An electron–boson glue function derived from electronic Raman scattering

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Abstract
Raman scattering cross sections depend on photon polarization. In the cuprates, nodal and antinodal directions are weighted more strongly in B₂g and B₁g symmetries, respectively. On the other hand, in angle-resolved photoemission spectroscopy (ARPES), electronic properties are measured along well-defined directions in momentum space rather than their weighted averages being taken. In contrast, the optical conductivity involves a momentum average over the entire Brillouin zone. Newly measured Raman response data on high-quality Bi₂Sr₂CaCu₂O₈₊δ single crystals up to high energies have been inverted using a modified maximum entropy inversion technique to extract from B₁g and B₂g Raman data corresponding electron–boson spectral densities (glue), and these are compared to the results obtained with known ARPES and optical inversions. We find that the B₂g spectrum agrees qualitatively with nodal direction ARPES while the B₁g results look more like the optical spectrum. A large peak around 30–40 meV in B₁g and a much less prominent one in B₂g are taken as support for the importance of \((\pi, \pi)\) scattering at this frequency.

(Some figures in this article are in colour only in the electronic version)
This leaves 90% of the effective spectrum which extends to 400 meV, much larger than any phonon energy, to possibly come from spin fluctuations.

So far $I^2\chi(\omega)$ has been recovered from tunneling [1, 18], nodal direction ARPES [6-8, 10], and optical data in the cuprates [11, 12, 19, 20, 13, 14]. In principle, $I^2\chi(\omega)$ is anisotropic and it will be different for each momentum direction. Also quasi-particle and optical spectral densities will not be the same. While both involve the same bosons different weighting electronic factors apply. This is also true for Raman for which different vertices apply for different polarization. In this case, the frequency of the incident (scattered) light, $\nu$, will not be the same. While both involve the same bosons the magnitude of which is $\chi_{\mu}(\omega)$, can be extracted quantitatively from $I^2\chi(\omega)$.

The basis of the analysis is the Raman response function $R_{\mu}(\omega)$, where $\mu$ represents the scattering symmetry. Here we focus on $\mu = B_{1g}$ and $B_{2g}$. The response is derived from the measured cross section $\sigma_{\mu} = (\omega_{\mu}/\omega_0)[1 + n(\omega, T)]^{-1}d\sigma_{\mu}/d\Omega d\omega_0$ [26]. Here, $\omega_0$ is the frequency of the incident (scattered) light. $\Omega$ is the solid angle of acceptance of the collection optics and $n(\omega, T) = \text{exp}[\hbar\omega/(k_B T) - 1]^{-1}$ is the Bose factor. $\sigma_{\mu}$ is corrected for the sensitivity of the instrument. Since the optical constants vary only little for visible light, interface effects can be absorbed in the constant $R_{\mu}$, the magnitude of which is irrelevant here. For revealing the pure symmetry components of the response in a reliable fashion we measured spectra at different parts of the BZ dictated by symmetry $\mu$ [26]. In the normal state and within a Kubo formalism we can show that the Raman spectrum [24] and the optical response [28–32] is related to the appropriate electron–boson spectral density, $I^2_{\mu}(\nu)$, to a good approximation through the equation [33–35]

$$
\Gamma_{\mu}(T, \omega) - \tau^{-1}_{\mu, \text{imp}} = \int_{0}^{\infty} d\nu K(\omega, \nu; T) I^2_{\mu}(\nu),
$$

where $\tau^{-1}_{\mu, \text{imp}}$ is a Raman impurity scattering rate and

$$
K(\omega, \nu; T) = \frac{\pi}{\omega} \left[ 2\omega \coth \left( \frac{\nu}{2T} \right) - (\omega + \nu) \coth \left( \frac{\omega + \nu}{2T} \right) \right],
$$

where $T$ denotes the temperature. For the conductivity which involves an average over all momentum directions $\theta$ the appropriate electron–boson spectral density is $I^2_{\mu}(\nu)$, the average over the directions $\theta$. For the Raman case with symmetry $\mu$ there is an additional weighting of $\cos^2(2\theta)$ for $B_{1g}$ and $\sin^2(2\theta)$ for $B_{2g}$. Here, $I^2_{\mu}(\nu, \theta)$ is the electron–boson spectral density associated with momentum direction $\theta$. There is another difference between quasiparticle and transport quantities associated with vertex corrections. These are expected to mainly change the magnitude of the distribution functions with change changes secondary.

Jiang and Carbotte [24] give the formula for the lowest order Raman susceptibility for an interacting electron system in the form

$$
\chi_{\mu}(i\nu_n) = -T \sum_{q, \mathbf{v}} \text{Tr}[\gamma^{\mathbf{v}}_{\mu}(\mathbf{q}) \tau_3 G(\mathbf{q}, i\omega_m) \times \tau_3 \tilde{G}(\mathbf{q}, i\omega_m - i\nu_n)],
$$

with $\omega_m$ and $\nu_n$ the fermionic and Bosonic Matsubara frequencies, respectively, $\mathbf{q}$ the momentum vector, $\tau_3$ the third Pauli matrix and $G(\mathbf{q}, i\omega_m)$ the electronic matrix Green’s function in Nambu notation. Equation (4) is valid in the superconducting as well as normal state. Here we consider only the latter. Equation (4) differs from the well known formula for the optical conductivity [28–31, 24] only through the factor $\gamma^{\mathbf{v}}_{\mu}(\mathbf{q})$, [24], which is to be replaced by $e^2 v^{\mathbf{v}}_{F, \mu}(\mathbf{q})$ where $v^{\mathbf{v}}_{F, \mu}(\mathbf{q})$ is the x-component of the Fermi velocity at momentum $\mathbf{q}$ which in the free electron model is assumed constant and $v^{\mathbf{v}}_{F, \mu}(\mathbf{q}) = v^{\mathbf{v}}_{F}/2$ in two dimensions. For infinite bands with constant electronic density of states $N(0)$, equation (4) can be written in a more convenient form as

$$
\chi_{\mu}(i\nu_n \rightarrow \nu + i0^+) = N(0) \int d\epsilon \int_{0}^{2\pi} \frac{d\theta}{2\pi} \times \frac{[f(\epsilon) - f(\epsilon - \nu)]\gamma^{\mathbf{v}}_{\mu}(\theta)}{\nu + i\tau_{\mu, \text{imp}}^{-1} + \Sigma^*(\epsilon, \theta) - \Sigma(\epsilon + \nu, \theta)},
$$

This equation has the same form as equation (22) of Sharapov and Carbotte [35] reported in their study of the effect of the
energy dependence of the quasiparticle density of states on the far-infrared absorption in underdoped cuprates. Equation (5) differs as there is now an integration over angles $\theta$, the additional Raman vertex $\gamma'_\mu(\theta)$, and a different numerical factor. The same algebraic manipulation as in applied in (33) gives:

$$\Gamma_\mu(\omega) = \tau_{\mu, \text{imp}}^{-1} - \frac{1}{\omega} \int_{-\infty}^{\infty} d\epsilon \left[ f(\epsilon) - f(\epsilon + \omega) \right]$$

$$\times \int_0^{2\pi} \frac{d\theta}{2\pi} \gamma_\mu^2(\theta) \text{Im}\left[ \Sigma_\mu(\epsilon + \omega, \theta) - \Sigma_\mu^*(\epsilon, \theta) \right], \quad (6)$$

where $\Sigma_\mu(\epsilon, \theta)$ is the quasiparticle self-energy due to the directional electron–boson spectral density $I^2_\mu(\Omega, \theta)$ and the $\ast$ indicates the complex conjugate. This leads directly to our fundamental equation (2) when we use the relationship for the imaginary part of the quasiparticle self-energy in terms of $I^2_\mu(\Omega, \theta)$, namely

$$\text{Im}\left[ \Sigma_\mu(\omega, \theta) \right] = \frac{\pi}{2} \int_0^{\infty} d\Omega \frac{I^2_\mu(\Omega, \theta)}{\Omega} \left[ 2 \coth \left( \frac{\Omega}{2T} \right) - \tanh \left( \frac{\omega + \Omega}{2T} \right) + \tanh \left( \frac{\omega - \Omega}{2T} \right) \right]. \quad (7)$$

In the approximation of equation (5) for $\chi_\mu(\omega)$ we can construct the corresponding Raman cross section from

$$\text{Im}\left[ \chi_\mu(\omega) \right] = \chi''_\mu(\omega) = \frac{\omega \Gamma_\mu(\omega)}{\left[ \omega - \lambda_{\mu}(\omega) \right] + 2\Gamma_\mu^2(\omega)}, \quad (8)$$

where $\omega \lambda_{\mu}(\omega)$ is the Kramers–Kronig transform (KK) of $\Gamma_\mu(\omega)$ as described by Opel et al [25] (equation (A8)) to whom we refer for a detailed discussion of how the scattering rate $\Gamma_\mu(\omega)$ is extracted from the data on the Raman cross section corresponding to $B_{1g}$ and $B_{2g}$ symmetries. In figure 1 we present our results for the electron–boson spectral density $I^2_\mu(\omega)$, figure 1(a) for $B_{1g}$ (antinodal) and figure 1(b) $B_{2g}$ (nodal) polarizations. We show for comparison equivalent results obtained previously from optics [17, 36], figure 1(c), and from nodal direction ARPES, [10], figure 1(d). In figure 2(a) we show our maximum entropy fits to the $B_{2g}$ Raman scattering rates for two temperatures. The light solid (blue) dots are experiment at $T = 98$ K and the heavy solid (black) curve our fit. The light open (black) dots give the data for $T = 298$ K with the heavy dashed (red) curve our fit. Note that the low frequency part of the data at 98 K is almost linear, a feature which is well captured by the theoretical fit except for the very lowest $\omega$ region. On the whole the general trend is well described by our data reconstruction. To compensate for the rather big zero frequency offset of $\Gamma_{B_{2g}}(\omega \rightarrow 0, T)$ an impurity scattering rate of $\Gamma_{B_{2g, \text{imp}}} = 44$ meV had to be introduced in equation (2). In figure 2(b) we show a comparison between data and theoretical fit for the $B_{2g}$ Raman cross section (see [25] for notation) which is the measured quantity from which the Raman scattering rate $\Gamma_{B_{2g}}(T, \omega)$ of figure 2(a) has been extracted. To get this quantity we applied equation (8), with $\lambda_{B_{2g}}(T, \omega)$ obtained from a Kramers–Kronig transform of our fitted Raman scattering rate $\Gamma_{B_{2g}}(T, \omega)$. We see good agreement except for a low frequency peak in the theoretical curve at $T = 98$ K (heavy solid (black) curve) not present in the data. This peak is traced to our use of an infinite band approximation. The experimental data on $\Gamma_\mu(T, \omega)$ develop peaks around 700 meV (not shown here) for both $B_{1g}$ and $B_{2g}$ symmetry which is understood to be the result of a reduction in the electron density of states as a band edge [10] is approached. This effect is not captured in our calculations and translates into important differences between calculated and experimental $\lambda_\mu(T, \omega)$. Furthermore, we observe that theory deviates consistently from experiment to lower values for energies greater that $\sim 350$ meV. This can also be traced to our use of the infinite band width approximation. A more
Figure 2. (a) Fit to the B\textsubscript{2g} Raman data for the scattering rate $\Gamma(\omega, T)$ from our maximum entropy reconstruction. (b) The corresponding fits to the measured B\textsubscript{2g} cross section.

complete discussion of the effect of finite bands in the inverted electron–boson spectrum can be found in [10].

Similar results are found for the B\textsubscript{1g} data reconstruction. $\Gamma_{B1g}(\omega, T)$ is reproduced equally well except for some deviations at low energies seen in the $T = 302$ K data because of a linear frequency dependence in the experimental $\Gamma_{B1g}(\omega, T)$ data extending from $\sim 100$ meV down to $\omega = 0$ which cannot be reproduced by theory. Such a low energy linear dependence at room temperature has not been observed in the B\textsubscript{2g} data (light open (black) dots in figure 2(a)). The zero frequency offset of $\Gamma_{B1g}(\omega \rightarrow 0, T)$ was compensated by an impurity scattering rate $\tau_{B1g, imp} = 81.6 \text{ meV}$ about twice as much as was necessary for B\textsubscript{2g}. The data reconstruction of the B\textsubscript{1g} Raman cross section also reveals deviations from experiment at low and high frequencies which, again, can be understood to be the result of our infinite band approximation on which equation (2) is based.

We turn now to a comparison of antinodal with nodal Raman results of figures 1(a) and (b). The shapes of the distributions obtained are quite distinct. For B\textsubscript{1g} there is a large peak at $\sim 29$ meV in the $T = 98$ K spectrum followed by a dip and then a second peak around 300 meV. As the temperature is increased there is a clear evolution of the spectrum with the low energy peak decaying in amplitude, broadening and moving towards higher energies. The same trend, although less pronounced, is observed for the second peak. The valley between the peaks becomes progressively filled in but it still very much remains, even for $T = 302$ K although the effects are much less pronounced. This is also seen when the B\textsubscript{2g} (nodal) Raman spectrum is considered instead of B\textsubscript{1g} (antinodal). The shape of $I_{B2g}^\mu(\omega)$ for B\textsubscript{1g} as well as its change with increasing temperature agrees well with previous trends for the optical case. In figure 1(c) we show results at $T = 100$, 200 and 295 K in a sample of Bi\textsubscript{2}212 based on data by Tu \textit{et al} [36]. The prominent peak is at a position ($\sim 44$ meV) slightly different from our Raman spectrum, but the overall shape at 100 K is in good qualitative agreement with the Raman result (figure 1(a)) including the second peak, the valley between the two peaks, and the temperature evolution. A more extensive set of data on a similar sample but doped with some yttrium is found in [12]. The data presented in their figure 2, top frame, is also in good qualitative agreement with our Raman results. The important observation is that both antinodal Raman and optical data show a prominent peak in the electron–boson spectral density around 30–40 meV which does not appear in ARPES and appears much less prominently in nodal Raman data. More specifically, the fractional area under this peak is only $\sim 3\%$ in B\textsubscript{2g} as compared with $\sim 23\%$ in B\textsubscript{1g}. As noted above, these two structures evolve with temperature in the same way which indicates their common origin. Part of this temperature evolution could be due to the reduced resolution intrinsic to our unbiased inversion method as the temperature is increased. (See [17].) This is consistent with the results obtained with the biased inversion method discussed later in figure 3 which shows less temperature dependence.

For nodal ARPES there is no peak at $\sim 30$ meV but one appears instead at much higher energy $\sim 65$ meV. This higher energy has often been identified with coupling to oxygen phonons [4–6, 15, 16] and we will return to this issue later. Such a peak is not seen in our Raman spectra which indicates that such effects are mainly confined to the nodal direction and do not appear in averaged quantities such as B\textsubscript{1g} and even B\textsubscript{2g} Raman even though for this latter polarization the Raman vertex peaks in the nodal direction. The fact that the peak at $\sim 30$ meV is stronger in B\textsubscript{1g} (antinodal) than in B\textsubscript{2g} (nodal) and is not seen in nodal ARPES is consistent with a boson which is associated with scattering through momentum transfer $q$ of $(\pi, \pi)$. Such a vector corresponds to transitions between those parts on the Fermi surface which lie also on the antiferromagnetic Brillouin zone and is, therefore, closer to the antinodal direction. Consequently, quantities that emphasize the area around $(\pi, 0)$ such as B\textsubscript{1g} Raman and, to some extent,
optics should show a strong peak. In contrast, the peak at \( \sim 30 \text{ meV} \) is expected to be weaker in B\(_{2g}\) symmetry weighing out the nodal part. This is what we observe and have shown in the data of figures 1(a) and (b).

Instead of using a maximum entropy technique to invert equation (2) van Heumen et al [14] used a histogram to characterize the electron–boson spectral density, \( I^2 \omega \chi(\omega) \), derived from optics. The histogram is then used to directly reconstruct the experimental data by inversion of equation (5) modified for the optical conductivity. They find less temperature dependence than we have here, for the position and width of the peak around 30 meV in B\(_{1g}\). So far we used an unbiased maximum entropy inversion of equation (2) in which the default model [17] is set to a constant at all temperatures. Another method is the so-called biased maximum entropy inversion in which the default model is set to the previous next lower temperature solution. We can expect the solutions of these two methods to be different because the inversion of equation (2) is an ill-posed problem which usually has more than one solution. It was pointed out by Yang et al [19] that in the case of HgBa\(_2\)CuO\(_4\)+\( \delta\) the biased inversion of optical data resulted in electron–boson spectra very similar to those reported by van Heumen et al [14] for the same material.

Results of such a biased inversion of the Raman B\(_{1g}\) and B\(_{2g}\) data are shown in figures 3(a) and (b), respectively. We see that the peak at \( \sim 30 \text{ meV} \) for the B\(_{1g}\) spectra shifts less with temperature than is shown in figure 1(a) although it still loses amplitude with increasing temperature \( T \). This points to the possibility that at least part of this peak comes from a phonon, and its contribution could be determined from the amplitude of this peak at room temperature. The situation is quite different for the B\(_{2g}\) polarization. Comparison of the spectra shown in figure 3(b) with the ones presented in figure 1(b) reveals that there is very little difference between the two sets of spectra. In particular, the position of the low energy peak \( \sim 40 \text{ meV} \) at 98 K shows almost the same temperature dependence and is almost smeared out at room temperature. Furthermore, the inversion method has little or no influence on the shape and size of the high energy part \( (\omega > 100 \text{ meV}) \) of the B\(_{1g}\) and B\(_{2g}\) spectra. Figures 3(c) and (d) show results for the often used mass enhancement parameter \( \lambda_{\mu} = 2 \int_{0}^{\infty} d\omega I^2 \omega \chi(\omega)/\omega \) as a function of temperature for B\(_{1g}\) and B\(_{2g}\) polarization, respectively. There is very little difference between the two methods of inversion.

Returning to figure 1(b), the nodal direction Raman spectrum (B\(_{2g}\)) has a very different characteristic shape as compared to B\(_{1g}\), figure 1(a). While it can be characterized as also having two peaks, the valley between them is not pronounced and its spectral weight is much more uniformly distributed below 300 meV with a relatively sharp drop off beyond this energy. This shape is much closer to what has been found in inversions of the nodal direction ARPES data reproduced in figure 1(d). This is expected since the B\(_{2g}\) Raman vertex peaks at the nodal direction. Note that ARPES is strictly directional and samples only the nodal direction while B\(_{2g}\) Raman probes an extended part of the BZ weighted by \( \sin^2(2\theta) \). Nevertheless, the agreement as to shape between B\(_{2g}\) Raman and nodal direction ARPES gives one confidence that both methods are measuring the same boson spectrum. Note that there is nothing which limits the application of this method to the high-\( T_c \) cuprates and our inversion technique has a more general applicability to other metals.

While our inversions provide us with a good handle on the size and qualitative shape of the spectral density, the question as to the origin of the boson involved is more difficult to answer in a definite way. Certainly if phonons are involved we would...
expect $I^2 \chi(\omega)$ to mirror the phonon frequency distribution while for spin fluctuations we should see an image of the local spin susceptibility. The fact that $I^2 \chi(\omega)$ shows very significant spectral weight up to 400 meV means that excitations other than phonons having energies below 100 meV are involved. For spin fluctuations the energy scale is set by the exchange coupling, $J$, which enters, e.g., the $t-J$ model as a parameter, and this is consistent with the large energy scale seen here. It is also consistent with recent numerical studies of the $t-J$ model by Maier et al. [37] in which an effective electron–boson spectral density associated with short range spin fluctuations is extracted and identified as the pairing glue. It displays many of the features seen in our empirical spectra. The cellular dynamical mean-field calculations of Kyung et al. [38] based on the Hubbard model also give qualitatively similar results for the spectral density and offer further microscopic support for an interpretation of our derived spectra as due largely to spin fluctuations.

On the other hand one does expect, and experimental data provide support for some contribution from the electron–phonon interaction [3, 5–7]. While the change in critical temperature $T_c$ on substitution of $^{16}$O $\rightarrow$ $^{18}$O is small for optimally doped samples, it is nonzero and it can be large for the underdoped case. However, the latter fact can also be understood as due to an energy dependence in the electronic density of states [39] or to a pseudogap formation [40] while at the same time the underlying contribution of phonons to the pairing interaction remains small. Recent ARPS experiments along the nodal direction [16] found a shift in the ‘boson kink’ in the renormalized dispersion curves of Bi2212 upon oxygen isotope substitution. This was assigned by Schachinger kink’ in the renormalized dispersion curves of Bi2212 upon oxygen isotope substitution. This was assigned by Schachinger [14] van Heumen E, Muhlethaler E, Kuzmenko A B, Eisaki H, Xu Z, Gu G, Ono S, Ando Y and Yazdani A 2008 Science 320 196.

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