Characteristics of oxygen isotope substitutions in the quasiparticle spectrum of Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$

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Abstract – There is an ongoing debate about the nature of the bosonic excitations responsible for the quasiparticle self-energy in high-temperature superconductors — are they phonons or spin fluctuations? We present a careful analysis of the bosonic excitations as revealed by the “kink” feature at 70 meV in angle-resolved photoemission data using Eliashberg theory for a $d$-wave superconductor. Starting from the assumption that nodal quasiparticles are not coupled to the $(\pi, \pi)$ magnetic resonance, the sharp structure at 70 meV can be assigned to phonons. We find that not only can we account for the shifts of the kink energy seen on oxygen isotope substitution but also get a quantitative estimate of the fraction of the area under the electron-boson spectral density which is due to phonons. We conclude that for optimally doped Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ phonons contribute $\sim 10\%$ and non-phononic excitations $\sim 90\%$.

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Experimental evidence based on the analysis of the optical properties of the high-$T_c$ cuprate superconductors suggested early on that the charge carriers were strongly coupled to a spectrum of bosonic excitations in the 40 to 60 meV range [1,2]. This was confirmed by the discovery of a slope change or kink in electronic dispersion of the quasiparticles (QP) by angle-resolved photoemission spectroscopy (ARPES) in the same energy range [3–5]. The boson coupling that manifests itself as peaks in the real part of the QP self-energy extracted from the ARPES dispersion curves can be seen in optical spectroscopy as features in the optical scattering rate, a term in the generalized Drude formula for the optical conductivity [1,2,6–11] and in tunneling [12,13]. Surprisingly, the nature of the bosons involved remains highly controversial [6–11,14–23]. The subject is of great interest since such a coupling, would give us a working model of superconductivity in these materials, at least within the Migdal-Eliashberg formalism with retarded interactions. In support of this approach, recent numerical solutions of the Mott-Hubbard model suggest that in the high-$T_c$ cuprates retarded interactions do provide most of the pairing [24,25], with the energy scale set by the size of the antiferromagnetic exchange constant $J$. These calculations do not support the suggestion by Anderson [26] that the energy scale for the retardation might be much higher and set by the Hubbard $U$ which would imply non-retarded pairing. Another mechanism considered in the literature is kinetic energy pairing for which the kinetic rather than the potential energy is lowered as the temperature is lowered below $T_c$. In an Eliashberg formalism this can be simulated by a phenomenological reduction in quasiparticle scattering which could be due to a reduction in the electron-boson spectral density [27,28]. But this, presumably, applies only to the superconducting state and would give no dispersion “kink” above $T_c$.

The controversy over the bosonic spectrum centers on the nature of the bosons: are they phonons or magnetic excitations. Neutron spectroscopy does not resolve this problem as it shows both phonon and magnetic excitations in the energy region of interest. What is needed is a fingerprint experiment, one that unambiguously identifies
the suspect bosons. In the past a number of candidate experiments have been proposed: doping and temperature evolution of the bosonic spectrum \[5,6,19,20\], substitution with magnetic and non-magnetic impurities \[21\], but the gold standard for the resolution of this problem is the oxygen isotope effect. A spectral feature caused by a lattice vibration involving oxygen will shift its frequency in a typical oxide by 6.5\% on \(^{16}\text{O}\rightarrow^{18}\text{O}\) substitution whereas a magnetic excitation will yield little or no change \[29\].

There have been many studies of the effect of oxygen isotope substitutions on the boson structure in the cuprates. While the early ARPES experiments could not resolve the expected 4.4 meV frequency shift in the kink, an optical study by Wang et al. \[30\] on the frequency of the shoulder in the reflectance of underdoped \(\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}\) found no evidence of an isotope effect. Recent scanning tunneling microscope (STM) work has shown a shift in the 52 meV structure of about the expected amount for an oxygen mode \[12\], but this may be explained in terms of inelastic tunneling through a barrier \[31–33\]. Very recently new high precision low-energy ARPES data of \(\text{Bi}_{2}\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}\) (Bi2212) with a \(T_c\) of 92 K have revealed a shift of the kink at 69 meV seen in the dispersion curves in the nodal direction for \(^{16}\text{O}\rightarrow^{18}\text{O}\) substitution with little change in the spectrum outside the immediate kink region. By providing a more detailed analysis of the spectra in ref. \[34\] we will show here that not only do these new measurements provide a fingerprint for the presence of a phonon contribution to the self energy of the charge carriers, but that they permit us to calculate the fraction of the bosonic coupling parameter \(\lambda\) associated with phonons and the fraction with magnetic excitations and conclude that the phonon contribution is \(\sim 10\%\) of the area under the spectral density. The major contribution to the glue is non phononic.

A key to an understanding of the bosonic structure in ARPES is the recognition that there are several features that can contribute to the kink in the dispersion curves and that the challenge is to separate their contributions. We will show that the new experiments of Iwasawa et al. \[34\] allow us to do that unambiguously. Calculations of scattering by spin fluctuations suggest that an important role is played by bosons that have a \((\pi,\pi)\) wave vector and they are particularly efficient in scattering charge carriers near the antinodes but have little effect on the nodal quasiparticles. Neutron scattering experiments show that the local (\(\text{q}\)-averaged) susceptibility has a strong peak in energy around 40 meV, but also that the \(\text{q}\)-dependent susceptibility is peaked at \((\pi,\pi)\) in this energy range. As a result, in frequency dependent measurements, the interaction with this magnetic peak is small in the nodal direction because Fermi surface to Fermi surface electronic transitions involving the spin one resonance with momentum transfer of \((\pi,\pi)\) cannot take place due to the particular Fermi surface geometry but this becomes more possible as we move towards the antinodal direction. This agrees with ARPES measurements that show the growth of a sharp feature as the momentum of the quasiparticles approaches the zone boundary at \((0,\pi)\) \[17\].

What then scatters the quasiparticles in the nodal direction? We suggest that spin excitations that are not centered at \((\pi,\pi)\) but spread out as a continuum and phonons that are also expected to be scattered more or less independent of QP vector \(\text{q}\). ARPES measurements \[23\] show that while there is a sharp spectral feature in the 65 meV region for nodal quasiparticles it is weak. Nevertheless, as we have argued above, it cannot be assigned to the \((\pi,\pi)\) spin resonance and we suggest this feature to be the residual phonon scattering superimposed on a broad magnetic background. In what follows we will show that this idea is fully consistent with all the data of Iwasawa et al. \[34\] and thus provides us with a quantitative tool to estimate the relative contribution of phonons and spin fluctuations to high temperature superconductivity.

The changes in the real and imaginary parts of the self energy with isotopic substitution are best illustrated with a simple model for the effect of a single Einstein mode at frequency \(\omega_E\) on the self energy. To simplify the algebra we first show the results for a mode of zero width. The electron-boson interaction spectral density \(I^2\chi(\omega) = A\delta(\omega - \omega_E)\) where \(A\) is the area under the Dirac \(\delta\)-function \(\delta(x)\). The resulting QP self-energy in the normal state at zero temperature \((T = 0)\) is

\[
\Sigma(\omega) = \Sigma_1(\omega) + i\Sigma_2(\omega) = A \ln \left| \frac{\omega_E - \omega}{\omega_E + \omega} - i\pi A \theta(|\omega| - \omega_E) \right|,
\]

where \(\theta(x)\) is a step function. The dotted and the dash-dotted curves in fig. 1(b) illustrate these functions where we have modified the spectral density by giving it a Lorentzian width. We note that under isotopic substitution \(^{16}\text{O}\rightarrow^{18}\text{O}\), the frequency of the mode is reduced to \(\omega_E \rightarrow \gamma\omega_E\), but the coupling constant too is reduced to \(A \rightarrow \gamma A = A_{\text{iso}}\) where \(\gamma = \sqrt{16/18} = 0.94\). The mass enhancement parameter \(\lambda = 2A/\omega_E\) remains unchanged. In the limit \(\omega \ll \omega_E\) the real part of the self energy is linear in \(\omega\) with slope \(\lambda\). As fig. 1 shows, isotopic substitution leaves two “fingerprints” in the self energy spectra, the well-known shift of the mode frequency by 6\% but a second, less known one, a reduction in amplitude of 6\%.

In the real part of the self-energy, fig. 1(a), the peak is shifted to lower energy and reduced in amplitude by the same factor while for the imaginary part, fig. 1(b), the onset of scattering is also moved to lower energies and its saturation value is reduced by the same \(\gamma\). The inflection points are found at \(\omega_L\) and \(\omega_{L,\text{iso}} = \gamma\omega_L\) respectively.

Many of the features of the model of a single bosonic mode shown in fig. 1 do not agree with recent data of Iwasawa et al. \[34\] even when we account for the changes in QP properties that are brought about by superconductivity. First, coupling, predominantly to a single mode peaked around \(\omega_{L,\text{iso}}\), whatever its origin, leads
Isotope substitution

Fig. 1: (Colour on-line) (a) Minus the real part of the QP self-energy, $-\Sigma_1(\omega)$ ($^{16}$O solid and $^{18}$O dashed lines) vs. energy $\omega$. The two thin dashed and dotted vertical lines indicate the position of the peak in the $I^2\chi(\omega)$ spectrum, $\omega_L$ and $\omega_{\text{iso}}$, respectively. (b) The same as (a) but now for $-\Sigma_2(\omega)$. Lorentzian model $I^2\chi(\omega)$ spectrum for $^{16}$O (dash-dotted line) and $I^2\chi_{\text{iso}}(\omega)$ for $^{18}$O (dotted line) right-hand scale.

to a saturation in the magnitude of the QP scattering rate, $\Sigma_2(\omega)$, at frequencies above $\omega_L$. Such a saturation is not seen in the published ARPES data obtained by fitting a Lorentzian form to momentum distribution curves, nor is it seen in optical data. Both methods find scattering rates [8,22] which continue to increase up to energies of order 400 meV. This is well beyond any phonon energy ($\sim 86$ meV in Bi2212 [35]) and shows that in addition to the sharp bosonic peak the charge carriers are also coupled to high-energy boson modes of some other origin. Secondly, the model predicts a reduction of the amplitude of the real part of the self-energy of 6% but the data of Iwasawa et al. shows little change in the range around 70 meV. While the frequency of the peak is shifted by the expected amount, the magnitude of the maximum in $-\Sigma_1(\omega)$ appears not to be significantly changed. This contradicts the single-phonon model of the self-energy.

We will next show that these contradictions can be resolved by assuming that in addition to a (weak) phonon contribution there is a strong contribution from magnetic scattering and that there is additional influence due to finite band widths. Taking this into account we are able to estimate the relative strength of the phonon and magnetic contributions to the QP self-energy.

To accomplish this we use the recent high resolution ARPES work of Zhang et al. [23]. These data were analyzed using maximum entropy inversion [9] in the superconducting state based on finite band width $d$-wave Eliashberg equations, in order to determine the spectral density $I^2\chi(\omega)$ of the bosons contributing to the nodal direction QP self-energy. In fig. 2(a) we show our fits, solid curve for $T = 17$ K (dashed curve for $T = 45$ K) to the ARPES data, open circles (open squares). Results for

Fig. 2: (Colour on-line) (a) Presents fits to the real part of the superconducting state QP self-energy ARPES data of ref. [23] as reported by Schachinger and Carbotte [9] for $T = 17$ K (solid line and open diamonds) and 45 K (dashed line and open squares). (b) The $I^2\chi(\omega)$ spectrum from maximum entropy inversion of the $T = 17$ K Zhang et al. [23] data (solid line) with $\lambda = 1.19$, and the area under the spectrum $A = 51.9$ meV. The (red) dashed line shows the spectrum $I^2\chi_{\text{iso}}(\omega)$ which was used to simulate the isotope effect (depicted also in the inset for clarity). Only the area of the peak equal to 6.15 meV or a partial $\lambda$ of 0.21 is shifted in energy by 6% with $\lambda = 1.19$ and $A_{\text{iso}} = 51.4$ meV.
the electron-boson spectral density $I^2\chi(\omega)$ at $T = 17$ K
and a bandwidth of 1.2 eV are shown in fig. 2(b) as the solid (black) line. This $I^2\chi(\omega)$ spectrum is very different
from the single sharp Lorentzian form used so far. It has a broad peak around $\omega_p = 65$ meV superimposed on a
large background extending to 400 meV. The cutoff in this spectrum depends critically on the choice of the
bare dispersion curve made in the ARPES study. The renormalizations end at the crossing between bare and
dressed dispersions and in the work of Zhang et al. [23] this was taken to be 400 meV. This choice is consistent
with optical data [7,8,10,11]. It is also consistent with the observation that the QP as well as optical scattering
rates still increase with increasing $\omega$ in the range of a few
100 meV.

Our model of the isotope substitution on the real part of the self energy is based on shifting only the peak in
$I^2\chi(\omega)$ shown by the (red) dashed curve in fig. 2(b) leaving the background unchanged as depicted in the inset, and results for the self-energy are shown in fig. 3(a). The solid curve is for $^{16}$O and the dashed curve for $^{18}$O. The results are in good overall agreement with the experimental data of Iwasawa et al. [34]. For instance, we predict a shift in the position of the peak of the order of 4.5% against the observation of $5 \pm 0.8\%$. We also predict a reduction in the amplitude of the self energy maximum of $\sim 3\%$ somewhat larger than is observed in the data. Note that the position of the peak in $-\Sigma_1(\omega)$ is not trivially correlated to the position of the peak in the spectrum, $\omega_p (\omega_{p,iso})$, nor to their values displaced by the gap amplitude, $\omega_p + \Delta (\omega_{p,iso} + \Delta_{iso})$.

Figure 3(b) presents the results of a numerical simulation using a smaller energy cutoff of 200 meV as done by Iwasawa et al. [34]. The solid and dashed lines are for $^{16}$O and $^{18}$O, respectively. The temperature is $T = 17$ K and the bandwidth had to be reduced to 0.6 eV. We also added experimental data for $^{18}$O (dash-dotted line) and for $^{16}$O (dotted line) from fig. 2(b) (cut zero) of Iwasawa et al. rescaled to meet our $\Sigma_1(\omega)$ data in amplitude. The qualitative agreement between data and numerical simulation is now even better. As a result of the new cutoff the peak position in our theoretical results moved down by $\sim 6$ meV. Also the difference in amplitude is now less than one meV and this explains why almost no difference in amplitude can be seen in the data. As a result of this simulation we can safely conclude that the data can be reproduced by a $I^2\chi(\omega)$ spectrum which has the same shape as the spectrum presented by the solid line in fig. 2(b) with the broad peak which will be shifted by isotope substitution now centered between 55 and 58 meV.

The area under the electron-boson spectral density which we have shifted in energy by $6\%$ corresponds to
6.13 meV or about 10% of the total area under the $I^2\chi(\omega)$. If we attribute this shift in area entirely to an electron-phonon coupling, the mass enhancement factor involved is $\lambda \approx 0.2$. This value is much smaller than estimated in ref. [34] but is of the order found in band structure
calculations on related cuprates [36–38]. It also compares favourably with the total value of mass enhancement $\lambda_{tot} = 0.23$ found in studies [39] of anisotropic electron-phonon coupling due to the oxygen buckling mode ($\sim 36$ meV). The coupling to the breathing mode ($\sim 70$ meV) was found to be much smaller: $\lambda_{tot} \approx 0.02$. Our value of $\lambda = 0.2$ corresponds to less than 20% of the total $\lambda$ found through maximum entropy inversion [9] of the nodal direction ARPES data of ref. [23].

In summary we analyzed recent nodal direction ARPES data of the effect of $^{16}$O $\rightarrow$ $^{18}$O substitutions on the QP self energy. We find that the data are consistent with a model where only $\sim 10\%$ of the self energy is through coupling to lattice vibrations, so that the major part of the glue function is of another origin, we believe spin fluctuations. We conclude that the new measurements need not imply that phonons play a large role in the superconductivity of these materials. If the mass enhancement
parameter $\lambda$ is used instead of the area under $I^2\chi(\omega)$ as a measure of the strength of the electronic renormalizations, the phonon contribution to $\lambda$ is 0.2 or less than 20% of the total $\lambda=1.19$ for our realistic model for the spectral density in Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ obtained from inversion of nodal direction ARPES data.

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