form of the charge-response function on the phonon softening. Actually, the following analysis shows that the observed behavior of the BS phonon sets strong constraints on the parameters of the charge-response model (2). Figure 1 shows the solution of Eqs. (4) and (5) for the choice of charge-mode dispersion,

$$\Omega_q = c + v\,\sin(q/2). \tag{10}$$

It describes a low-energy gapped positively dispersing excitation that saturates towards the Brillouin-zone boundary, roughly similar to the one found in the microscopic approach of Ref. 28. In our calculations all the energy quantities have been scaled with bandwidth ($W\simeq 2$ eV, so that the value $\omega_0\simeq 70$ meV becomes 0.035), the parameter c has been estimated above to be in the MIR. A rough estimate of the parameter $\beta_q\equiv\alpha\eta_q$ that appears in Eq. (6) at $q=\pi$ is obtained for the parameters of the charge excitation estimated above.

$$\beta_\pi = \frac{\omega_0^2-\omega_\pi^2}{\Omega_\pi^2-\omega_\pi^2}\frac{(\Omega_\pi^2-\omega_\pi^2)^2+(\Gamma_\pi\omega_\pi)^2}{\omega_0^2\sin^2(\pi/2)} \simeq 0.01-0.02.$$

From our calculations it follows that a flattened bottom of the phonon dispersion at q close to π can be obtained assuming a flattened dispersion of the charge mode at short wavelengths. This implies that the energy variation determined by the value of v must be comparable to phonon frequency at large enough q. Recent experiments on perovskites[1,6,7] show a minimum of phonon dispersion at intermediate $q(\sim 0.25-0.3)$ reciprocal lattice units rather than a flattened region. Thus to obtain such a minimum in our model, one needs to assume a stronger q dependence of Ω_q than in Eq. (10). It should be mentioned that different explanations of the minimum at intermediate q are found in literature, namely, based on charge inhomogeneities, stripes, etc.[7,9] Another physical mechanism for such a minimum can be related to strong electron correlations and their effect on a momentum-dependent coupling constant. In Figs. 1 and 2 the parameter β_q has been taken constant. However, in the strong-coupling calculations of Ref. 28 within the t-J model, where order $1/N$ corrections in the slave-boson method have been taken into account, it has been shown that the low-energy charge response is dominated by a polaron peak at low momentum. Similar arguments have been discussed in connection to electron-phonon vertex renormalization due to strong electron correlations.[11,13,52] It has been shown that vertex corrections lead to an enhanced forward scattering, so that the effective coupling constant is obtained by multiplying the structure factor of the BS phonon with the vertex factor. The resulting coupling constant is thus peaked not at the border of the Brillouin zone, but at intermediate momentum.[13,28,37] When we take into account forward scattering, or a β_q peaked at low momentum (see Fig. 2) one obtains a phonon dispersion having a minimum at intermediate momentum q in agreement with experiments. Another observation is that the effective coupling constant should not be very singular at low momentum, otherwise causing strong phonon softening already at small q, as seen in Fig. 2. For example, a behavior $1/q$ is not compatible with the observed phonon softening. From Eqs. (4) and (5) we also notice that the maximum in phonon damping occurs in the vicinity of q where the quickest drop in the phonon dispersion takes place. A similar correlation is observed in experiments (see Fig. 3 of Ref. 8). From Eq. (3) this behavior of the damping is obtained in the static approximation for the charge response

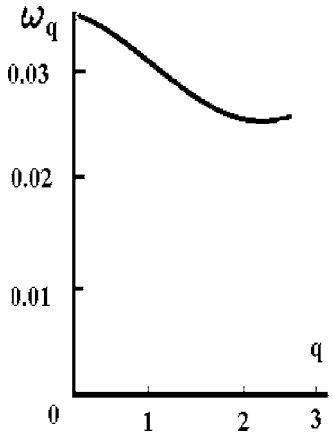

FIG. 2. Phonon dispersion assuming the coupling constant with enhanced forward-scattering correction: $\beta_q\sim 1/\sqrt{q}$ and the same parameters as in Fig. 1.

$$\omega_q \simeq \omega_0\left(1-\frac{\beta_q\sin^2(q_x/2)}{\Omega_q^2}\right)^{1/2},$$

$$\gamma_q \simeq \Gamma_q\omega_0^2\frac{\beta_q\sin^2(q_x/2)}{\Omega_q^4}. \tag{11}$$

Equation (11) shows that phonon softening is proportional to the inverse square of the charge-mode dispersion and its linewidth to Ω_q^{-4}. By assuming a smooth momentum dependence of the effective coupling constant β_q, it is possible to explain

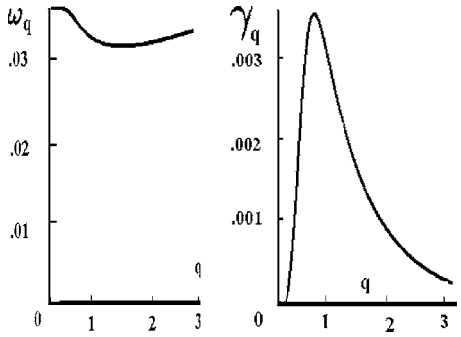

FIG. 3. Phonon dispersion and linewidth assuming interaction with acoustic plasmon: $\Omega_q=\sqrt{aq+bq^2}$, $a=0.006$, $b=0.005$, $\beta=0.01$, $\Gamma=0.1$.

the anisotropy in the phonon softening in terms of the charge dispersion. It follows from Eq. (11) that the dispersion of the charge excitation should be rather modest from the center to the border of the Brillouin zone in the (100) direction. In contrast, a steep dispersion in the (110) direction would explain why the full breathing mode is almost not softened along the diagonal. Finally, we have analyzed the effect of coupling to an acoustic plasmon with dispersion (see Fig. 3),

$$\Omega_q = \sqrt{aq + bq^2}, \tag{12}$$

where the parameters a,b have been chosen in such a way to reproduce a phonon softening and linewidth compatible with experiments. From the above analysis we conclude that the charge mode should belong to MIR and have a flattened dispersion along the (100) and (010) directions at short wavelengths.

It should be mentioned that our single-oscillator ansatz cannot be directly compared to a many-peak structure of the charge response in microscopic models, which involves a certain distribution of spectral weight, and a different position of the peak centers. For instance, the charge response obtained in Refs. 27 and 28 consists of two peaks. The small-weight narrow peak can be even lower than the BS phonon (70 meV) while the high-weight broad peak scales with the bandwidth and has an energy twice as large as MIR, e.g., more than 0.8 eV. A similar comparison holds for the models discussed in Refs. 24 and 34; where the high-energy feature is associated with acoustic plasmon. For a more quantitative analysis of the charge response following from the semiphenomenological model it is necessary not only to determine its parameters, but also to check the consistency of the model. This can be accomplished if additional experiments are carried out, as discussed in the next section. Our analysis is in qualitative agreement with the results in Ref. 37, where a slave-boson approach is employed to analyze the renormalization of the electron-phonon interaction within the t-J model. Here it is shown that the inclusion of vertex correction in phonon self-energy results in a formation of a dispersion minimum away from the boundary of the Brillouin zone along the (100), (010) directions as in Fig. 2.

IV. EFFECTS OF ISOTOPE SUBSTITUTION

Differently from other approaches, we argue that the phonon modes could serve as a probe of the low-energy momentum-resolved charge dynamics and some important information could be extracted on the basis of semiphenomenological models. In particular, we propose to estimate the effect of isotope substitution on the dispersion of the BS phonon to probe the characteristics of the charge mode coupled to it. The isotope shift of the breathing (110) mode does not depend on momentum. In contrast, the momentum dependence of the isotope effect on dispersion of the (100) and (010) BS modes is significant. This effect is shown in Fig. 4 where the values of the parameters have been chosen to have a stronger momentum dependence of softening. As shown in Fig. 5, the oxygen isotope substitution causes a stronger q dependence of the damping. These features can be inferred already from the static approximation for the charge response [Eq. (11)], and are enhanced due to dynamic cor-

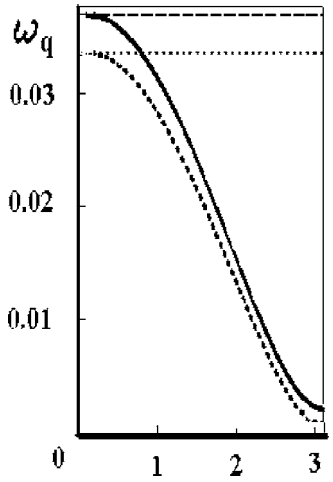

FIG. 4. Isotope O^{16}-O^{18} shift of the BS phonon dispersion in the (100) direction. As shown the effect is reduced towards the border of the Brillouin zone compared to the center. The short-dashed line corresponds to the heavier mass. The straight lines (dashed and dotted) represent the isotope effect on the breathing mode in the (110) direction (the dotted line corresponds to heavier isotope).

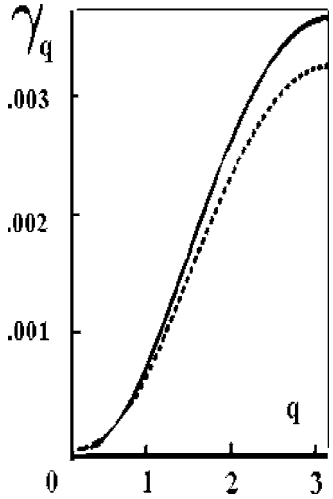

FIG. 5. The calculated isotope shift of the BS phonon linewidth in the (100) direction of the Brillouin zone. The short-dashed line corresponds to the heavier isotope.

rections. The latter are expected to increase with doping towards optimal due to the shift of the MIR charge-response structure to lower energies, as explained in Sec. II.

To describe the property discussed above one can define the oxygen isotope coefficient according to the equation

$$\frac{d\omega_q}{\omega_0} = -\bar{\alpha}_q \frac{dm}{m}, \tag{13}$$

where $dm=m(O^{18})-m(O^{16})$, $d\omega_q=\omega_q(O^{18})-\omega_q(O^{16})$, $\bar{\alpha}_q$ is a function of momentum in contrast to the standard definition of isotope coefficient $d\omega_0/\omega_0=-\alpha_0 dm/m$.[50,53] An estimate of $\bar{\alpha}_q$ is possible on the basis of Eqs. (4)–(6) by neglecting dynamical effects, i.e., considering ω_0/Ω_q as a small parameter. The renormalized phonon frequency is determined by the equation

$$\omega_q^2 = \omega_0^2\left(1+\beta_q \frac{\sin^2(q_x/2)(\omega_q^2-\Omega_q^2)}{(\omega_q^2-\Omega_q^2)^2+(\Gamma_q\omega_q)^2}\right). \tag{14}$$

By considering the derivative $d\omega_q/d\omega_0$ in Eq. (14) and keeping only the main term of the expansion, we obtain

$$\bar{\alpha}_q \simeq \alpha_0 \sqrt{1-\frac{\beta_q \sin^2(q_x/2)}{\Omega_q^2}}. \tag{15}$$

Such an expression, although obtained in a standard way, conveys the property of an anisotropic isotope coefficient that could be tested in experiments. It should be noted that in cuprates the effect of isotope substitution on phonon dispersion has not been studied systematically. Experiments on optical reflectance find that α_0 can be significantly different from 0.5.[33] Therefore α_0 should be identified as the value of $\bar{\alpha}_q$ for $q\to 0$. As noted earlier, $\bar{\alpha}_q$ can then be estimated from the measured values of phonon dispersion at $q=\pi$ $[\omega(q=\pi)/\omega_0 \simeq 0.8]$ as $\bar{\alpha}(q=\pi)\simeq 0.8\alpha_0$.

In a similar way, using relation (3) for the phonon linewidth, one can derive an equation of the isotope effect on γ_q,

$$\frac{m}{dm}\frac{d\gamma_q}{\gamma_q} = -2\alpha_0\left(1+2\frac{\bar{\alpha}_q}{\alpha_0}\frac{\omega_q\omega_0(\Omega_q^2-\omega_q^2-\Gamma_q^2)}{(\Omega_q^2-\omega_q^2)^2+(\Gamma_q\omega_q)^2}\right). \tag{16}$$

It is easy to see that the effect of isotope substitution on the phonon linewidth is roughly two times larger than on its dispersion,

$$\frac{d\gamma_q}{\gamma_q} \simeq -2\alpha_0\frac{dm}{m}. \tag{17}$$

Although these expressions have been obtained in a static approximation (i.e., we have assumed that the charge and phonon modes are not too close to resonance) the experimental test of Eqs. (15) and (17) could possibly decide on the validity of the considered model.

Nevertheless, these equations do not fully determine the charge mode. In fact, they do not lead to a third independent relation in addition to Eqs. (3) and (6), which is required to reconstruct the charge excitation from the properties of the BS phonon. This is obtained by the quantity:

$$\Delta\alpha_q \equiv \alpha_q - \alpha_0,$$

where we define $\alpha_q \equiv -(md\omega_q/\omega_q dm)$. It gives the difference between renormalized and bare isotope coefficients. Using Eq. (14) and keeping only the main term of the ω_0/Ω_q expansion we find

$$\Delta\alpha_q/\alpha_0 = \beta_q \sin^2(q_x/2)\omega_0^2\frac{(\Gamma_q/\Omega_q)^2-1}{\Omega_q^4}. \tag{18}$$

The differential-isotope coefficient on the left-hand side of Eq. (18) can be obtained experimentally (e.g., by inelastic neutron or x-ray scattering). From the above discussion it follows that its value should be close to zero for the (110) phonon mode and can be significant for the (100) and (010) modes. One can see that this coefficient is rather sensitive to the effective coupling constant β_q, the frequency of the charge excitation Ω_q and its ratio to the linewidth Γ_q. Depending on the latter, $\Delta\alpha_q/\alpha_0$ can even change its sign. In particular, one can see from the definition that $\Delta\alpha$ can be rewritten as

$$\Delta\alpha_q = -\frac{m\omega_0^2}{2\omega_q^2}\frac{d(\omega_q^2/\omega_0^2)}{dm},$$

and therefore the condition $\Gamma_q/\Omega_q > 1$ in Eq. (18) implies that the ratio ω_q^2/ω_0^2 decreases with mass, i.e., a smoother dispersion for the heavier isotope is expected. This is what suggest also the available data for $q\simeq 0$ (Refs. 39 and 54) according to which the inequality $\Gamma/\Omega > 1$ is observed for the midinfrared part of the spectrum in the relevant doping range.

We are not aware of experiments on isotope substitutions on the BS phonon. From the above arguments these experiments could give important information about the characteristic of the charge mode coupled to the phonon. In fact, one expects a different behavior on isotope substitution in the framework of a resonance scenario or incommensurate charge-density wave with a charge peak in the far IR picture.

V. CONCLUSIONS

In conclusion, we have presented an analysis of the bond-stretching phonon softening in different copper-oxide superconductors on the basis of a semiphenomenological model that couples the bond-stretching phonon modes to charge fluctuations parametrized by a single damped oscillator. Taking into account the experimentally observed dispersions and linewidth of the bond-stretching phonons, we have shown that the relevant charge mode is located in the mid-infrared–near-infrared region, 0.3–0.6 eV, i.e., above the phonon-energy scale (70–90 meV) and below the charge-transfer excitation gap, 2 eV, or optical plasmon energy, 1–2 eV. The anomalous behavior of the charge response in this part of the spectrum has been known for a long time, although the experimentally achievable resolution at such low energies still does not allow one to resolve its momentum dependence. In this context, the bond-stretching phonon that couples in a nonlocal way to charge fluctuations could be a source of important information. As observed experimentally, the cen-

ter of the midinfrared region shifts to lower energies with doping until overdoped region, quasisaturating at optimal values. This observation is in agreement with our study that requires a modest dispersion of charge response to reproduce the phonon-softening anomalies along (100) or (010) directions. At the same time, as it is known from the studies of microscopic strong-coupling models and in agreement with the present model, there exists a strong anisotropy of the charge response within the main directions of the Brillouin zone. In particular, it has been found that the charge-mode dispersion in the diagonal direction should be much stronger than in the (100) or (010) directions. This allows one to explain the observed anisotropy of the phonon softening. Finally, we have proposed that neutron and x-ray-scattering experiments on the effect of oxygen isotope substitution on the phonon mode could provide important information on the charge-excitation spectrum and lead to a better understanding of the origin of the phonon anomalies. In particular, the isotope coefficient defined above could be used to determine the characteristics of the charge mode. Equations (14)–(18) could also serve to verify the validity of the proposed semiphenomenological model as the number of constraints is larger than the number of parameters. We stress that the conclusion about the MIR location of the charge mode has been reached on the basis of the analysis of incomplete experimental data and that the experiments suggested in our analysis could indicate whether further refinements of the model are necessary. It is worth noting that the coupling of bond-stretching phonon to charge excitations in the midinfrared region could offer an explanation of phonon softening in different perovskites in a unified framework.

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