Effect of the electron-phonon interaction on spectroscopies of graphene

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In collaboration with: V.P. Gusynin, J.P. Carbotte, E.J. Nicol

Talk is based on: J.P. Carbotte, E.J. Nicol, S.G. Sharapov, unpublished and V.P. Gusynin, S.G. Sharapov, J.P. Carbotte, to appear in New J. Phys. (graphene issue, editor N.M.R. Peres)

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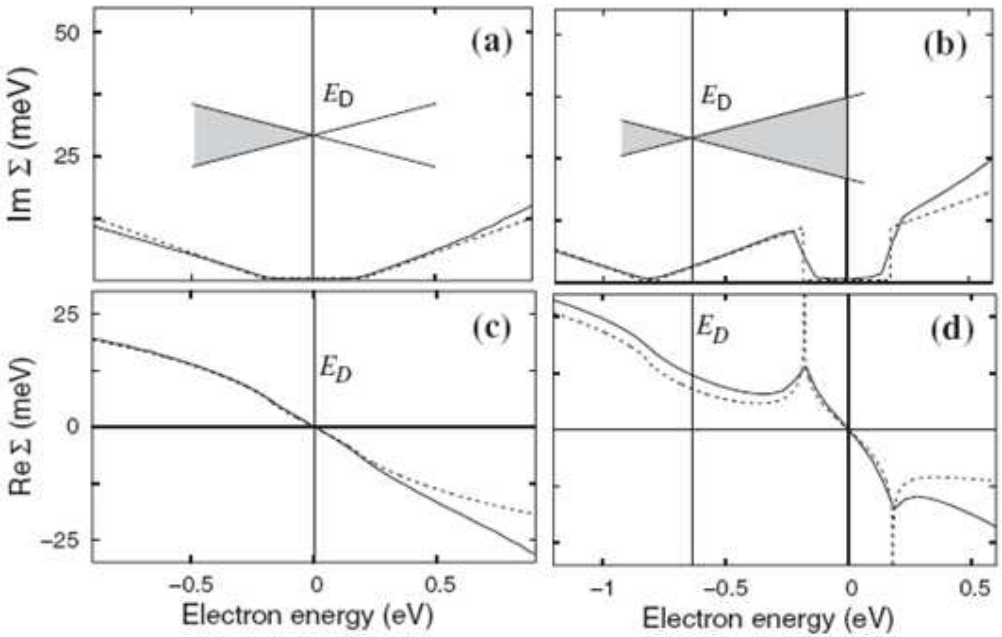
Outline

1. Model for electron-phonon interaction in graphene
2. Velocity and chemical potential renormalization
3. The influence of self-consistency
4. DOS modification by phonons
5. More realistic phonon spectra: magic formula
6. Renormalization and ARPES
7. Electrical conductivity in the presence of phonons
8. AC background and evolution of Drude peak $B \neq 0$

Model for e-ph interaction in graphene

Park, Giustino, Cohen, Louie PRL 99, 086804 (2007) did full first-principles calculations for the electron-phonon interaction in graphene.



Found self-energy could be well-approximated (within 15%) by in-plane Einstein phonon spectrum at frequency $\omega_E = 200\,\text{meV}$
The self-energy is independent of electron momentum and band index.

Notice, $\mathrm{Im}\Sigma$ has the wrong sign

Solid lines – first-principles
Model – dashed lines
left – intrinsic; right – electron-doped

Main equations

Consider $T = 0$.

$$\Sigma(\omega) = \int_{-\infty}^{+\infty} d\omega' \frac{N(\omega')}{N_\circ} \frac{A}{W_C} \left[\frac{\theta(\omega')}{\omega - \omega' - \omega_E + i0^+} + \frac{\theta(-\omega')}{\omega - \omega' + \omega_E + i0^+} \right],$$

where $A = 250\,\text{meV}$ is the coupling, $W_C = \sqrt{\pi\sqrt{3}}t = 7\,\text{eV}$ is the cutoff on the Dirac cone (preserves BZ volume), and $N_\circ = 2/\pi\hbar^2 v_0^2$ with the bare Fermi velocity v_0.

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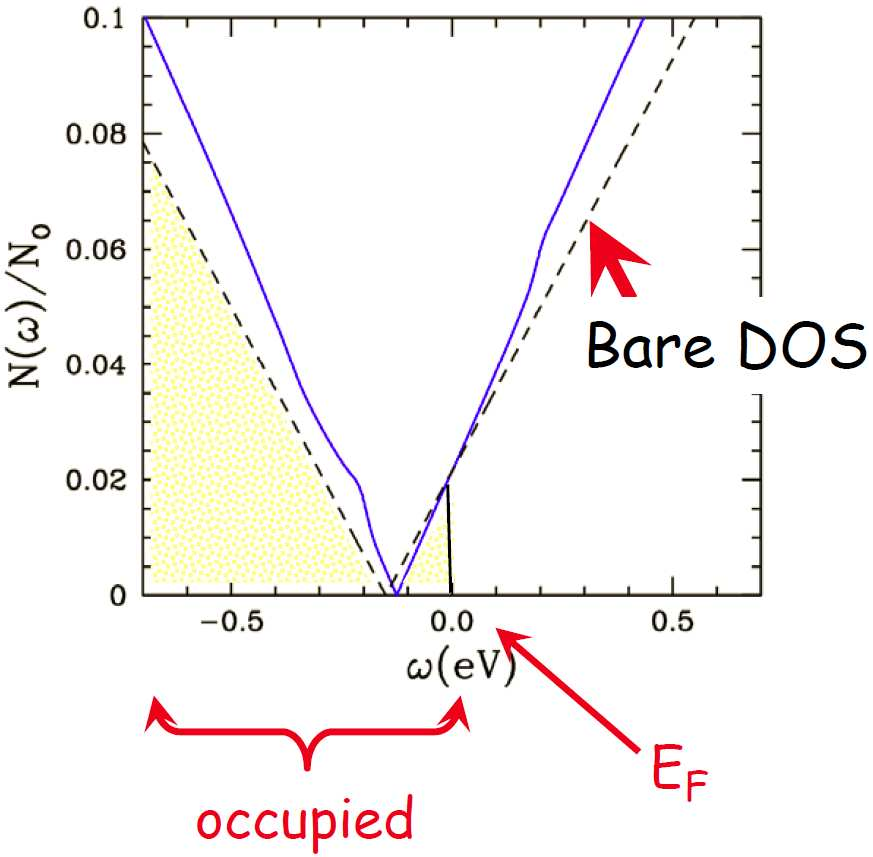
$$\frac{N(\omega)}{N_\circ} = \int_{-W_C}^{W_C} d\epsilon \frac{|\epsilon|}{\pi} \frac{-\text{Im}\Sigma(\omega)}{[\omega - \text{Re}\Sigma(\omega) + \mu - \epsilon]^2 + [\text{Im}\Sigma(\omega)]^2}.$$

Here μ is the chemical potential of interacting system and μ_0 (will appear below) is the chemical potential for the bare bands.

Main equations: conventions and features

By choice $\omega = 0$ corresponds to the Fermi level in noninteracting and interacting cases.



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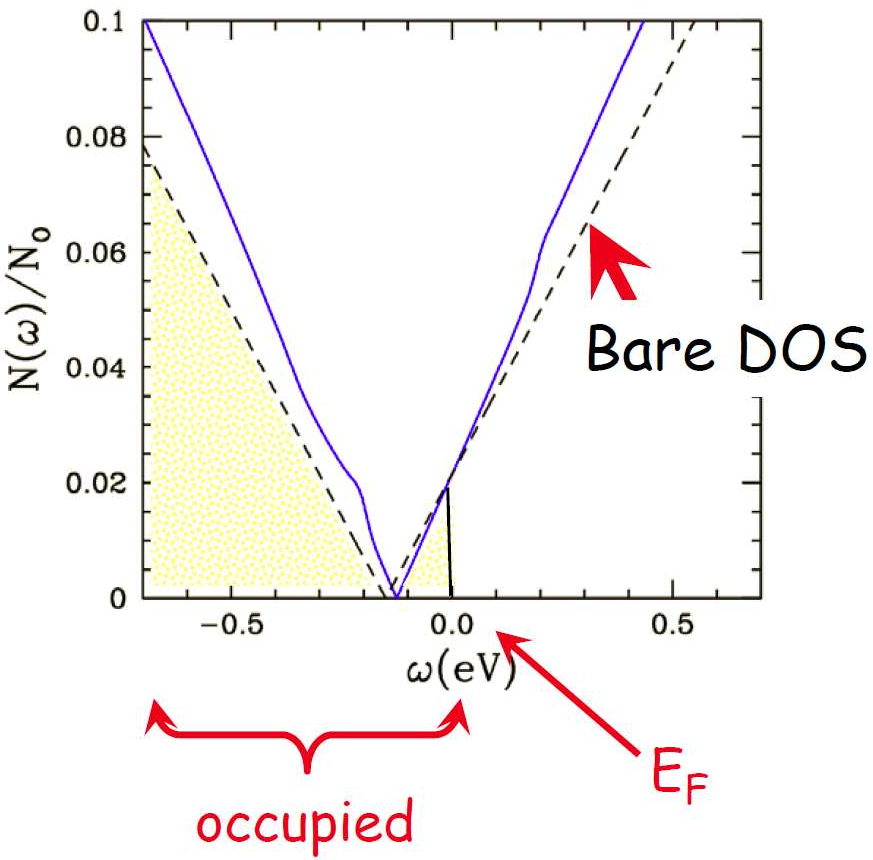

The value of the DOS at the Fermi surface remains pinned to its noninteracting value:

$$\frac{N(\omega=0)}{N_\circ} = \mu - \mathrm{Re}\Sigma(\omega = 0) = \mu_0$$

Will show why last equality holds.

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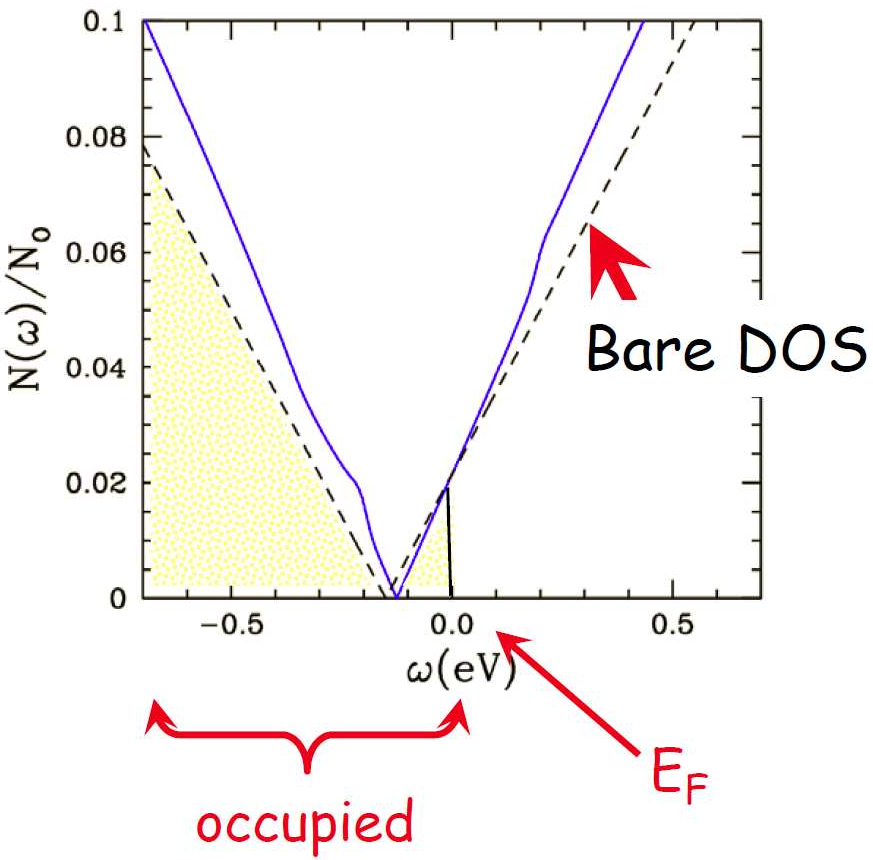

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Notice, in some papers including our own in New J. Phys. use different convention with $\omega = \mu$!

First approximation: bare DOS

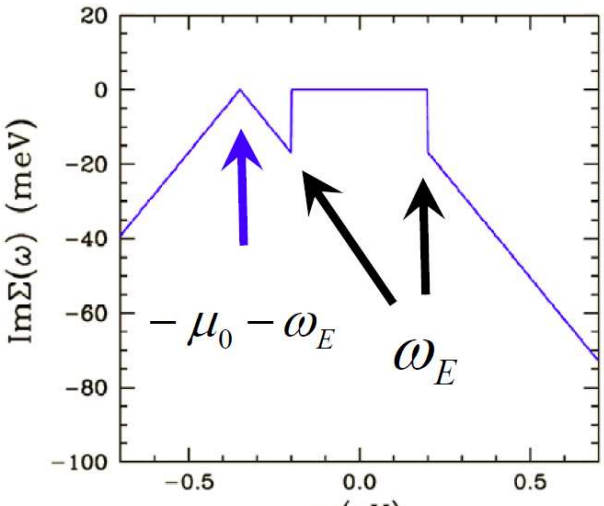
Getting used to the self-energy

$$\frac{N(\omega)}{N_\circ} = \begin{cases} |\omega + \mu_0|, & -W_C - \mu_0 < \omega < W_C - \mu_0, \\ 0, & \text{otherwise}, \end{cases}$$

In this case self-energy $\Sigma(\omega; \mu_0)$ can be calculated analytically:

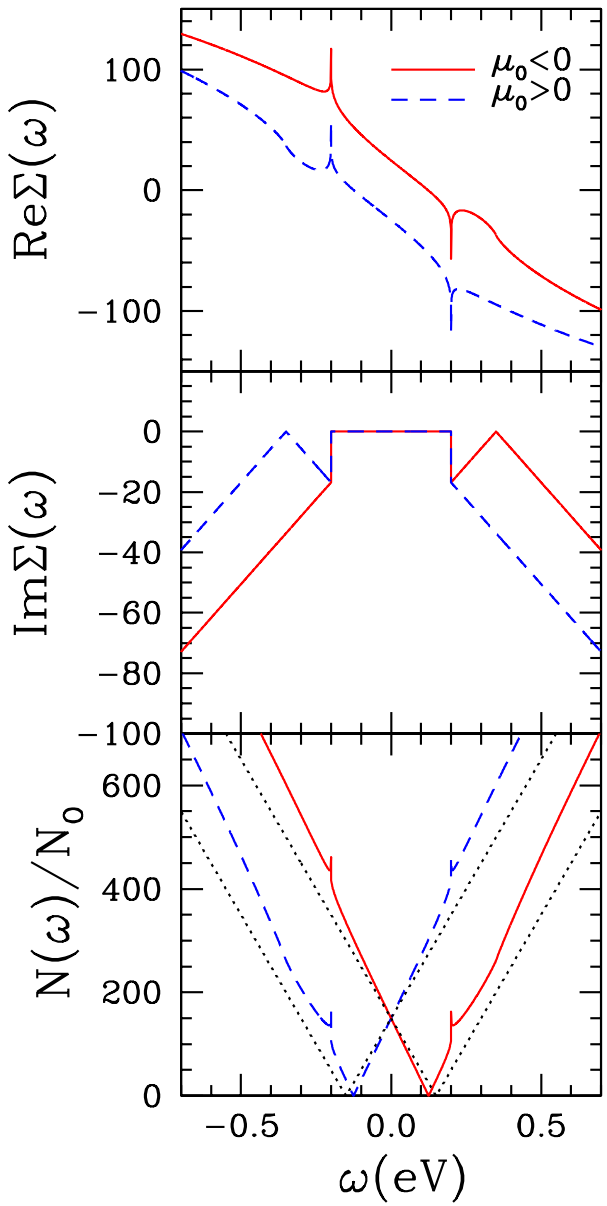
$$-\mathrm{Im}\Sigma(\omega) = \begin{cases} \frac{\pi A}{W_C}|\omega - \omega_E + \mu_0|, & \omega_E < \omega < W_C - \mu_0 + \omega_E, \\ \frac{\pi A}{W_C}|\omega + \omega_E + \mu_0|, & -\omega_E > \omega > -W_C - \mu_0 - \omega_E, \end{cases}$$

$\mathrm{Re}\Sigma(\omega)$ is lengthy and explicitly depends on $\ln W_C$



M. Calandra and F. Mauri, PRB **76**, 205411 (07); W.-K. Tse and S. Das Sarma PRL **99**, 236802 (07); T. Stauber and N.M.R. Peres, J.Phys. Cond. Matt. **20**, 055002 (2008).

First iteration approximation



Real (top) and imaginary (middle) part of $\Sigma(\omega)$ and DOS [first iteration] $N(\omega)/N_\circ$ (bottom) (all in units of meV) as a function of ω in eV for $|\mu_0| = 150\,\mathrm{meV}$. Dashed is for $\mu_0 > 0$ and solid is for $\mu_0 < 0$. The bare band DOS is indicated by the dotted curve.

Mass (velocity!) and μ_0 renormalization

Carrier effective mass renormalization $\lambda^{\rm eff}$ due to the e-ph interaction: $\mathrm{Re}\Sigma(\omega) = -\lambda^{\rm eff}\omega + \mathrm{Re}\Sigma(\omega = 0)$ for $\omega \to 0$. From known analytical expression for $\Sigma(\omega)$ we obtain

$\lambda_{eff} = \frac{2A}{W_C}\left(\ln\frac{W_C}{|\mu_0+\omega_E|} - 1 + \frac{|\mu_0|}{\omega_E}\right).$

Energies E_k are obtained from the pole condition:

$\omega - \mathrm{Re}\Sigma(\omega) + \mu - \epsilon = 0 \Rightarrow E_k + \lambda^{\rm eff} E_k - \mathrm{Re}\Sigma(\omega = 0) + \mu = \pm v_0|\mathbf{k}|$

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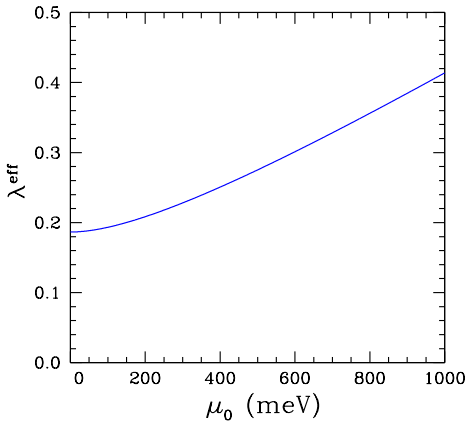
Thus the dispersion becomes $E_k = \dfrac{\pm\hbar v_0 k - \mu_0}{1 + \lambda^{\rm eff}}$.

Main conclusions for renormalizations: 1-2

1) For the massless carriers in graphene the role of λ_{eff} is to renormalize their velocity: $v_0 \to v_0/(1+\lambda_{eff})$.
If we take $A = 250\,\text{meV}$, obtain $\lambda_{eff} \sim 0.19$ which is larger than calculated in density functional theory (Park), but smaller than measured by ARPES.



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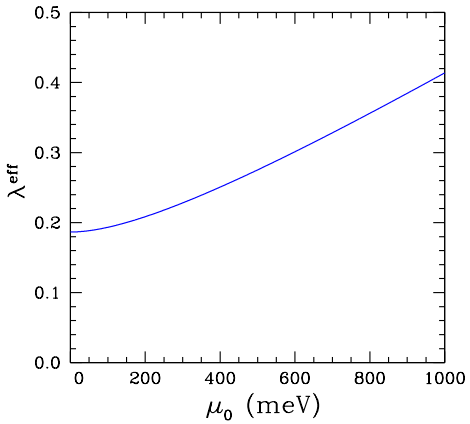

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2) The value of μ_0 is renormalized (J.M. Luttinger and J. C. Ward, Phys. Rev. **118**, 1417 (1960)). Since $\mathrm{Re}\Sigma(\omega = 0) = -\mu_0\lambda^{\mathrm{eff}}$ then

$$\mu = \mu_0(1-\lambda^{\mathrm{eff}}) \simeq \frac{\mu_0}{1+\lambda^{\mathrm{eff}}}$$

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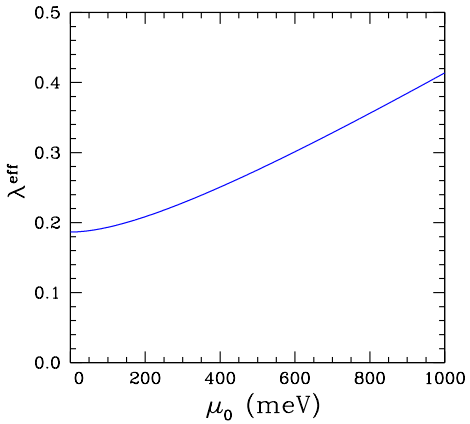

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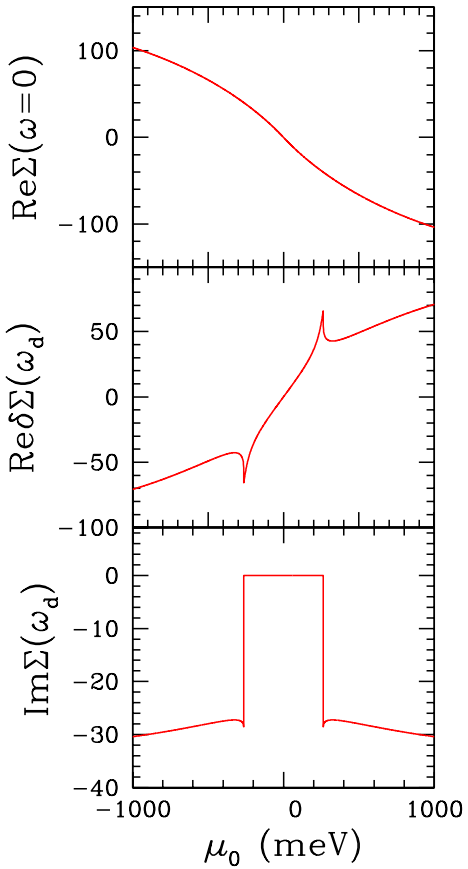
Main conclusions for renormalizations: 3-4

3) If we include imaginary part due to impurities, $\eta = -\mathrm{Im}\Sigma$: $\eta \to \eta/(1+\lambda_{\mathrm{eff}})$

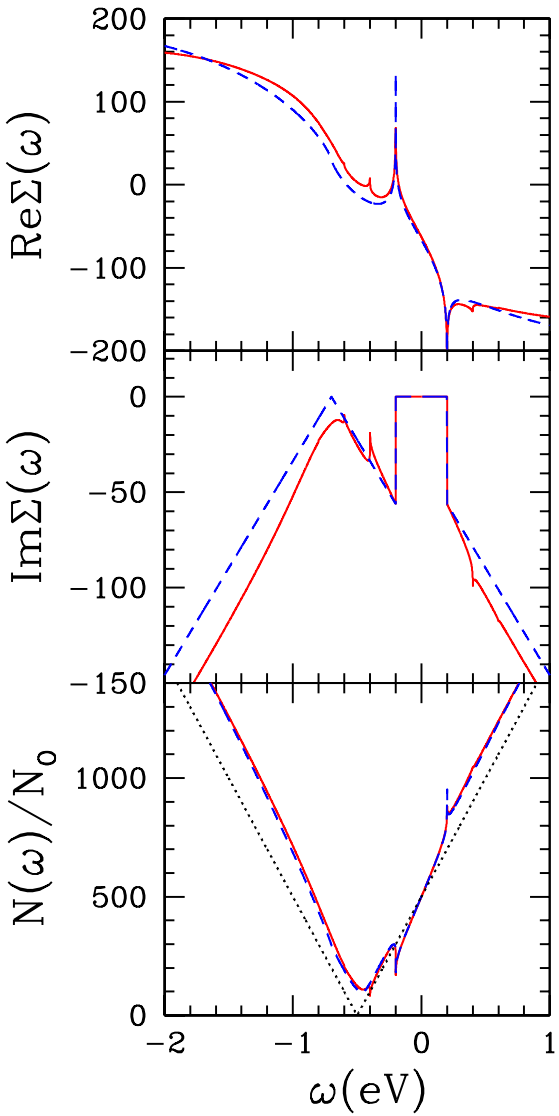
4) The position of the Dirac point is also shifted from $\omega = -\mu_0$ to $\omega_d \simeq -\mu_0/(1+\lambda_{\mathrm{eff}})$.



$\omega_d = -\mu_0 + \mathrm{Re}\delta\Sigma(\omega_d)$

Results of a first iteration for the shift in chemical potential $\mathrm{Re}\Sigma(\omega=0)$ (top frame), the shift in position of the Dirac point $\mathrm{Re}\delta\Sigma(\omega_d)$ (middle frame) and the imaginary part of the self-energy $\mathrm{Im}\Sigma(\omega_d)$ at the Dirac point (lower frame). All of these quantities are shown in units of meV.

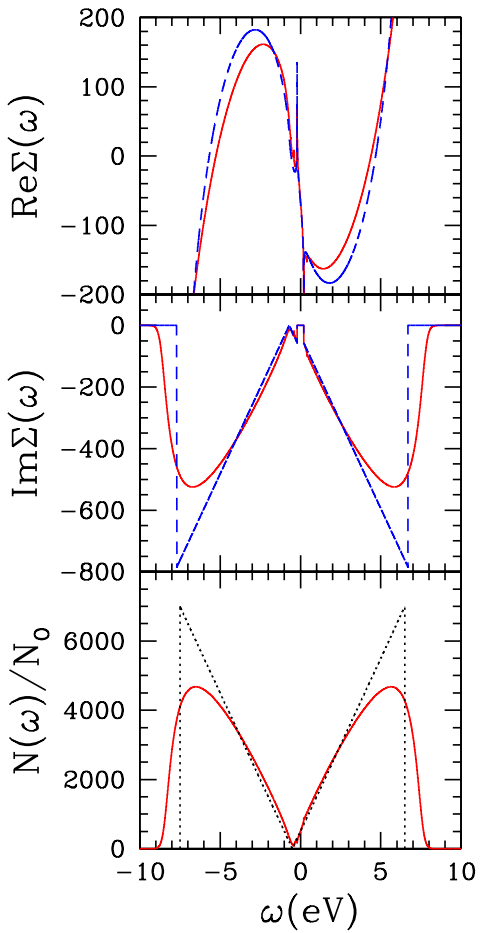
The effect of self-consistency I



$\Sigma(\omega)$ and DOS have been self-consistently iterated and $\mu_0 = 500\,\text{meV}$ which is greater than ω_E. The red solid curves are for the iterated case and the blue dashed curves are for the initial uniterated results. The dotted curve is the bare density of states. All quantities on the y-axis are in meV.

The effect of self-consistency II

Looking closer at the band edge

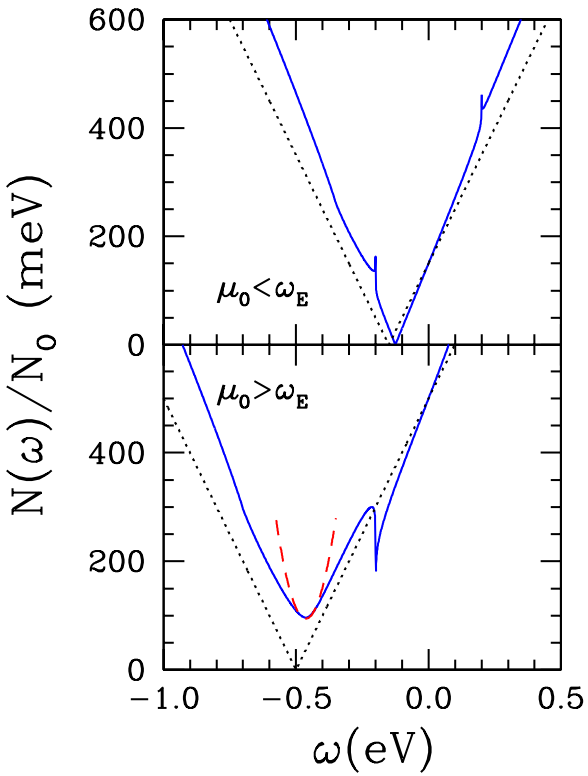


Top of the renormalized band extends to higher energies as compared with the bare band and the bottom extends to lower energies, F. Dogan and F. Marsiglio, PRB 68, 165102 (03); A. Knigavko and J.P. Carbotte, PRB **72**, 035125 (05). The phonon energy sets the scale for this smearing beyond the bare band edge. $\Sigma(\omega)$ and DOS have been self-consistently iterated and $\mu_0 = 500\,\text{meV}$ which is greater than ω_E. The red solid curves are for the iterated case and the blue dashed curves are for the initial uniterated results. The black dotted curve is the bare density of states. All quantities on the y-axis are in meV.

DOS modification by phonons I

For ω near ω_d and ϵ near $\epsilon = 0$, the spectral functions that determine the DOS are
$A(\pm\epsilon,\omega) \simeq \frac{1}{\pi}\frac{-\mathrm{Im}\Sigma(\omega_d)}{[\omega-\mathrm{Re}\Sigma(\omega_d)-\mathrm{Re}\Sigma'(\omega_d)(\omega-\omega_d)+\mu\pm\epsilon]^2+[\mathrm{Im}\Sigma(\omega_d)]^2}$.
Denoting $1-\mathrm{Re}\Sigma'(\omega_d)$ by Z, where $\Sigma'(\omega_d) \equiv d\Sigma(\omega)/d\omega|_{\omega=\omega_d}$, and $-\mathrm{Im}\Sigma(\omega_d)$ by Γ, we obtain $A(\pm\epsilon,\omega) \simeq \frac{1}{\pi}\frac{\Gamma}{[(\omega-\omega_d)Z\pm\epsilon]^2+\Gamma^2}$.
Then for $\mu_0 > \omega_E$ and $|(\omega-\omega_d)Z| \ll \Gamma$ we get that the Dirac point is lifted and the DOS becomes quadratic at that point due to finite scattering rate: $\frac{N(\omega)}{N_0} = \frac{2\Gamma}{\pi}\ln\left|\frac{W_C}{\Gamma}\right| + \frac{(\omega-\omega_d)^2Z^2}{\pi\Gamma}$.
For $\mu_0 < \omega_E$ as $\Gamma \to 0$ recover linear in ω DOS.

DOS modification by phonons II



$N(\omega)$ (solid blue line) vs. ω for $\omega_E = 200\,\text{meV}$, $\lambda = 2A/\omega_E = 2.5$. The top frame is for $\mu_0 = 150\,\text{meV} < \omega_E$ and the bottom for $\mu_0 = 500\,\text{meV} > \omega_E$. The dotted curve is the bare band case. For $\mu_0 > \omega_E$, $N(\omega)$ at the Dirac point is nonzero and becomes quadratic. A comparison of this approximate quadratic behavior is shown as the dashed (red) curve.

More realistic phonon spectra

Magic formula

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Consider distribution of phonon energies: $\Sigma_{\rm lor}(\omega) = \int_{-\infty}^{\infty} P(\nu)\Sigma(\omega,\nu)d\nu$, where we used truncated Lorentzian (F. Dogan and F. Marsiglio, PRB **68**, 165102 (03)) to model the widths of the phonon peaks.

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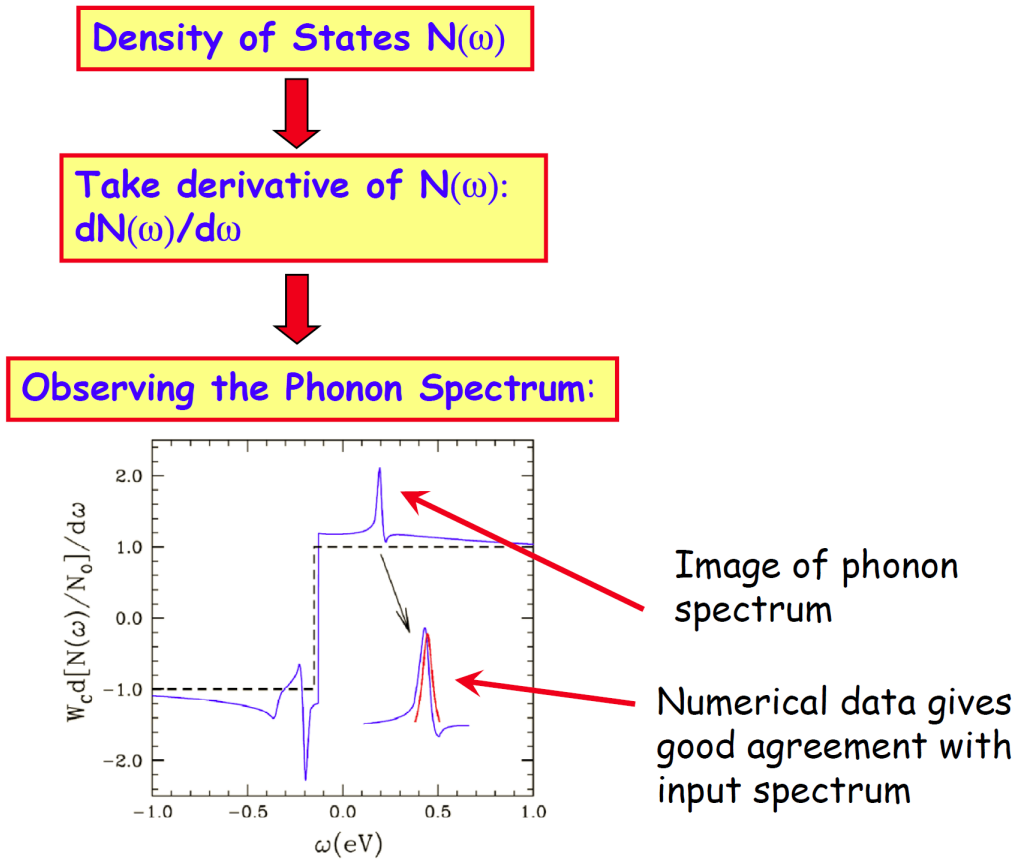

Renormalization and ARPES I

Spectral function $A(\mathbf{k},\omega) = A(\epsilon,\omega), \quad \epsilon \pm \hbar v_0 |\mathbf{k}|$ can be measured by ARPES. Begin with ideal case $\mathrm{Im}\Sigma = 0$, so that the dressed energy E_k is found from the equation $E_k - \mathrm{Re}\Sigma(E_k) + \mu - \epsilon_k = 0$.

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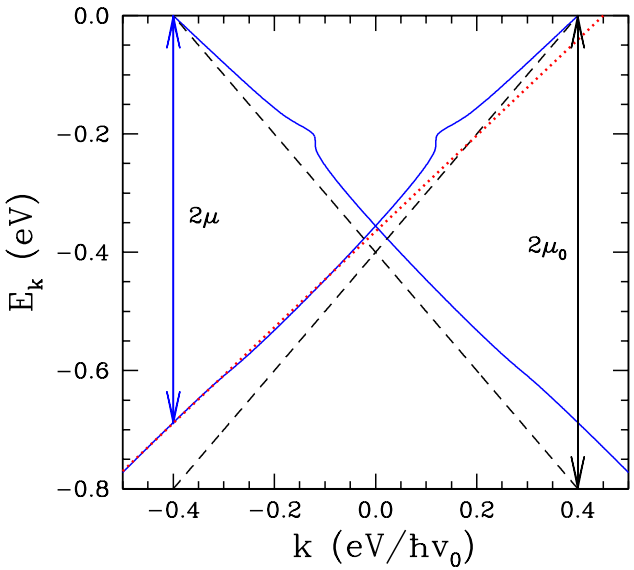

Renormalized energies (solid blue curves) E_k as a function of k in units of $\mathrm{eV}/(\hbar v_0)$ for $\mu_0 = 400\,\mathrm{meV}$. Twice the bare $2\mu_0$ and dressed 2μ chemical potential are indicated by vertical arrows.

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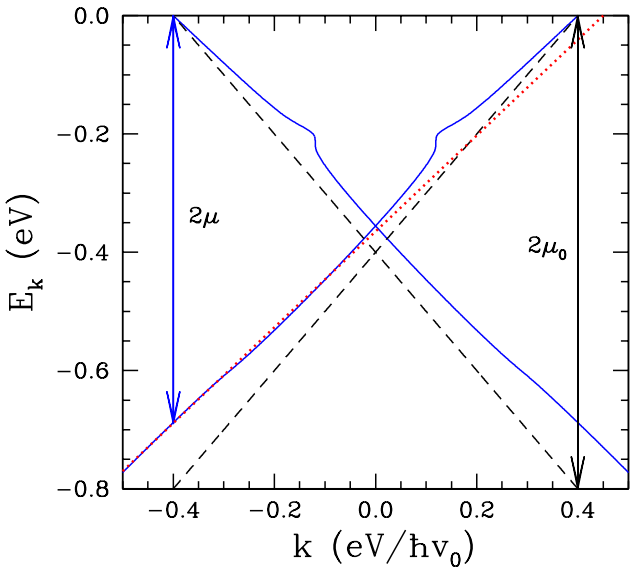

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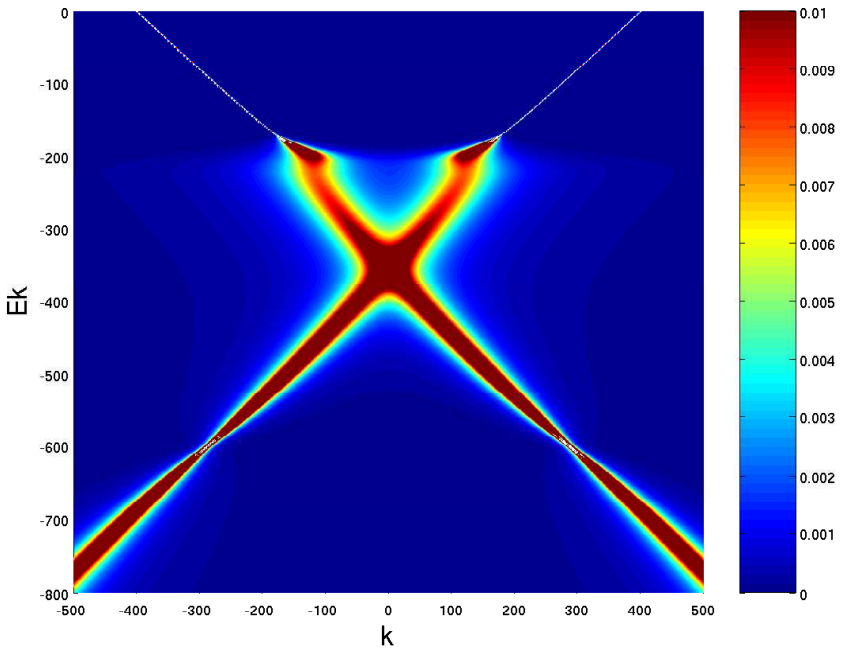
Renormalization and ARPES II

Restore $\mathrm{Im}\Sigma$.



Color map (units of meV^{-1}) of interacting dispersions E_k in units of meV as a function of $\mathbf{k}$ in units of $\mathrm{meV}/(\hbar v_0)$. The bare chemical potential $\mu_0 = 400\,\mathrm{meV}$ and a Lorentzian phonon spectrum with $\omega_0 = 200\,\mathrm{meV}$ were used. Broadening makes determination of μ somewhat ambiguous.

Electrical conductivity

Consider the limit the opposite to the universal one $T = 0$, but $\mu - \mathrm{Re}\Sigma(\omega = 0) \gg \eta$, where $1/\tau = 2\eta$ is the transport scattering rate. The electron-phonon interaction drops out of the DC conductivity:

$$\sigma_{DC}(T = 0) = \frac{e^2}{h} \frac{2[\mu - \mathrm{Re}\Sigma(\omega = 0)]}{2\eta} = \frac{e^2}{h} \frac{2\mu_0}{2\eta}.$$

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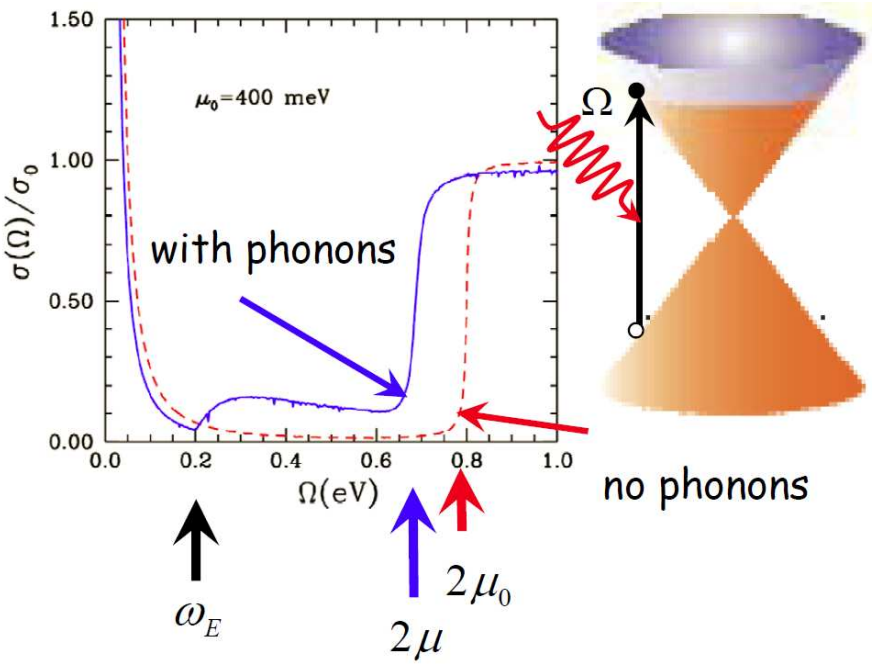
Low frequency conductivity:

$$\sigma_{intra}(\Omega) = \frac{\pi e^2}{2h}\frac{4|\mu_0|}{\pi}\frac{2\eta}{\Omega^2(1+\lambda^{\mathrm{eff}})^2 + 4\eta^2}, \qquad \Omega, \eta \ll \mu_0.$$

Drude form with effective optical scattering rate $2\eta/(1+\lambda^{\mathrm{eff}})$ and effective plasma frequency of $4\mu_0/[\pi(1+\lambda^{\mathrm{eff}})]$.

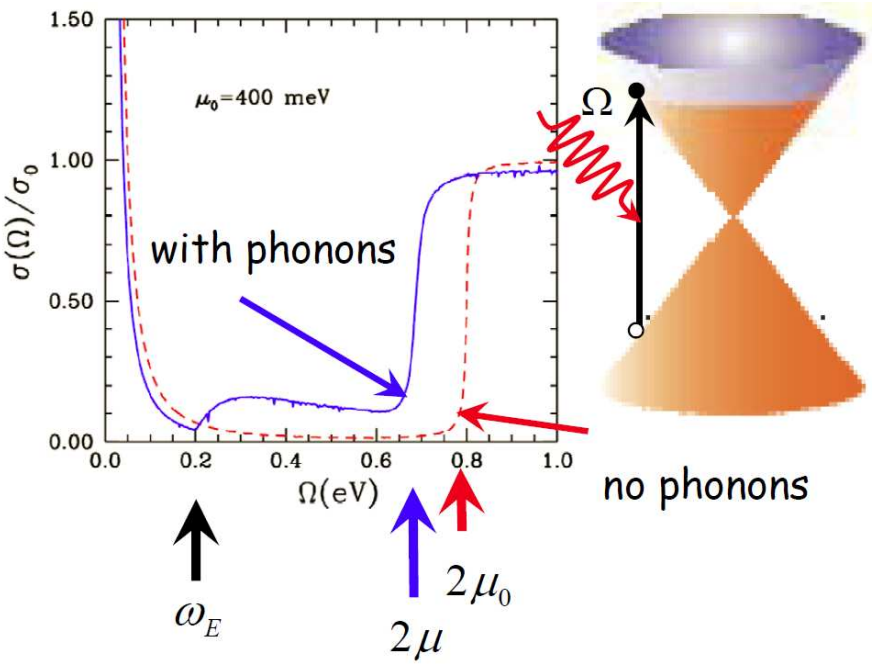
Optical conductivity



$\mathrm{Re}\sigma_{xx}(\Omega)$ in units $\pi e^2/2h$
There is an absorption $\Omega > \omega_E$
(Holstein sideband).

T. Stauber and N.M.R. Peres, J.Phys. Cond. Matt. 20, 055002 (08) did not include a shift of the absorption edge from $2\mu_0$ to 2μ, but also get the reduction of $\sigma(\Omega)$.

Optical conductivity

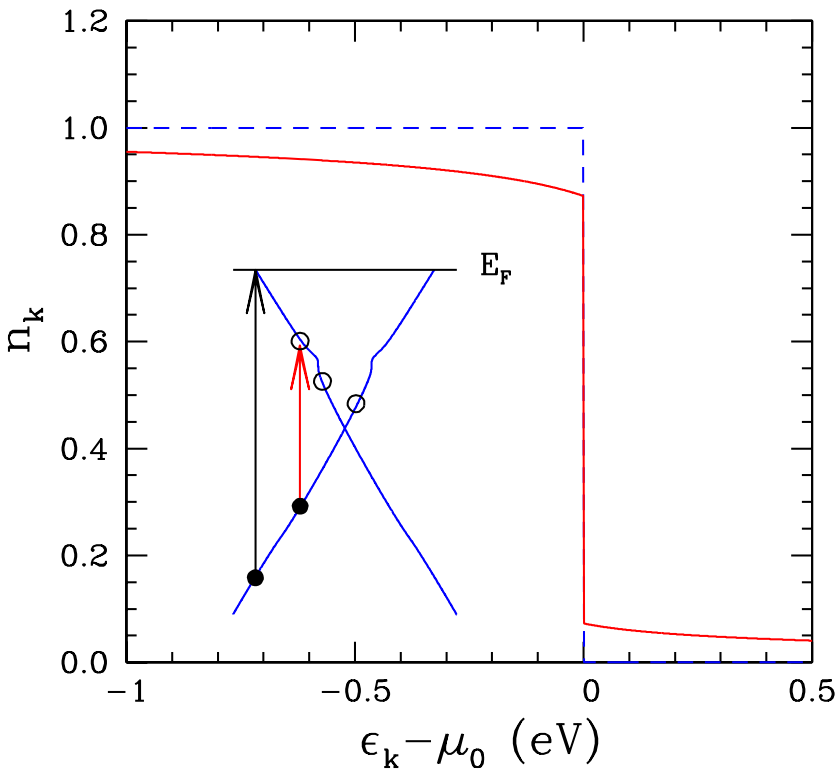


$\mathrm{Re}\sigma_{xx}(\Omega)$ in units $\pi e^2/2h$
There is an absorption $\Omega > \omega_E$ (Holstein sideband).

T. Stauber and N.M.R. Peres, J.Phys. Cond. Matt. 20, 055002 (08) did not include a shift of the absorption edge from $2\mu_0$ to 2μ, but also get the reduction of $\sigma(\Omega)$.

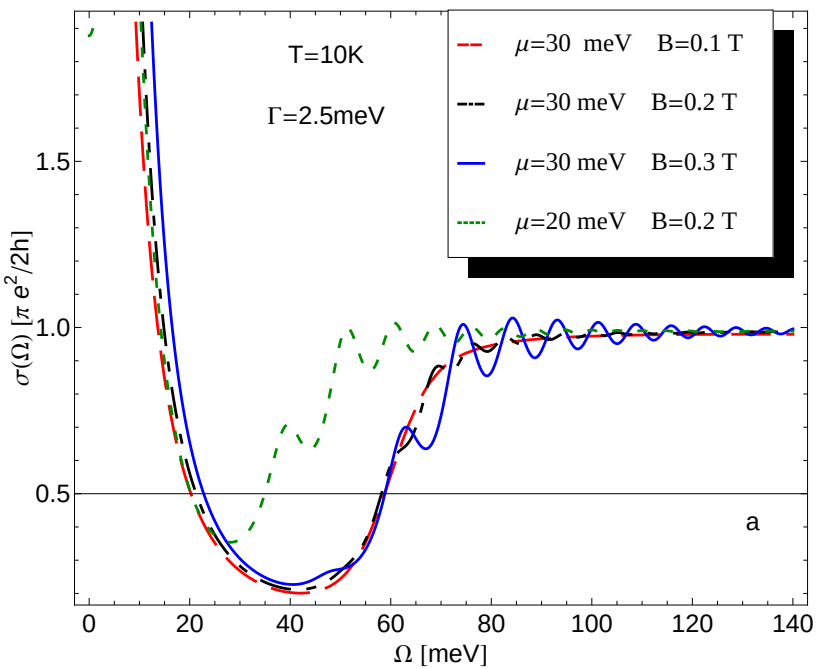
The e-p interaction has reduced the value of the universal background slightly below $\sigma_0 = \pi e^2/2h$ (for finite μ!).
The electron-phonon interaction has a profound effect on the band structure in the energy region around the band edge. The DOS is considerably depleted below its noninteracting value and to conserve states tails appear beyond the bare cut off W_C.
Thus in optical experiments, spectral weight is removed below the bare optical cut off which is transferred to higher energies.

Understanding Holstein sideband



The probability of occupation of a state $\mathbf{k}$, $n_{\mathbf{k}} = \int_{-\infty}^{\infty} f(\omega) A(\mathbf{k}, \omega) d\omega$ for two cases: bare band (dashed blue curve) and with electron-phonon interaction included (solid red curve). The inset is a schematic which illustrates the renormalized energy bands filled to the Fermi level E_F with finite probability for some holes to exist below the Fermi level. Interband transitions are now possible for energies below 2μ.

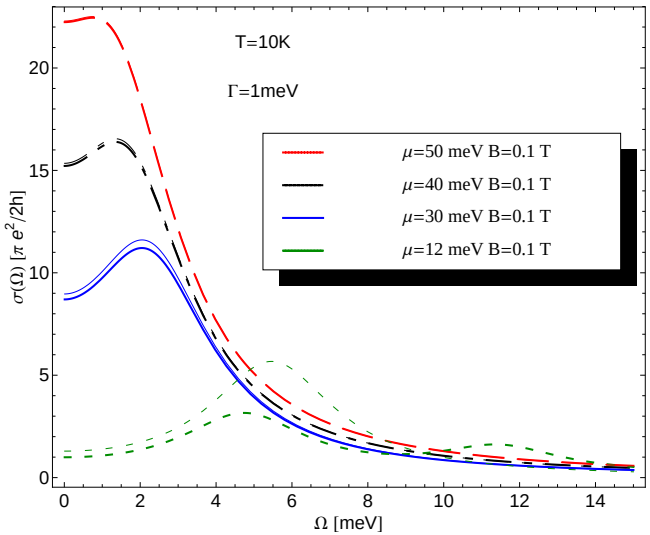
AC background in magnetic field



$\mathrm{Re}\sigma_{xx}(\Omega)$ in units $\pi e^2/2h$ (universal background) for $T = 10\,\mathrm{K}$, scattering rate $\Gamma = 2.5\,\mathrm{meV}$.

It is not surprising that for low B the universal AC background survives. The figures is plotted on the base of the formula which includes the sum over transitions between Landau levels:
V.P. Gusynin, S.G Sh and J.P. Carbotte, J. Phys. Cond. Mat. **19**, 026222 (07).
Is there a simpler low-field representation for conductivity?

Evolution of the Drude peak



$\mathrm{Re}\sigma_{xx}(\Omega)$ in units $\pi e^2/2h$ for $B = 0.1\,\mathrm{T}$, $T = 10\,\mathrm{K}$, and scattering rate $\Gamma = 1\,\mathrm{meV}$. For green line μ is getting close to $E_1 = 11.5\,\mathrm{meV}$.

All thick lines are computed using the full expression with the sum over Landau levels and the thin lines using a simple expression with the "relativistic" cyclotron frequency, $\omega_c = |eB|v_F^2/(c|\mu|)$:

$$\mathrm{Re}\sigma_{xx}(\Omega) = \frac{2e^2}{h} T \ln\left(2\cosh\frac{\mu}{2T}\right) \times \left[\frac{2\Gamma}{(\omega_c-\Omega)^2+4\Gamma^2} + \frac{2\Gamma}{(\omega_c+\Omega)^2+4\Gamma^2}\right]$$

Valid for $E_1 = L(B) = \sqrt{2eB\hbar v_F^2/c} \lesssim |\mu|$.

Summary

- Any conclusive comparison with experiment would need to include additional interactions! So we did not try to fit the data.
- Because the bare band DOS is linear in energy rather than constant, an image of the phonons is retained in dressed DOS and a first derivative of $N(\omega)$ provides an ideal baseline to study boson structures.
- Renormalization of v_0, μ, η, ω_d by $1 \to 1/(1+\lambda_{eff})$.
- Drude peak evolves into a peak at the cyclotron frequency.

Thank you very much for inviting and listening!