Bond stretching phonon softening and isotope effect in a phenomenological model for cuprate superconductors

S. Cojocaru\textsuperscript{1,2}, R. Citro\textsuperscript{3,4} and M. Marinaro\textsuperscript{3,5}

\textsuperscript{1} National Institute of Physics and Nuclear Engineering, Bucharest-Magurele 077125, Romania
\textsuperscript{2} Institute of Applied Physics, Chişinău 2028, R. Moldova
\textsuperscript{3} Dipartimento di Fisica “E. R. Caianiello” and C.N.I.S.M., Università degli Studi di Salerno, Baronissi (Sa)-84081, Italy
\textsuperscript{4} Laboratorio SuperMat, CNR-INFM, Salerno, Italy
\textsuperscript{5} I.I.A.S.S., Via G. Pellegrino 19, Vietri sul Mare (SA) 84019, Italy

E-mail: scojocaru@theory.nipne.ro

Abstract. Lattice and charge degrees of freedom of cuprate superconductors remain an intensively debated topic, both experimentally and theoretically, because of the strong anomalies observed in some of the phonon branches. In particular, the bond-stretching phonon modes show a knee like softening and a large damping for momenta around 0.25–0.3 r.l.u. along, e.g., (1, 0, 0) direction of the Brillouin zone. We discuss a phenomenological model where these anomalies are caused by coupling to electron charge fluctuations and explore a possibility to extract the spectrum of the latter from the phonon data. In particular, we predict a significant deviation from the standard value of the oxygen isotope substitution effect on the phonon spectrum itself. The phonon dispersion and linewidth are shown to contain complementary information related to the isotope effect which could allow a detailed description of the electronic spectrum.

1. Introduction

The half-breathing in-plane $Cu - O$ bond-stretching mode (BS) is ubiquitous for high-Tc cuprate superconductors [1] and corresponds to the highest frequency (of the order of 80 meV) phonon branch which is almost exclusively due to oxygen vibrations (see [2] for a review). Its coupling to charge carriers is therefore considered to be the main source of anomalies [3]: large momentum dependent softening (over 20\%) and line broadening (over 10 meV) which grow with doping even in the overdoped region. The decrease in the phonon frequency from $\Gamma = (0, 0, 0)$—point towards the border of the Brillouin zone is highly anisotropic an inhomogeneous, it has a maximum around $q = (0.25, 0, 0)$ r.l.u. and its symmetrical counterpart. A number of microscopic scenarios explaining this behavior have been proposed, e.g., related to quasistatic or dynamic stripes[4], charge dynamics of magnetic polarons in near resonance with the BS phonon [5], vibronic mechanism [6] and others. In [7] we have proposed a semi-phenomenological model which, instead of deriving the phonon properties, takes the experimental data the BS mode as an input for the reconstruction of the electronic density response function in the form of a damped oscillator. It then became clear that the existing data are insufficient for a complete description and one would need new kind of measurements to be carried out. Nevertheless, the model has...
allowed to estimate the possible location of the electronic excitation in the mid-infrared region, 0.3 − 0.5 eV, known as MIR anomaly in the optics of high-Tc cuprates [8]. A possible source of additional information on the coupling of lattice vibrations to charge carriers could be the effect of substitution of the oxygen isotope $O^{18}$ for $O^{16}$ on the BS phonon properties. In the present work we report the description of this effect which can be directly applied to the analysis of the suggested experiments. It turns out that in the case of the "MIR scenario" one should expect a rather large measurable isotope effect despite of energy separation between the phonon and electron excitations. It should be mentioned that the existing possibility of a site-selective isotope substitution requires a separate treatment because it is related to a modification of the phonon eigenvectors and electron-phonon coupling [9].

2. The isotope effect

The dynamic electron density response function

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

is defined by three parameters: linewidth $\Gamma_q$, frequency $\Omega_q$ and oscillator strength $\eta_q$. It describes that part of the response which is involved in the BS phonon self-energy:

$$ (2\omega_0) D^{-1}(q, \omega) = \omega^2 - \omega_0^2 (1 + \lambda \sin^2(qz) P(q, \omega)), $$

Where $D$ is the phonon Green’s function, $\omega_0$ the bare phonon frequency and $\lambda$ is the dimensionless coupling constant [5]. The dependence on oxygen mass is contained explicitly in the relation $\omega_0 \sim M^{-1/2}$. The r.h.s. as obtained from experimental data is determined by $\tilde{\gamma}_q, \tilde{\omega}_q$, the phonon linewidth and frequency respectively: $\omega^2 - \tilde{\omega}_q^2 + i\tilde{\gamma}_q \tilde{\omega}_q$. In our model the same quantities are found by solving for the real and imaginary parts of the zeros of (2), $\omega = \omega_q - i\gamma_q$.

That establishes two simple relations $\tilde{\omega}_q = \sqrt{\omega_q^2 - 4\gamma_q^2}$ and $\tilde{\gamma}_q \tilde{\omega}_q = 2\omega_q \gamma_q$ and results in two independent equations. After eliminating the factorized constants $\lambda \eta_q$ we find a single equation for the unknown parameters $\Gamma_q$ and $\Omega_q$:

$$ \Gamma_q^{-1} (\Omega_q^2 - (2\omega_q^2 - 2\gamma_q^2 - \omega_0^2)) = (2\gamma_q)^{-1} (\omega_q^2 - \omega_0^2 + 3\gamma_q^2). $$

The two equations mentioned above are then used to derive the expressions for the isotope coefficients (IC) quantifying the isotope effect (IE)

$$ \alpha_q = -\frac{d \ln \omega_q}{d \ln M}, \quad \beta_q = -\frac{d \ln \gamma_q}{d \ln M}. $$

We then get two additional but somehow lengthy expressions (see [10]) relating the IC to the parameters of the model. Thus, if both coefficients are known, it would allow not only to calculate the parameters of the model but also to verify its consistency. To focus on the effect of coupling it is convenient to introduce the IC describing the deviation from the reference values $\alpha_0 = 0.5$ and $\beta_0 = 1$, [11],

$$ a_q = \alpha_q - 0.5, \quad b_q = \beta_q - 1. $$

Indeed, it can be shown that these values are obtained from (4) when the two energy scales are far apart $\Omega_q \gg \omega_q$. Moreover, in many materials where the bulk of the isotope effect is due to disorder or lattice anharmonicity, the measured data are close to $\alpha_0$ and $\beta_0$. Calculation shows that when the main role is played by the electron-phonon coupling, some special conditions are necessary to have a significant deviation from $\alpha_0$ and $\beta_0$. This is naturally the case when the electronic excitation is close to resonance with the phonon mode. However, there is also another
possibility when the systems is far from resonance, but the coefficient \(b_q\) can still be large. In this respect it is important to point out a qualitative distinction between the two IC. It can be illustrated by the analytic expression derived in the assumption that the energy and linewidth of the electronic excitation are the largest parameters in the problem. We first scale all the energy quantities in the equations with the electronic frequency \(\Omega_q\):

\[
x_q = \omega_q \Omega_q^{-1}, \quad y_q = \gamma_q \Omega_q^{-1}, \quad g_q = \Gamma_q \Omega_q^{-1}.
\]

(6)

Keeping only the first non-vanishing terms, one obtains (the \(q\)–index has been dropped)

\[
a \simeq y \left(2x^2/g + g + 2g (y/x)^2 - y/x^2 - g^{-1}\right); \quad b \simeq 2yg (1 - g^{-2}) + 2x^2 (1 - g^2/2).
\]

(7)

It is now clear that while the dispersion derived IC \(a_q\) vanishes with phonon linewidth \(y_q\) (proportional to the electron-phonon coupling constant), the coefficient \(b_q\) derived from the phonon linewidth, IC remains finite. This is because \(b_q\) also depends on the energy separation between the two coupled excitations (the second term in the last line of (7)). In other words, for a vanishing phonon linewidth one should expect a saturation of the coefficient \(b_q\) at a finite value determined by the ratios like \(\omega_q/\Omega_q\) and \(\Gamma_q/\Omega_q\). Note also that the condition \(g \gtrsim 1\) determines the negative sign of \(b\) in (7). At the same time the IC derived from the phonon dispersion, \(a_q\), is close to its "standard" value.

To reveal the dependence of the electronic parameters of the model on the characteristics of the BS phonon mode we have solved the full set of equations discussed above to obtain \(a_q\) and \(b_q\) by plugging in the data obtained from inelastic neutron and X-ray scattering on \(La_{1.85}Sr_{0.15}CuO_4\) ([1]) for several momenta in the Brillouin zone (see Table). At the \(\Gamma\)–point the frequency of the BS phonon is \(\omega_0 = 85.5\) meV.

| \(q\) (r.l.u.) | \(\gamma_q\) (meV) | \(\omega_q\) (meV) | \(\Omega\) (meV) | \(a\) | \(b\) | \(g\) |
|----------------|-----------------|-----------------|-------------|-----|-----|-----|
| (0.25, 0, 0)   | 13              | 73              | 350         | 0.028 | -0.055 | 2.1 |
| (0.25, 0, 0)   | 13              | 73              | 250         | 0.02  | 0.024 | 1.47 |
| (0.25, 0, 0)   | 13              | 73              | 120         | -0.06 | 0.63  | 0.57 |
| (0.22, 0, 0)   | 9               | 78              | 350         | 0.022 | -0.13  | 2.35 |
| (0.22, 0, 0)   | 9               | 78              | 250         | 0.018 | -0.039 | 1.6  |
| (0.22, 0, 0)   | 9               | 78              | 120         | -0.027 | 0.8  | 0.56 |
| (0.2, 0, 0)    | 6.5             | 81              | 350         | 0.019 | -0.24  | 2.8  |
| (0.2, 0, 0)    | 6.5             | 81              | 250         | 0.017 | -0.15  | 1.9  |

The calculations have been repeated for a few different choices of \(\Omega_q\) to illustrate the scale and the main tendencies shown by the IC. In the figure \(x_q\) and \(y_q\) are considered as independent variables in a certain finite interval and the frequency \(\Omega_q\) is chosen in the MIR range, 300 meV. This interval is centered around the "physical" point in \((\omega_q, \gamma_q)\) taken from the data in the table to illustrate the tendency of \(|b_q|\) to a rapid growth with softening and broadening of the BS phonon mode. Note the relation between the sign of the coefficient \(b_q\) and the corresponding values of the ratio \(g_q\) in the table. Both the numerical analysis and the analytical result discussed above show that this coefficient is negative and significantly larger than \(a_q\) (which is positive) for the "MIR scenario", described in terms of an overdamped oscillator, \(\Gamma_q \gtrsim \Omega_q\). In the "resonance scenario" the signs of the two coefficients are reversed, the oscillator which produces the measured effect becomes coherent \(\Omega_q < \Gamma_q\), but again \(|a_q|\) is much smaller than \(|b_q|\). Interestingly, it is the mass dependence of the phonon dispersion which has been examined in most of the experiments on other materials, while the present analysis clearly shows that the linewidth offers a much more sensitive instrument. Nevertheless, both quantities contain independent and complementary information on IE, as already mentioned. We also find that the maximum of \(a_q\) is reached
at $q_x = 0.25$ r.l.u., the momentum corresponding to the largest softening and damping. In contrast, the IE on the phonon linewidth is larger on the portion corresponding to the largest slope. This is related to the arguments explained above that unlike $a_q$, which mainly depends on the magnitude of the linewidth which is in turn proportional to softening, $b_q$ depends strongly on the separation between the frequencies $\omega_q$ and $\Omega_q$. For the MIR scenario $b_q$ can be over 20% of the reference value $\beta_q = 1$. Since $b_q$ is negative this corresponds to suppression of IE ($\beta_q < 0.8$).

To conclude, we have shown that the IE on the anomalous BS phonon can be used to extract the momentum resolved dynamical electron density response function, which is expected to be strongly inhomogeneous and anisotropic in agreement with the strong momentum dependence of the BS phonon softening. Even in the case when the energies of the phonon and electron density excitation responsible for its softening are far apart, one can expect a large IE on the phonon linewidth, provided the electronic excitation is incoherent. This points out to the so-called mid-infrared anomaly of the optical spectra in high-Tc cuprates, suggesting an interesting possibility to uncover its finite wavelength behavior using the phonons as a probe.

**Note added by S.C. and R.C.** We report with our deepest regret the passing away of Prof. Maria Marinaro after the completion of this work.

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