Electron-phonon effects on the Raman spectrum in MgB$_2$

E. Cappelluti

Istituto dei Sistemi Complessi”, CNR-INFM, v. dei Taurini 19, 00185 Roma, Italy,
and Dipart. di Fisica, Università di Roma “La Sapienza”, P.le A. Moro, 2, 00185 Roma, Italy

(Dated: May 10, 2018)

The anomalous features of the Raman spectroscopy measurement in MgB$_2$ represent a still unresolved problem. In particular highly debated are the origin of the huge $E_{2g}$ phonon linewidth, the nature of the low energy ($\omega < \omega_{E_{2g}}$) background and the evolution of the Raman spectra with Al doping. In this paper we compute the self-energy of the $E_{2g}$ phonon mode in a fully self-consistent way taking into account electron-phonon effects on the electronic properties. We show that all the anomalous features can be naturally understood in a framework where the whole electron-phonon spectrum $\alpha \omega F(\omega)$ gives rise to significant damping processes for the electronic excitations and consequently for the $E_{2g}$ phonon itself. The two-peak structure as function of the Al doping is ascribed to finite bandwidth effects arising as the Fermi level approaches the $\sigma$ band edge.

PACS numbers: 74.70.Ad, 74.25.Kc, 63.20.Kr, 78.30.-j

There is nowadays a general consensus that the electron-phonon (el-ph) interaction is responsible for the superconductivity in MgB$_2$ 

There is nowadays a general consensus that the electron-phonon (el-ph) interaction is responsible for the superconductivity in MgB$_2$. Interestingly, the two-gaps scenario, spanning with Al doping a wide range of hole doping of the compound, still represents a highly puzzling anomaly. The main open questions on this compound still represent a highly puzzling anomaly.

The origin of the broad phonon linewidth in Raman spectroscopy has been widely discussed in literature. Common feeling on this subject is that the linewidth of a phonon mode $\gamma_\mathbf{q}$ reflects the strength of the electron-phonon coupling $\lambda_\mathbf{q}$ of this specific mode, in the spirit of the Allen’s formula $\gamma_\mathbf{q} = 2\pi N(0)\lambda_\mathbf{q} \omega_\mathbf{q}$, where $N(0)$ is the electron density of states (DOS) coupled with the phonon mode and $\omega_\mathbf{q}$ is the phonon frequency. First-principle calculations however indicate that the electron-phonon coupling alone of the $E_{2g}$ phonon mode is not sufficient to explain the large experimental Raman linewidth. In addition, the direct employment of the Allen’s formula is strongly questioned in the $\mathbf{q} = 0$ case, relevant for the Raman spectroscopy, where a more careful analysis shows that the damping of a $\mathbf{q} = 0$ phonon mode scattering with a non-interacting electronic system is strictly zero.

In contrast to the above scenario, a Quantum Field Theory analysis specifically addressed to the $\mathbf{q} = 0$ case has been developed in Refs. Although these studies were mainly aimed to investigate phonon anomalies in the superconducting state, this approach can be as well employed in the normal state. Along this line, for instance, Marsiglio et al. remarked that the imaginary part of a $\mathbf{q} = 0$ phonon self-energy in the weak-coupling limit is simply

$$\Pi''(\mathbf{q} = 0, \omega) \propto \omega_\mathbf{q} \lambda_\mathbf{q} = \frac{4\omega \Gamma_{\text{imp}}}{\omega^2 + 4\Gamma_{\text{imp}}^2},$$ (1)

where the impurity scattering rate $\Gamma_{\text{imp}}$ was assumed to be the only damping source of the electronic propagator: $G(\mathbf{k}, \omega) = 1/[\omega - \epsilon_\mathbf{k} + i\Gamma_{\text{imp}}]$.

Eq. (1) explicitly shows that the damping processes of a $\mathbf{q} = 0$ phonon are triggered by corresponding damping processes of the electronic charge response which screens the phonon, whereas the el-ph coupling $\lambda_{\mathbf{q}=0}$ of this particular mode rules the magnitude of these effects. This results holds true also when the main source of the
the electronic excitation damping is the electron-phonon interaction itself. In this case it is also important to note that in principle all the phonon modes, and not only the specific $\lambda_{q=0}$ mode, contribute to the electronic damping. Indeed, as we are going to discuss, the imaginary part $\Pi''(\omega)$ of the phonon self-energy is roughly related to the integral up to the energy $\omega$ of the imaginary part of the electronic self-energy, $\gamma(q = 0, \omega) \propto \lambda_{q=0} \int_0^\omega d\omega' \Gamma(\omega')$. For optical phonons close to the top of the phonon spectrum we have $\int_0^\omega d\omega' \Gamma(\omega') \propto \omega' \lambda$, $\lambda$ being the total electron-phonon coupling constant, so that in intermediate-strongly coupled systems the phonon linewidth can be of the same order of the phonon frequency itself. In conventional low-$T_c$ superconductors however the total electron-phonon coupling $\lambda \sim 1$ is spread over several phonon branches and $q$ modes, $\lambda = \sum_{q,\nu} \lambda_{q,\nu}$, so that the contribution from a single mode is quite small. Typical values of $\lambda_{q=0,\nu}$ for the $A_g$ phonon modes in cuprates are for instance $\lambda_{q=0,\nu} \leq 0.034 \text{meV}$, resulting in experimental Raman linewidths of few meV. The opposite extreme case of only one strong coupled $q = 0$ phonon mode $\omega_\lambda$ carrying the whole electron-phonon coupling would also predict extremely small phonon linewidth since, although $\lambda_{q=0} \sim 1$, the amount of electron-phonon coupling smaller than $\omega_\lambda$ is practically negligible and $\int_0^{\omega_\lambda} d\omega \Gamma(\omega) \approx 0$.

Along this scenario MgB$_2$ presents very peculiar characteristics since it presents at the same time two (degenerate) strong coupled $q = 0$ $E_{2g}$ phonon modes and a relevant fraction of the total electron-phonon coupling strength spread over other different modes. We employ a fully self-consistent many-body approach to investigate the normal state $E_{2g}$ Raman spectrum of MgB$_2$. In more explicit way we first solve iteratively the Marsiglio-Schosmann-Carbotte equations with the Eliashberg spectral function $\alpha_\sigma^2 F(\omega)$ obtained by first-principle calculations (Fig. 4) to evaluate the real-axis electronic self-energy $\Sigma(\omega)$ of the $\sigma$ bands. The Eliashberg function $\alpha_\sigma^2 F(\omega) = \alpha_{\sigma,\nu}^2 F(\omega) + \alpha_{\sigma,\nu}^2 F(\omega)$ describes the total electron-phonon scattering of the $\sigma$ electrons with both the $\sigma$ ad $\pi$ bands. Note that the $\alpha_\sigma^2 F(\omega)$ extracted from first-principle techniques represents the el-ph spectral function where phonons are already renormalized. In order to better compare with the experiments, we include also explicitly possible effects of impurity disorder. We schematize the $\sigma$-bands as two degenerate bands with constant DOS, $\sum_k G(k, \omega) = N_\sigma(0) \int d\epsilon G(\epsilon, \omega)$, where $N_\sigma(0)$ represents the $\sigma$-band electron density of states per spin and per band, $N_\sigma(0) \approx 0.075$ states/(eV $\cdot$ cell). The so-obtained electronic Green’s function is then employed as input to calculate the full frequency dependence of the $q = 0$ phonon self-energy $\Pi(\omega)$, whose imaginary part reads:

$$\Pi''(\omega) = \frac{\pi N_s N_b}{N_c} \sum_k |g_{k, E_{2g}}|^2 \int d\omega' A(k, \omega' + \omega) \times A(k, \omega') \left[ f(\omega' + \omega) - f(\omega') \right],$$

where $N_c$ is the number of sampling point in the Brillouin zone, $A(k, \omega) = (1/\pi) \text{Im}[1/(\omega - c_k - \Sigma(\omega))|$ is the electronic spectral function and $N_s = 2$ and $N_b = 2$ represent respectively the spin degeneracy and the $\sigma$-band degeneracy. Note that Eq. 2 does not account the double degeneracy of the two $E_{2g}$ modes which on the other hand contribute to the superconducting pairing. A similar expression is obtained for the real part of the phonon self-energy, also attainable from the Kramers-Krönig relations. The electron-phonon matrix elements $g_{k, E_{2g}}$ can be estimated from the $E_{2g}$ deformation potential $D_{E_{2g}} \simeq 12$ eV/A [11, 50, 31], which is in good approximation $k$-independent close to the Fermi level, and from evaluating the zero point motion lattice displacement $g_{E_{2g}} = D_{E_{2g}} \sqrt{\langle q^2 \rangle}$ = 0.39 eV, taking into account anharmonic effects [22].

We finally evaluate the phonon spectral function $B(\omega)$ from the imaginary part of the phonon propagator

$$B(\omega) = -\frac{1}{\pi} \text{Im} \left[ \frac{2 \Omega_{E_{2g}}}{\Omega_{E_{2g}}^2 - \omega^2 + 2\Omega_{E_{2g}} \Pi(\omega)} \right],$$

where $\Omega_{E_{2g}}$ is the unrenormalized phonon frequency, while the renormalized phonon frequency $\omega_{E_{2g}}$ and the phonon linewidth $\gamma_{E_{2g}}$ are simply related to the real and imaginary parts of $\Pi(\omega)$,

$$\omega_{E_{2g}}^2 = \Omega_{E_{2g}}^2 + 2\Omega_{E_{2g}} \Pi'(\omega_{E_{2g}}),$$

$$\gamma_{E_{2g}} = -2(\Omega_{E_{2g}}/\omega_{E_{2g}}) \Pi''(\omega_{E_{2g}}).$$

We assume the unrenormalized $E_{2g}$ phonon frequency $\Omega_{E_{2g}} = 100$ meV, close to the top of the phonon spectrum.

In Fig. 11 we plot the imaginary part of the phonon self-energy, and the fully renormalized phonon spectrum. The filled symbols mark the values of imaginary parts of the phonon self-energy evaluated at the renormalized phonon frequency $\omega_{E_{2g}}$, obtained from the self-consistent solution of Eq. 4. At low temperature and in the absence of impurity scattering, the imaginary part of the phonon self-energy shows a monotonic behavior which reflects the corresponding increasing of the electronic damping processes (Fig. 11). Already in this case we predict a relevant value of $\Pi''(\omega_{E_{2g}})$ and of the phonon linewidth $\gamma_{E_{2g}} \simeq 6$ meV, which is essentially only due to the spectral weight of $\alpha_\sigma^2 F(\omega)$ for $\omega \leq \omega_{E_{2g}}$. Things are even more drastic when finite temperature effects or impurity scattering are taken into account. As is well known the imaginary part of the electronic self-energy starts from a finite value at $\omega = 0$ which is reflected in a
sudden increase of $|\Pi'(\omega)|$ and in a significant broadening of the phonon spectrum. Note also the appearance of an incoherent background for $\omega \lesssim \omega_{E_{2g}}$ and of a broad shoulder at very low temperature, in striking agreement with the Raman experimental data. Similar effects were pointed out in Refs. [33, 34] in the context of the electronic Raman scattering. We would like to stress that the sharp increase of $|\Pi'(\omega)|$ and the onset of the low temperature shoulder in the phonon spectrum are intrinsic features of the charge response function. As a matter of fact, as noted in Ref. [25], at a first approximation the ratio $-\Pi'(\omega)/\omega$ is qualitatively similar to the real part of the optical conductivity $\sigma'(\omega)$. The low energy sudden enhancement of $|\Pi'(\omega)|$ and the corresponding shoulder in the phonon spectrum, which arise from the finite value of $\Sigma'(\omega = 0)$, have thus a strict connection with the appearance of a Drude-like peak in $\sigma'(\omega)$. Along this line, the experimental observation of a significant Drude-like scattering rate $1/\tau \simeq 9 - 37$ meV [35, 36] and of the Raman shoulder at low temperatures $T \sim 40 - 45$ K [18] points out the actual presence of a small amount of impurity scattering. From the comparison between our results and experimental data we estimate an impurity scattering rate $\Gamma_{\text{imp}} \simeq 5.5$ meV.

As a last point of our analysis we address the evolution of the Raman spectra as function of the Al doping. Ab-initio calculations predict that the electron doping induced by the Al content would fill the two $E_{2g}$ bands, leading to a reduction of the electron-phonon coupling. This is expected to result in a steady hardening of the $E_{2g}$ phonon frequency [31]. Actual Raman measurements are however in substantially disagreement with this picture.

FIG. 2: (a) Phonon spectral function for $T = 50, 100, 150, 200, 250, 300$ K; (b) temperature dependence of the renormalized phonon frequency $\omega_{E_{2g}}$; (c) temperature dependence of the phonon linewidth $\gamma_{E_{2g}}$. We considered here a small amount of impurity concentration corresponding to $\Gamma_{\text{imp}} = 5.5$ meV.
behavior can be understood by looking at the real part
accompanied by a narrowing of the phonon linewidth. This
ability. For a qualitative insight we consider thus in the
main role is played by the presence of a finite cut-off for the electronic excitations.
However the correct treatment of the charge conserva-
tion and the impurity scattering rate \( \Gamma_{\text{imp}} \propto N(0) \) vanishes.
The resemblance of this framework with the experimental results [10, 17] is anyway striking suggesting that finite bandwidth effects are actually the natural explanation of the evolution of the Raman spectrum with Al doping.

In conclusion in this paper we have investigated the phonon Raman spectroscopy data by computing the phonon self-energy of the \( E_{2g} \) mode. We show that the anomalous features of the Raman measurements, namely the huge phonon linewidth, the low energy background, the two-peak structure as function of the Al doping, can be naturally explained by the interplay of the \( E_{2g} \) phonon mode with the whole electron-phonon spectrum which gives rise to damping processes in the electronic excitation and in the \( E_{2g} \) mode itself.

We thank L. Pietronero, G.B. Bachelet, P. Postorino, M. Lavagnini and D. Di Castro, for interesting discussions. We also acknowledge financial support from the MIUR projects FIRB RBAU017SS8R and COFIN 2003.

[1] J.M. An and W.E. Pickett, Phys. Rev. Lett. 86, 4366 (2001).
[2] J. Kortus et al., Phys. Rev. Lett. 86, 4656 (2001).
[3] T. Yildirim et al., Phys. Rev. Lett. 87, 037001 (2001).
[4] A.Y. Liu, I.I. Mazin, and J. Kortus, Phys. Rev. Lett. 87, 087005 (2001).
[5] Y. Kong et al., Phys. Rev. B 64, 020501 (2001).
[6] H.J. Choi et al., Nature 418, 758 (2002).
[7] F. Bouquet et al., Phys. Rev. Lett. 87, 047001 (2001).
[8] P. Szabo et al., Phys. Rev. Lett. 87, 137005 (2001).
[9] F. Giubileo et al., Phys. Rev. Lett. 87, 177008 (2001).
[10] S. Tsuda et al., Phys. Rev. Lett. 91, 127001 (2003).
[11] R.S. Gonnelli et al., Phys. Rev. Lett. 89, 247004 (2003).
[12] J.S. Slusky et al., Nature 410, 343 (2001).
[13] K.-P. Bohnen, R. Heid, and B. Renker, Phys. Rev. Lett. 86, 5771 (2001).
[14] J. Hlinka et al., Phys. Rev. B 64, 140503 (2001).
[15] A.F. Goncharov et al., Phys. Rev. B 64, 100509 (2001).
[16] P. Postorino et al., Phys. Rev. B 65, 020507 (2002).
[17] B. Renker et al., Phys. Rev. Lett. 88, 067001 (2002).
[18] J. W. Quilty et al., Phys. Rev. Lett. 88, 087001 (2002).
[19] H. Martinho et al., Solid State Commun. 125, 499 (2003).
[20] A. Shukla et al., Phys. Rev. Lett. 90, 095506 (2003).
[21] M. Lazzeri, M. Calandra, and F. Mauri, Phys. Rev. B 68, 220509 (2003).
[22] M. Calandra and F. Mauri, Phys. Rev. B 71, 064501 (2005).
[23] P.B. Allen, Phys. Rev. B 6, 2577 (1972).
[24] R. Zeyher and G. Zwicknagl, Solid State Commun. 66, 617 (1988).
[25] F. Marsiglio, R. Akis, and J.P. Carbotte, Phys. Rev. B 45, 9865 (1992).
[26] C.O. Rodriguez et al., Phys. Rev. B 42, 2692 (1990).
[27] B. Friedl, C. Thomsen, and M. Cardona, Phys. Rev. Lett. 65, 915 (1990).
[28] F. Marsiglio, M. Schossmann, and J. P. Carbotte, Phys. Rev. B 37, 4965 (1988).
[29] A.A. Golubov et al., J. Phys.: Condens. Matter 14, 1353 (2002).
[30] L. Boeri et al., Phys. Rev. B 65, 214501 (2002).
[31] G. Profeta, A. Continenza, and S. Massidda, Phys. Rev. B 68, 144508 (2003).
[32] L. Boeri, E. Cappelluti, and L. Pietronero, Phys. Rev. B 71, 012501 (2005).
[33] A. Zawadowski and M. Cardona, Phys. Rev. B 42, 10732 (1990).
[34] T. Dahm, D. Manske, and L. Tewordt, Phys. Rev. B 59, 14740 (1999).
[35] J.J. Tu et al., Phys. Rev. Lett. 87, 277001 (2001).
[36] R.A. Kaindl et al., Phys. Rev. Lett. 88, 027003 (2002).