MAX-PLANCK-GESELLSCHAFT

Many-Body effects in the Near-Field Optics of Graphene

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J.P.F. LeBlanc et al. Phys. Rev. B 84, 165448 (2011)

Overview

Many-Body Interactions

- Electron-electron interaction (EEI)
  - Plasmarons in spectral function and DOS
- Electron-phonon interaction (EPI)
  - Impact on energy and DOS

- Near-Field Optics – Current-Current Correlation Function
  - Finite Momentum Optical Conductivity (Longitudinal)
    - Removal of Pauli Blocking
    - Correspondence to Quasiparticle Peaks
  - Many-Body, EEI and EPI

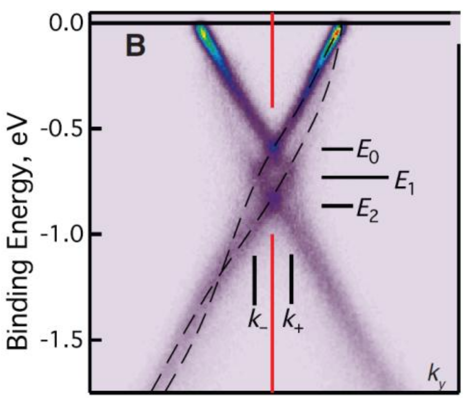
Strain (arXiv:1303.0131)

- EEI Correlations
  - Single Particle Spectral Density
  - Plamaron Ring Structure

Observation of Plasmarons

Angle Resolved Photoemission Spectra

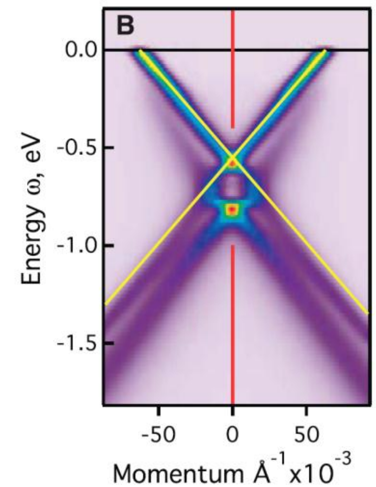
Bostwick et al Science **328**, 999 (2010)



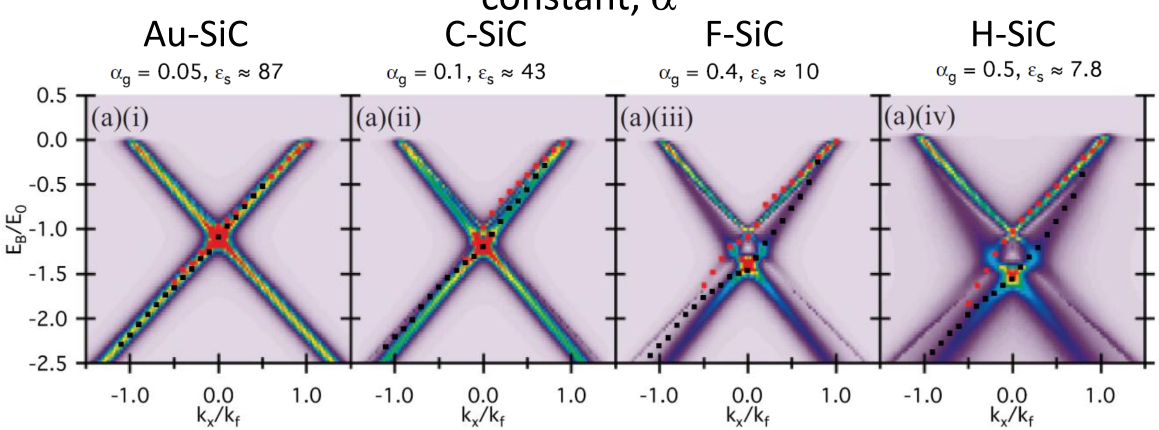
Well described through G_0W-RPA calculation for electron-electron interactions (EEI)

Renormalizations are frequency, ω, and momentum, **k**, dependent

G_0W-RPA



Plasmaron 'Ring' used to extract effective fine structure constant, α



How do plasmaron features manifest in the electronic density of states?

How does one separate EEI from other interactions?

Walters et al. PRB **84**, 085410 (2011)

Electron-Phonon Interactions (EPI)

LDA calculations suggest EPI independent of electron momentum

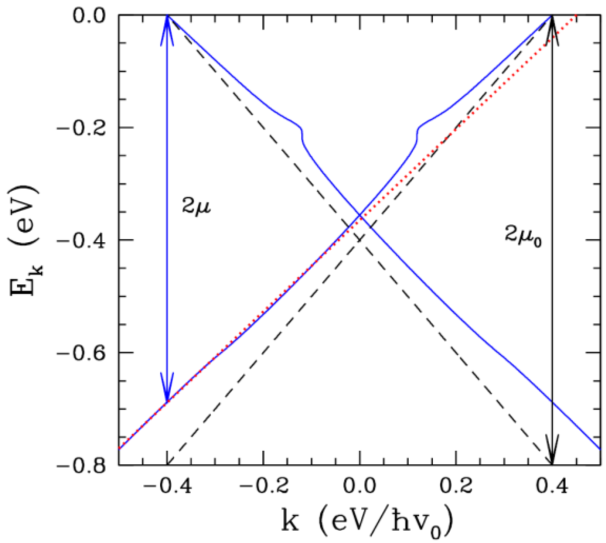
Park et al. PRL **99,** 086804 (2007)

α^2F spectrum distributed at 200meV

Kink feature indicating phonon mode

Renormalized energy

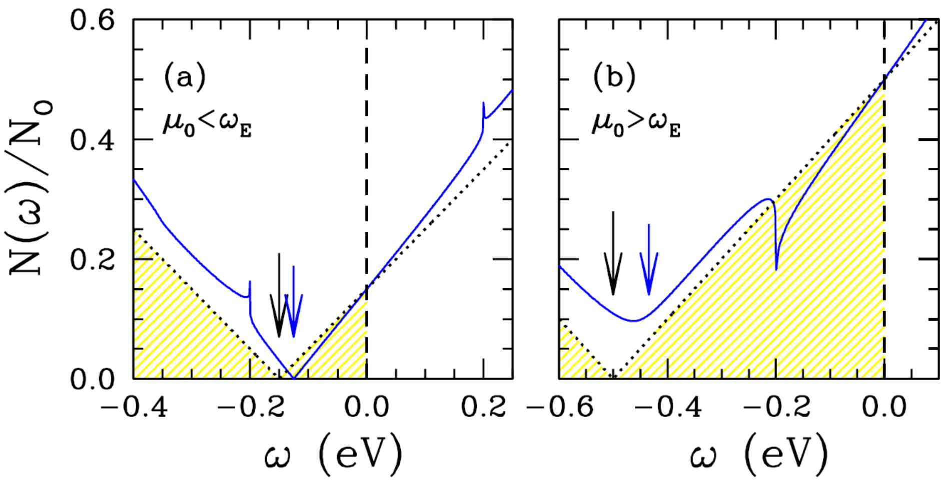
$$E_k = \frac{\epsilon_k}{(1+\lambda)}$$



Density of states, N(w)

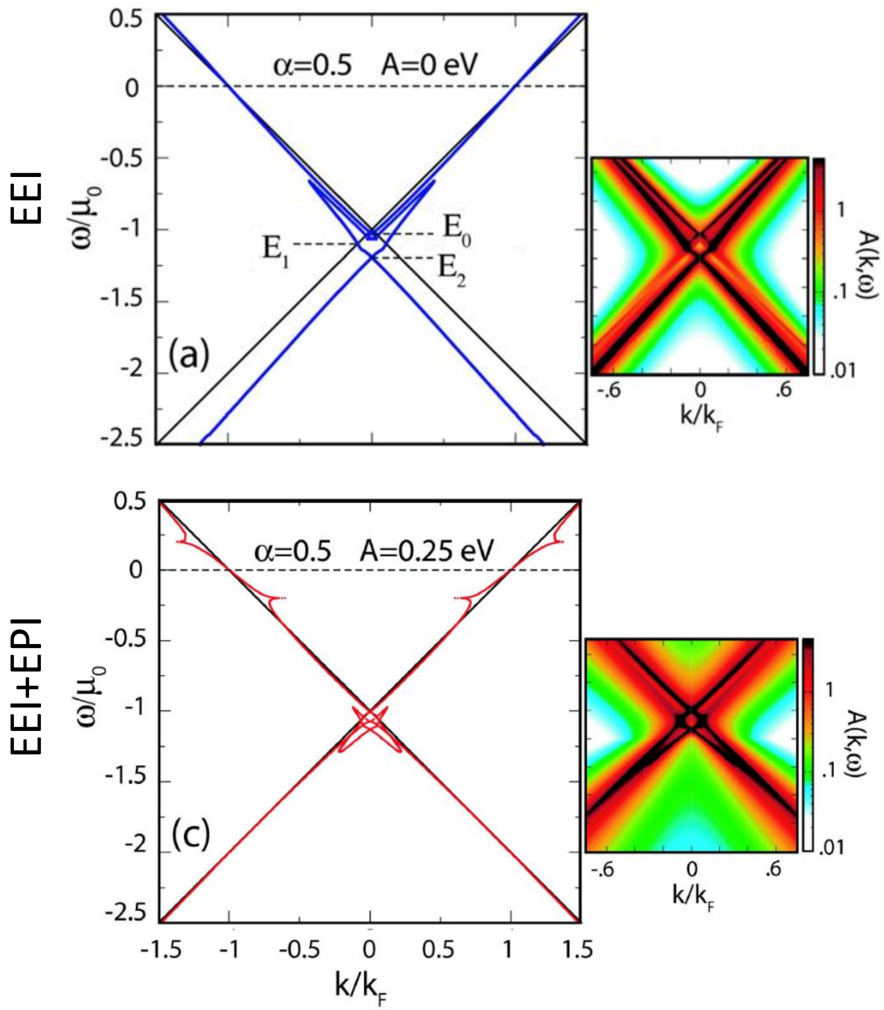
-N(0) unchanged

-Increase in slope at ω=0



Nicol et al. PRB **80**, 081415(R) (2009) *Nicol et al.* PRB **81**, 045419 (2010)

Modified Collective Modes



$$E_k = \epsilon_k + \mathrm{Re}\,\Sigma(k,\omega)$$

G_0W-RPA

$$\Sigma(k,\omega) = \Sigma^{EEI}(k,\omega) + \Sigma^{EPI}(\omega)$$

Real part of phonon self energy can shift spectral peaks. Particularly evident on the lower Dirac cone.

G_0W-RPA

<u>*Polini et al.*</u>

Phys. Rev. B **77**, 081411(R) (2008)

<u>*Hwang and Das Sarma*</u>

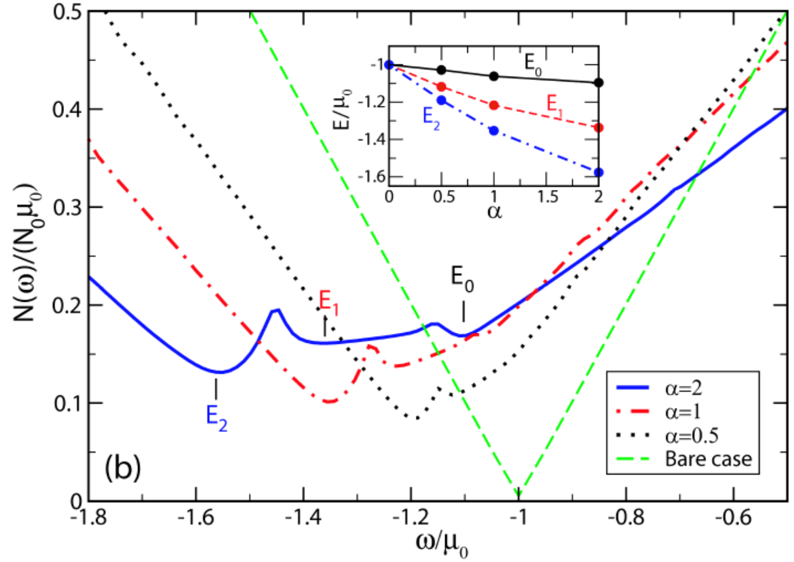
Phys. Rev. B **75**, 205418 (2007)
Phys. Rev. B **77**, 081412(R) (2008)

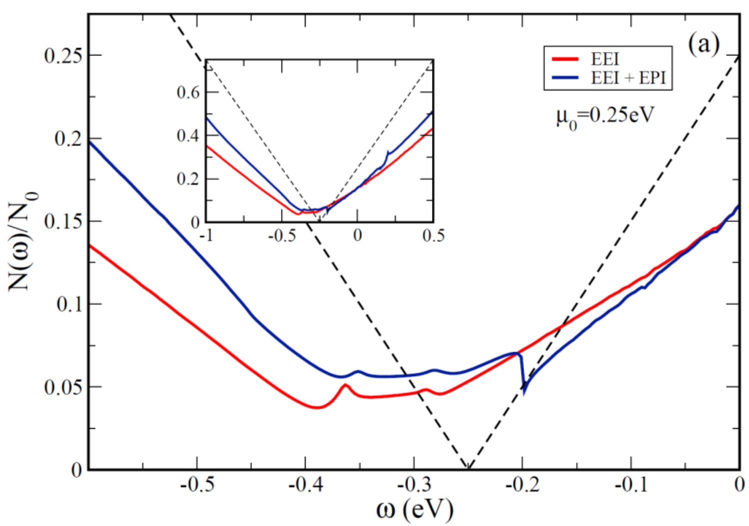
 J.P.F. LeBlanc et al. Phys. Rev. B 84, 165448 (2011)

Electronic Density of States

Electron-electron interactions (EEI)

- -Splitting of Dirac points into two
- -Each has parabolic signature in $N(\omega)$
- -Slope modified with increasing α
- -Features scale with chemical potential, μ_0
- -Verified, *Principi et al S.S. Comm.* ***152****, 1456 (2012)*





EEI+EPI

- N(0) can be significantly depressed from bare case due to EEI
- EPI does not change N(0)
- EPI increases the slope, opposite to EEI

J.P.F. LeBlanc et al. Phys. Rev. B 84, 165448 (2011)

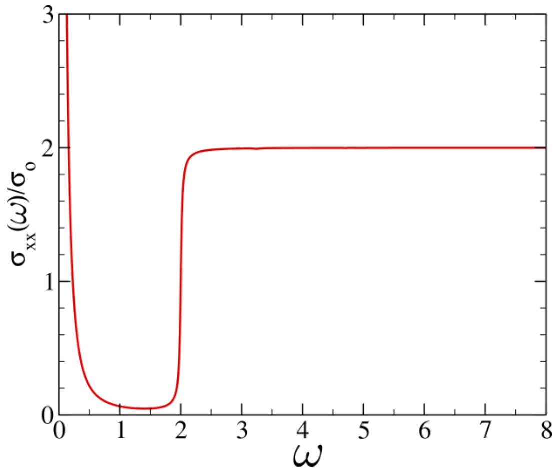
Density-Density vs Current-Current

Generally Exploited in Standard Optics

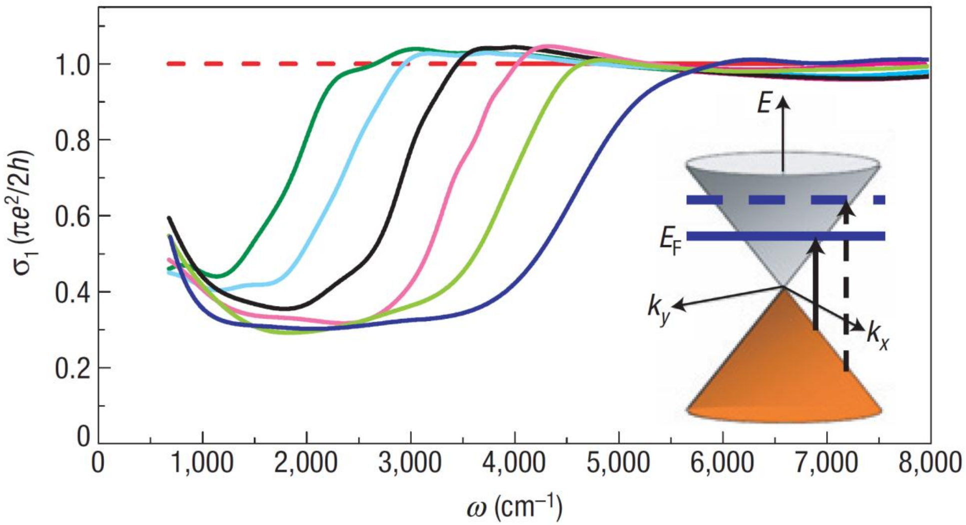
$$\Pi(q \to 0, \omega) = \frac{gq^2}{8\pi\hbar\omega}\left[\frac{2\mu}{\hbar\omega} + \frac{1}{2}\ln\left|\frac{2\mu - \hbar\omega}{2\mu + \hbar\omega}\right| - i\frac{\pi}{2}\Theta(\hbar\omega - 2\mu)\right]$$

$$\sigma(\omega) = \lim_{q \to 0} ie^2 \frac{\omega\Pi(\mathbf{q}, \omega)}{\mathbf{q}^2}$$

Non-Interacting Theory



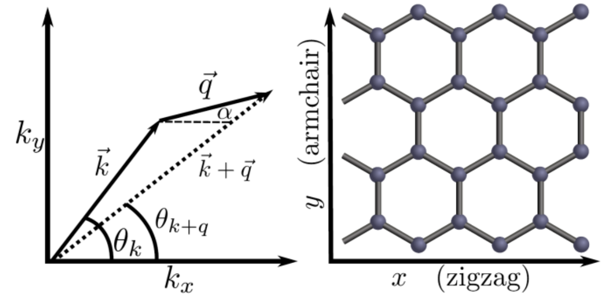
Experiment



Basov et al. Nature Phys. ***4****, 532 (2008)*

Key Difference in Band Overlap

Interacting Self-Energy in $A(\mathbf{k},\omega)$



$$A(\mathbf{k},\omega)=\frac{1}{\pi}\frac{\left|\mathrm{Im}\,\Sigma_s(\mathbf{k},\omega)\right|}{\left[\omega-\epsilon_k^s-\mathrm{Re}\,\Sigma_s(\mathbf{k},\omega)\right]^2+\left[\mathrm{Im}\,\Sigma_s(\mathbf{k},\omega)\right]^2}$$

$$\frac{\sigma(\boldsymbol{q},\omega)}{\sigma_0}=\frac{8}{\omega}\int_{-\omega}^{0}d\omega'\int\frac{d^2\boldsymbol{k}}{2\pi}\sum_{s,s'=\pm}F_{ss'}(\phi)A^{s}(\boldsymbol{k},\omega')A^{s'}(\boldsymbol{k}+\boldsymbol{q},\omega'+\omega)$$

Current-Current

$$\phi=\theta_k+\theta_{k+q}$$

$$F_{ss'}(\phi)=\frac{1}{2}\left[1+ss'\frac{k\cos(2\theta)+q\cos(\theta+\theta_q)}{\sqrt{k^2+q^2+2kq\cos(\theta-\theta_q)}}\right]$$

Density-Density

$$\phi=\theta_k-\theta_{k+q}$$

$$F_{ss'}(\phi)=\frac{1}{2}\left[1+ss'\frac{k+q\cos(\theta-\theta_q)}{\sqrt{k^2+q^2+2kq\cos(\theta-\theta_q)}}\right]$$

Never Equivalent for finite q unless no-scattering

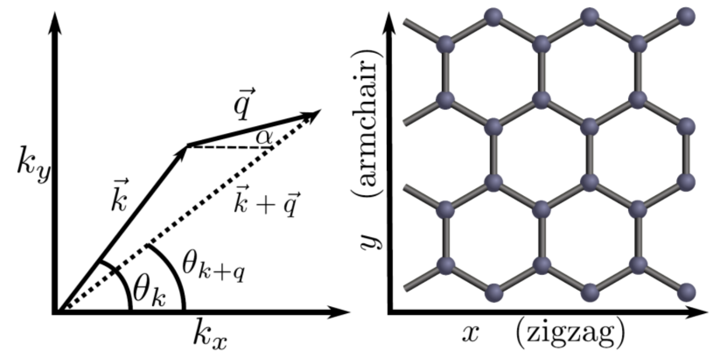
Longitudinal-Transverse

$$\sigma_{\mu\nu}(\mathbf{q},\omega) = \frac{q_\mu q_\nu}{\mathbf{q}^2}\sigma^L(\mathbf{q},\omega) + \left(\delta_{\mu\nu} - \frac{q_\mu q_\nu}{\mathbf{q}^2}\right)\sigma^T(\mathbf{q},\omega)$$

Convenient Basis

Longitudinal - Conductivity in direction of q-scattering

$$\sigma_{xx}(\mathbf{q},\omega) = \frac{q_x q_x}{\mathbf{q}^2}\sigma^L(\mathbf{q},\omega) = \sigma^L(\mathbf{q},\omega)$$



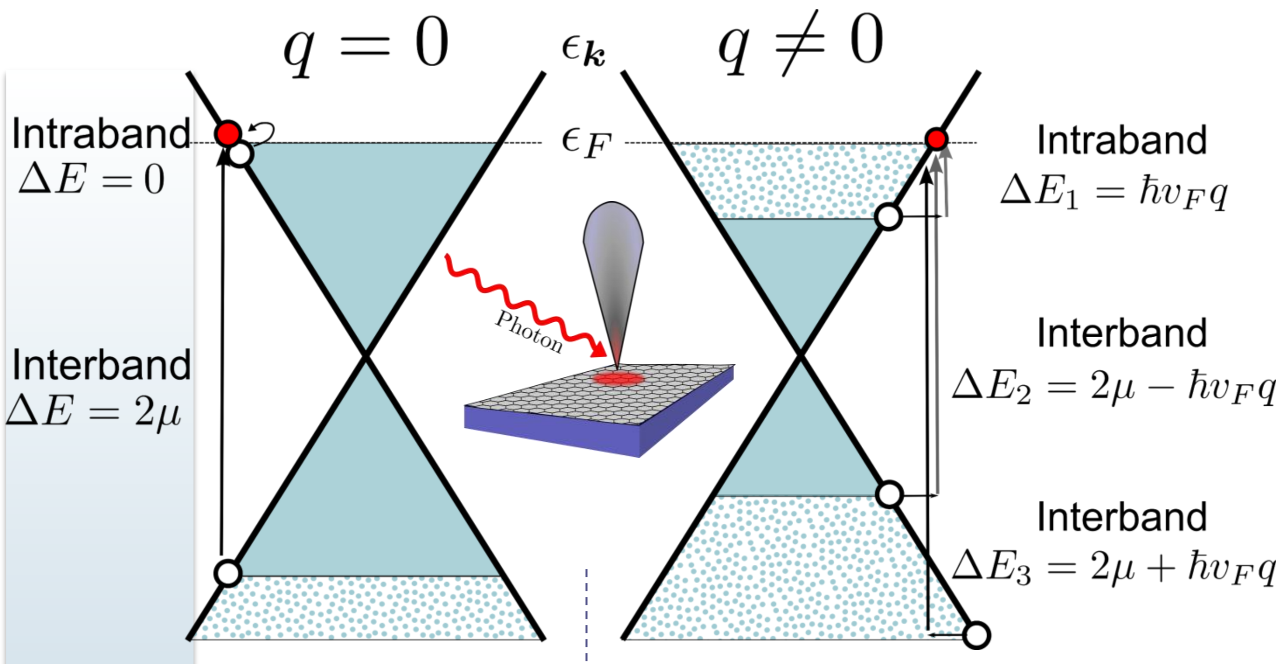
Transverse - Conductivity perpendicular to direction of q-scattering

$$\sigma_{yy}(\mathbf{q},\omega) = \sigma^T(\mathbf{q},\omega)$$

Experiment May Average Over Directions of ***q***

Focus on Strong Features (Longitudinal)

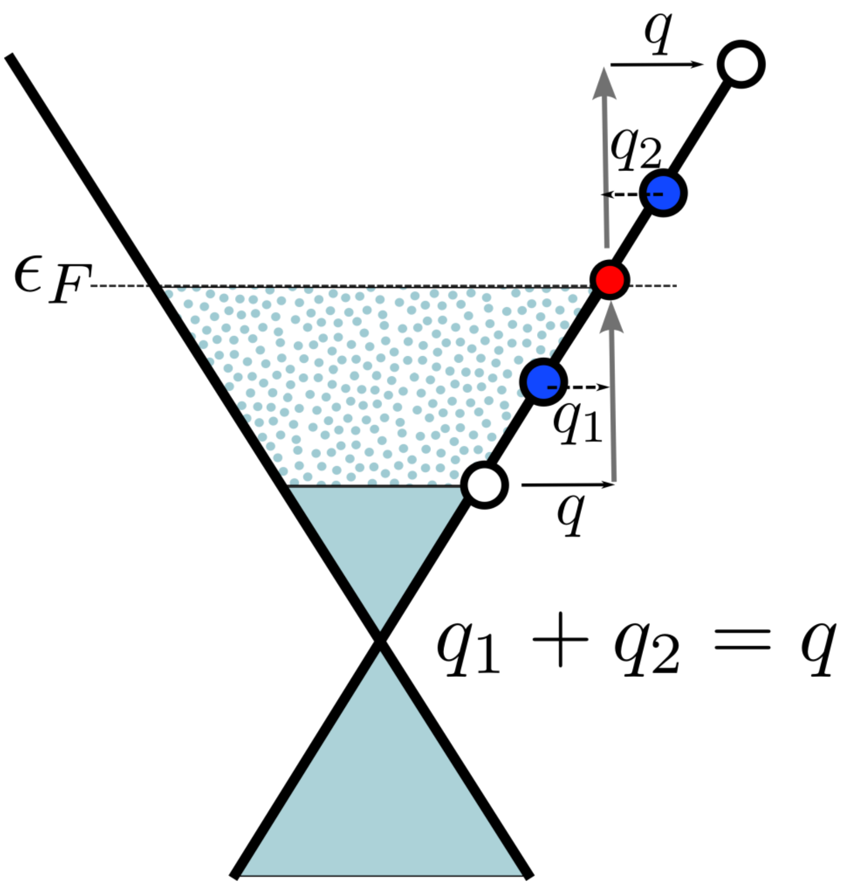
Pauli Blocking



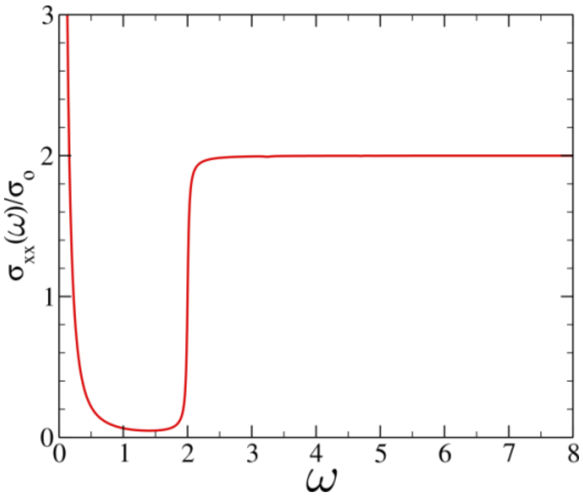
Single Optical Onset

Several Key Transition Energies

Vector Nesting



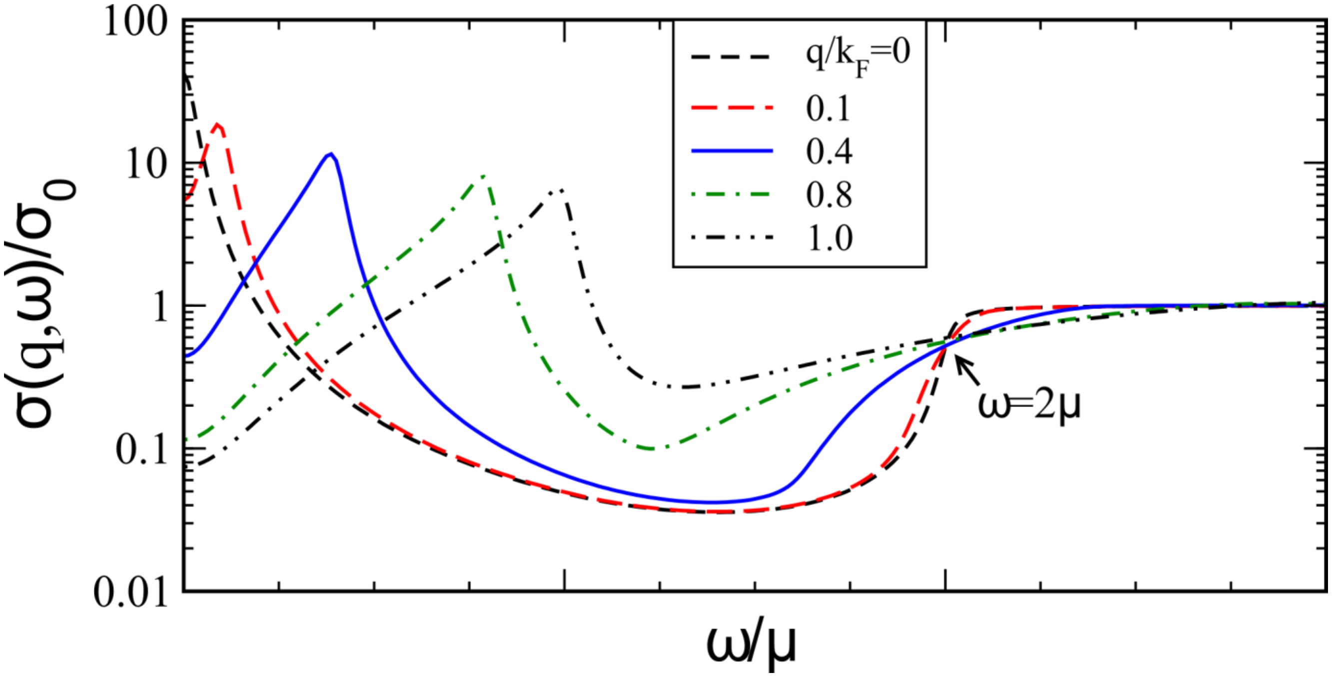
In limit of q to zero, this is Drude Response



Non-Interacting

Drude-Like Response is now shifted to

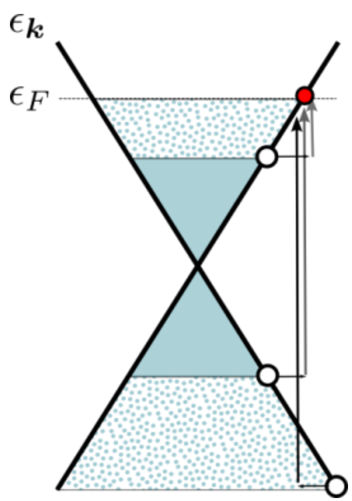
$\omega = q$

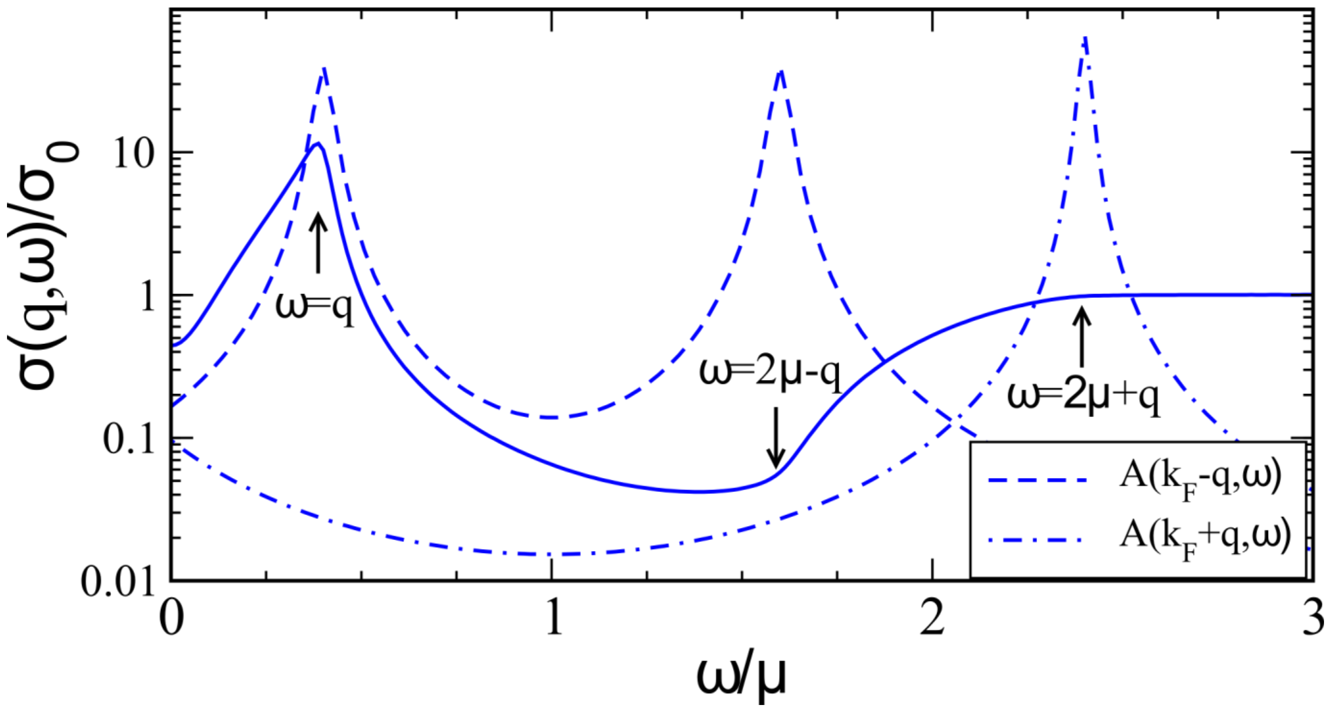


Relation to Spectral Peaks

$$\sigma(q,\omega) \Leftrightarrow A(k,\omega) \rightarrow A(k_F - q,\omega)$$

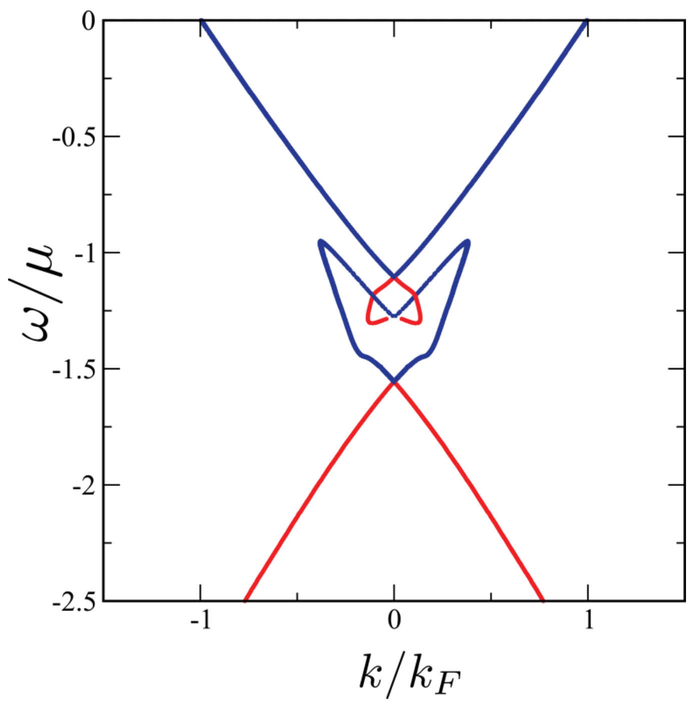
Simple Correspondence Between Spectral Peaks and Optical Peaks

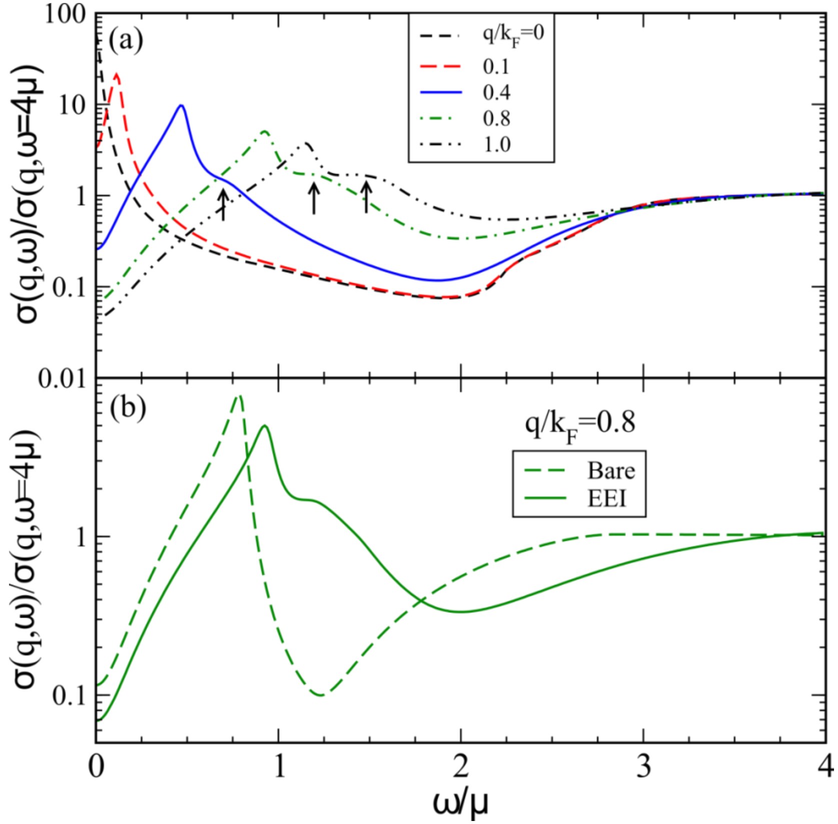




EEI Interactions

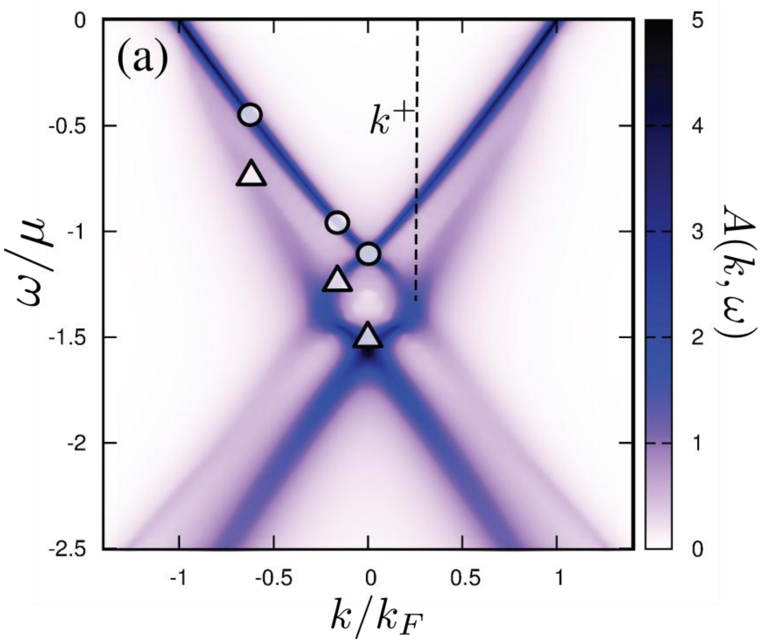
Poles in Green's Function – Peaks in $A(k,\omega)$

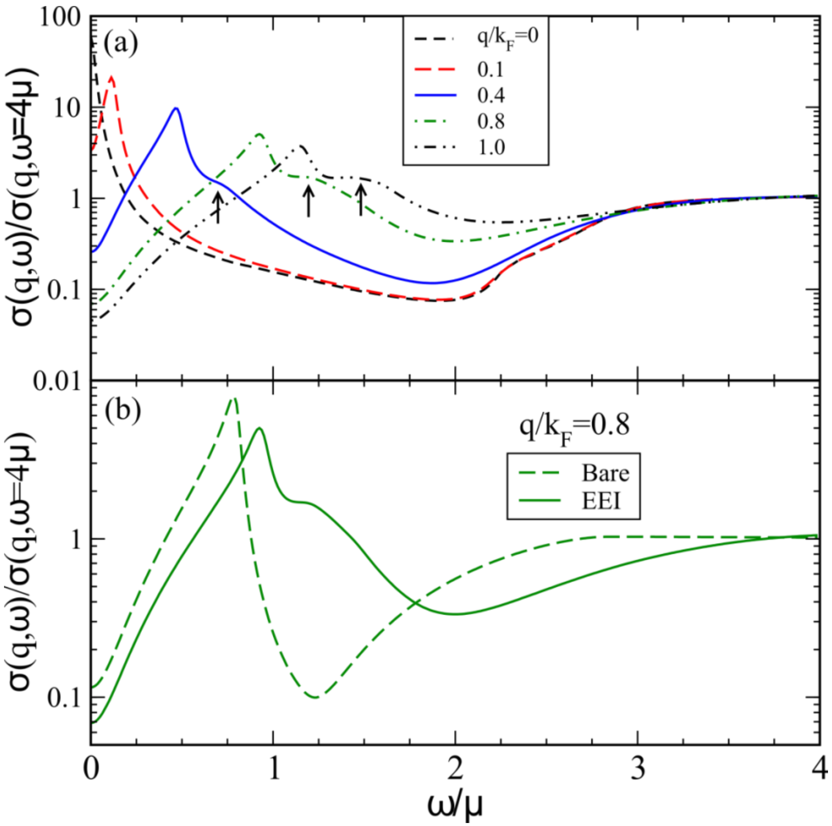




EEI Interactions

Poles in Green's Function – Peaks in A(k,ω)

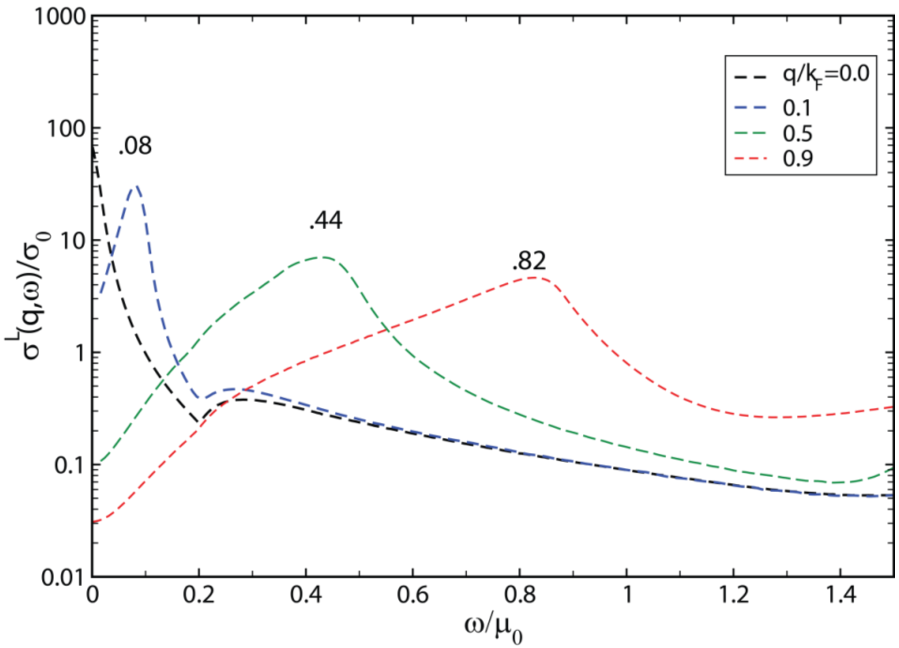


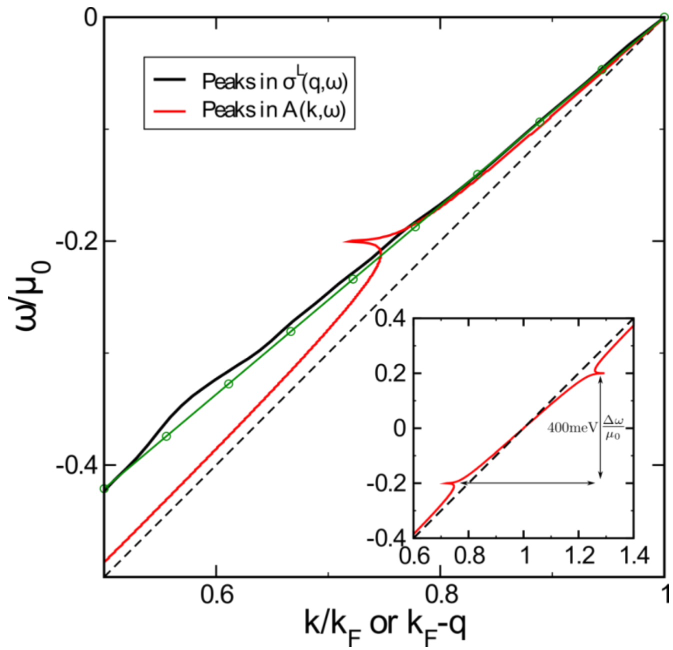


EPI - Renormalized Spectral Peaks

Electron-Phonon Interaction:

- Broadening for $\omega > \omega_E$
- Holstein sideband vanishes for $q > \omega_E$





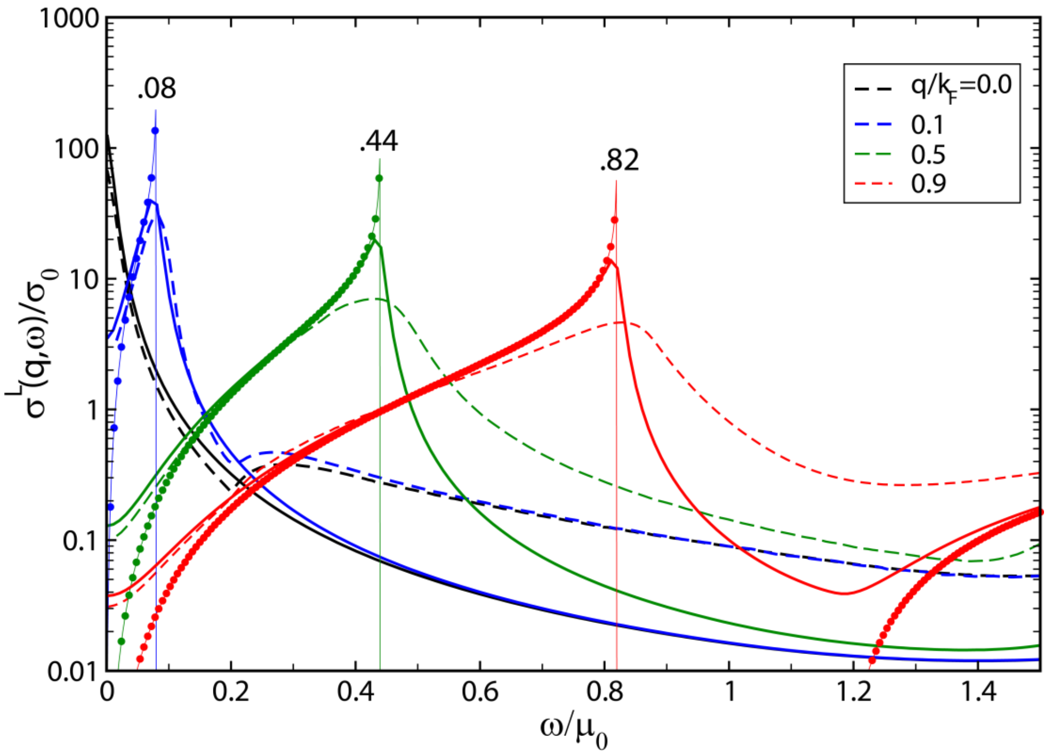
All optical peaks gain EPI renormalization factor

Analytic Result

Intraband:

$$\frac{\sigma^L(q,\omega)}{\sigma_0} = \frac{8\mu_0}{\pi}\left(\frac{\omega}{\bar{q}}\right)^2 \frac{1}{\sqrt{\bar{q}^2-\omega^2}}\frac{1}{1+\lambda}$$

$$\bar{q} = q/(1+\lambda)$$



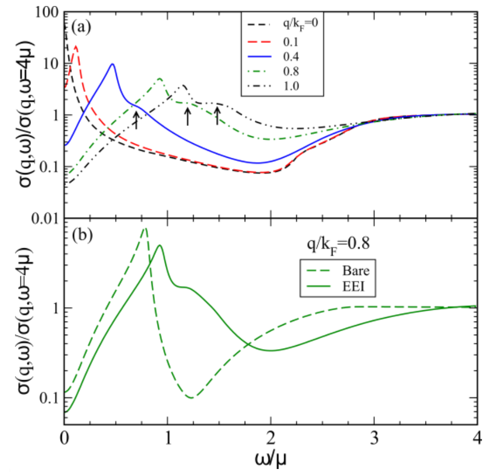
Simple Renormalization Factors Allow for Approximate Result for EPI Interaction

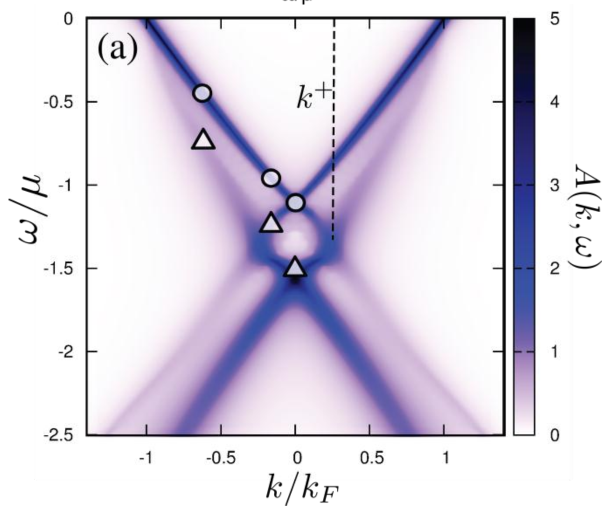
Holstein Side Band Does Not Interfere With EEI Region for Larger q

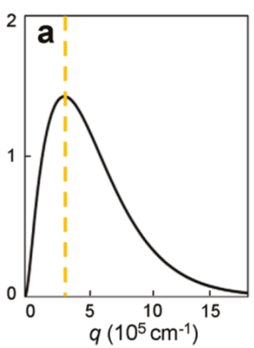
Recap

<u>Interactions on $\sigma(q,\omega)$</u>

- In the presence of scattering, the density-density and current-current correlation functions for finite momentum are not simply related
- Shown correspondence between quasiparticle spectral peaks and Intraband piece of finite q conductivity
- All Features scale with Chemical Potential
- EPI adds renormalization, λ, factors





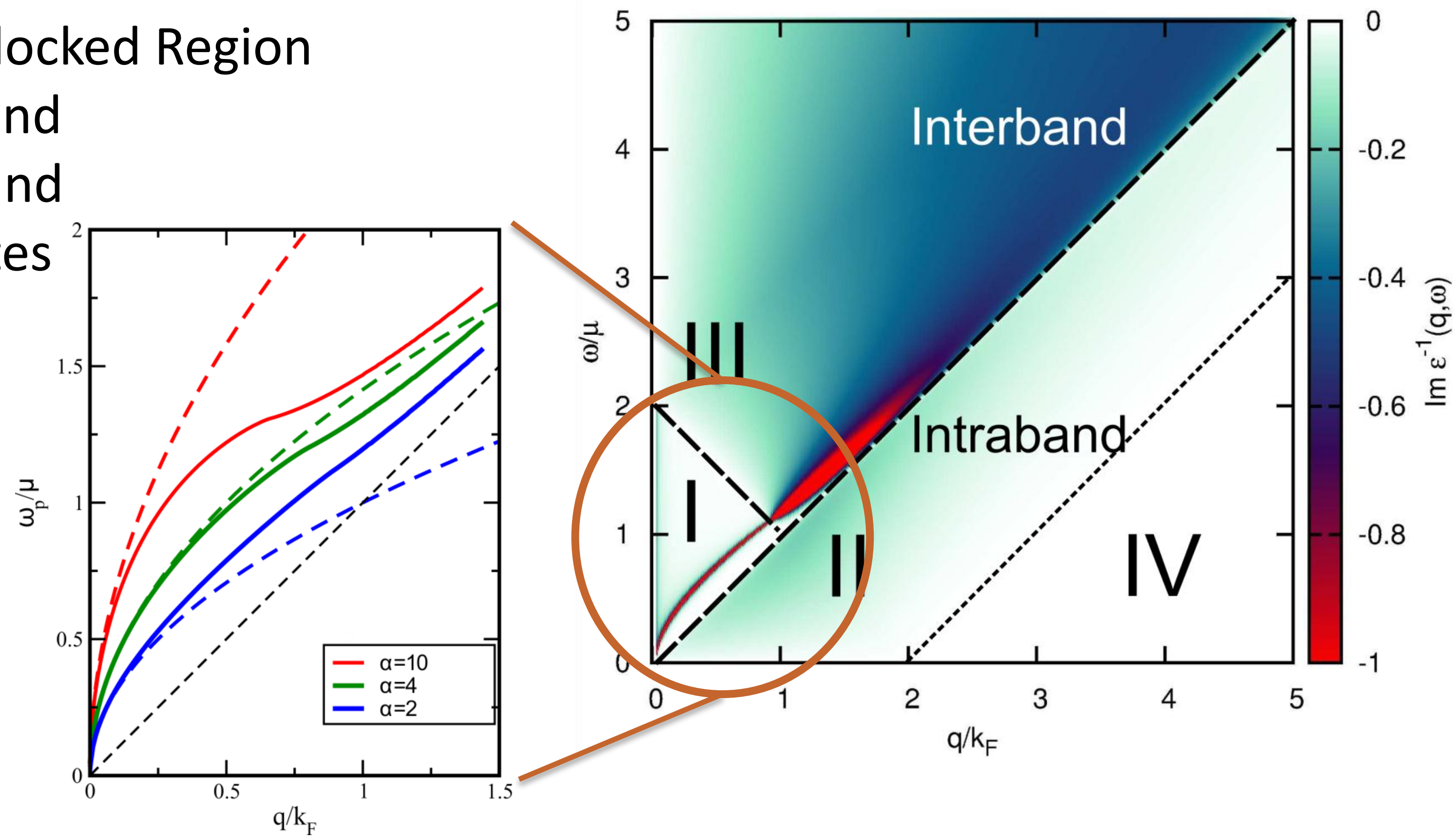


$$\frac{\sigma^L(q,\omega)}{\sigma_0} = \frac{8\mu_0}{\pi}\left(\frac{\omega}{\bar{q}}\right)^2 \frac{1}{\sqrt{\bar{q}^2-\omega^2}}\frac{1}{1+\lambda}$$

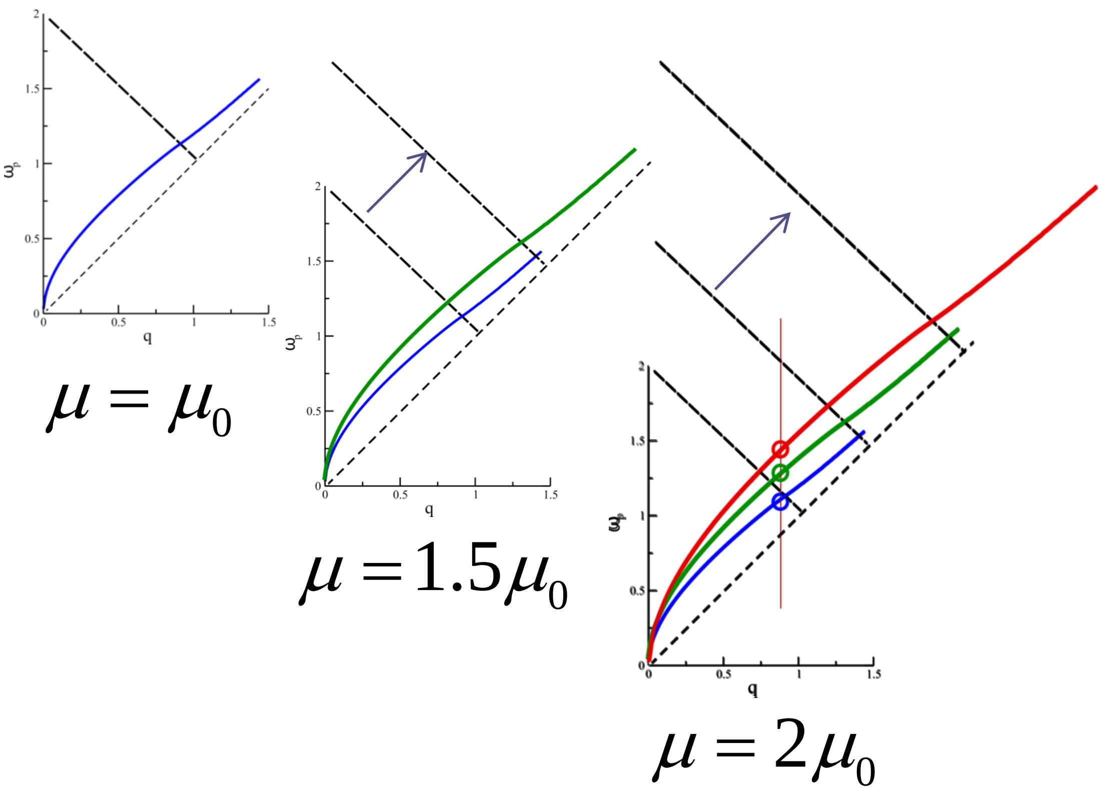
The Heart of Screening - RPA

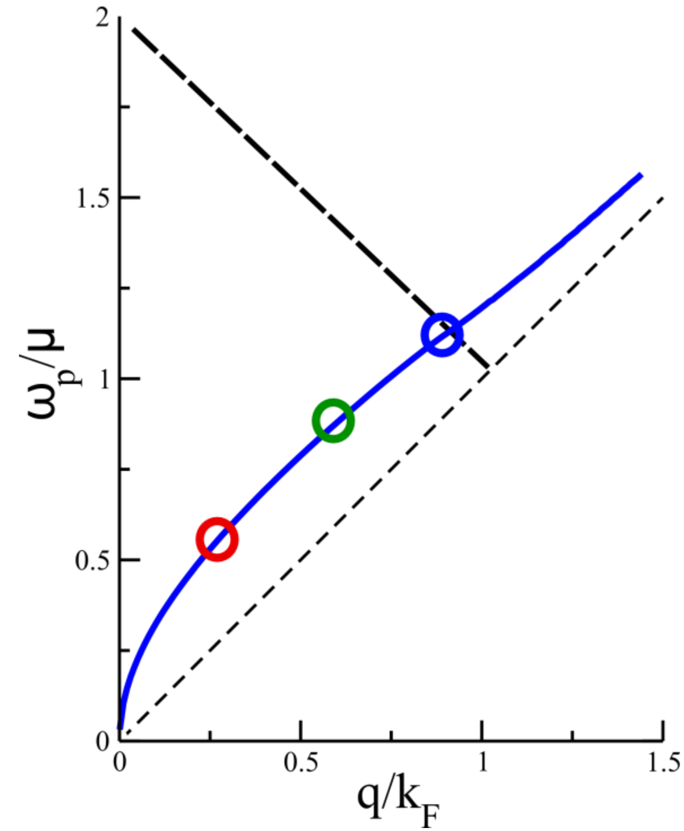
$$\varepsilon^{-1}(q,\omega) = \frac{1}{1 - V_q \Pi(q,\omega)} = \frac{q}{q - \alpha \Pi(q,\omega)}$$

I. Pauli Blocked Region
II. Intraband
III. Interband
IV. No states



Scaling with μ



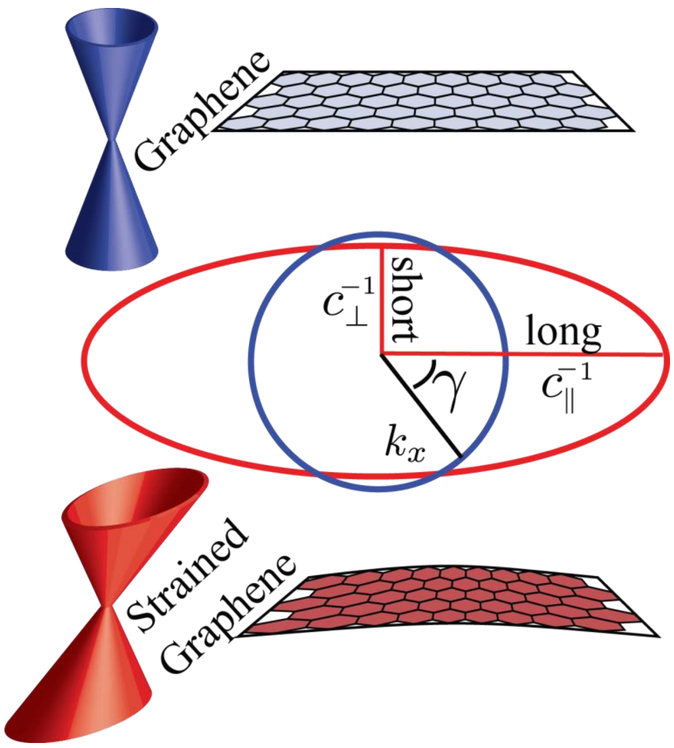


Can Exploit Scaling to provide information without varying q

What Other Tuning Mechanisms?

Strain

<u>Non-Interacting</u>
Purely Geometric Modification
To Bare Bands



$$\bar{\boldsymbol{k}} = A(\gamma)\boldsymbol{k}$$

$$A(\gamma) = R(\gamma)S(\varepsilon)R(-\gamma)$$

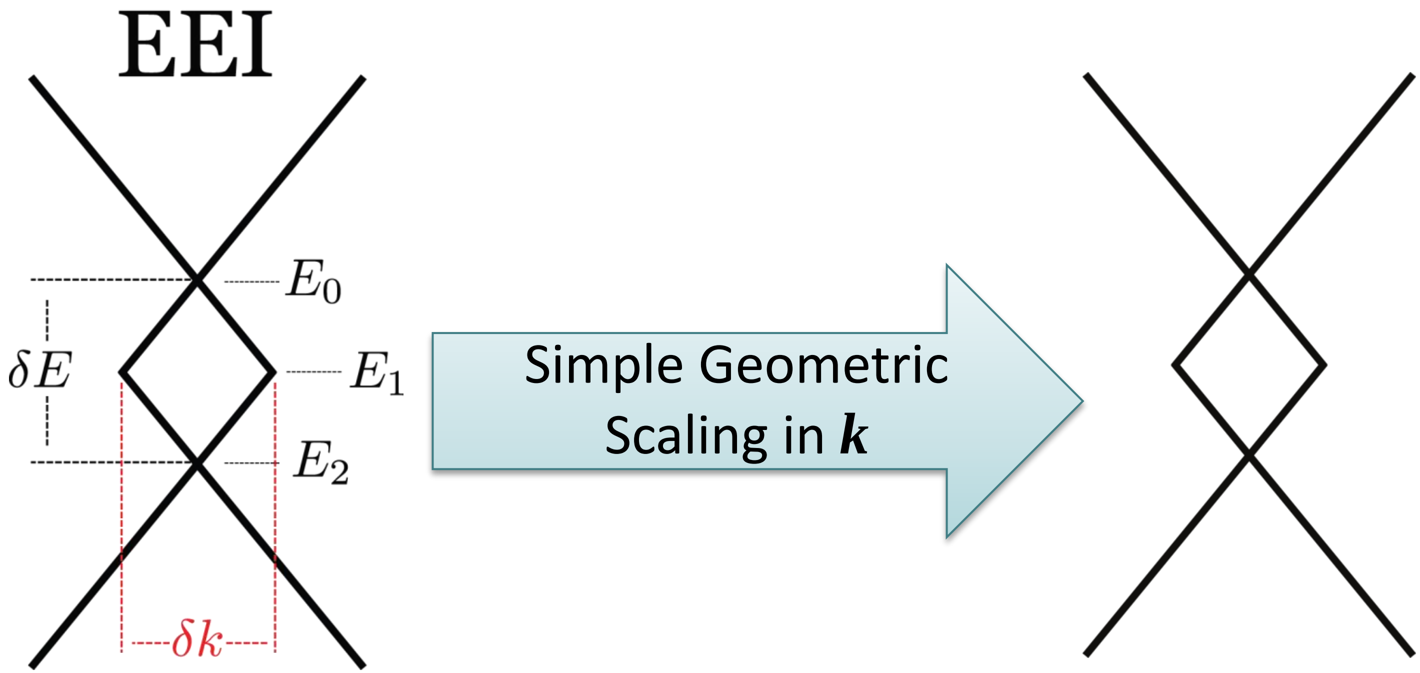
$$S(\varepsilon) = \begin{pmatrix} c_\parallel & 0 \\ 0 & c_\perp \end{pmatrix}$$

Transformation to
Maintain Linear
Dispersion

$$H = \hbar v_F \boldsymbol{\sigma} \cdot \bar{\boldsymbol{k}}$$

Strain on the Plasmaron Ring

What is the simplest thing you can do?



Not good for interacting system
We can do better than this

G_0W-RPA

Geometrically Strain the non-interacting dispersion

$$\bar{\mathbf{q}} = A(\gamma)\mathbf{q}$$

Calculate the Polarization

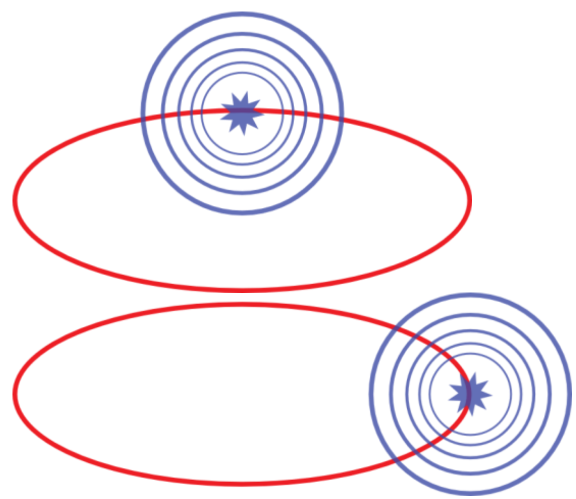
$$\Pi(q,\omega) = \left[\det S(\epsilon)\right]^{-1} \Pi^0(\bar{q},\omega)$$

Recalculate the G_0W-RPA Self Energies

$$\Sigma = \Sigma^{line} + \Sigma^{Res}$$

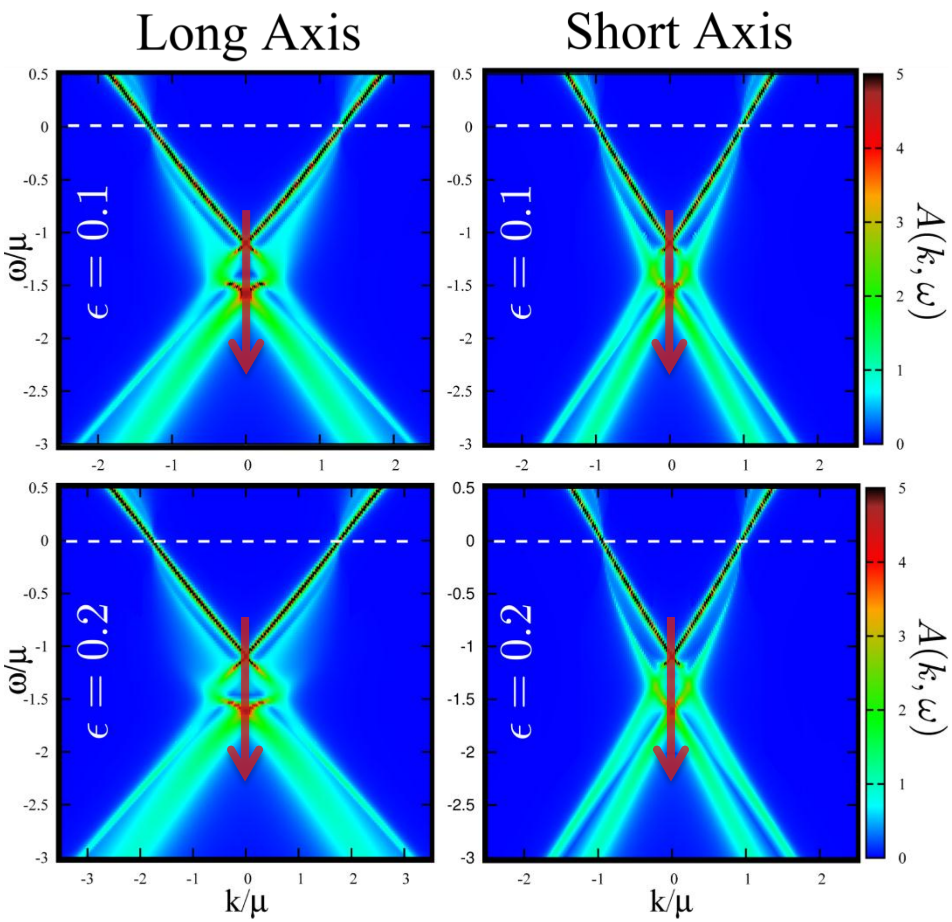
$$\Sigma_s^{RES}(\boldsymbol{k},\omega) = \sum_{s'=\pm 1} \int_0^{\infty} \int_0^{2\pi} \frac{dq d\theta_{\boldsymbol{q}}}{2\pi} \frac{\alpha}{g} \varepsilon^{-1}(q, \omega - \epsilon^{s'}_{\boldsymbol{k}+\boldsymbol{q}}) F_{ss'}(\beta_{\bar{\boldsymbol{k}}\bar{\boldsymbol{k}}'}) \left[\Theta(\omega - \epsilon^{s'}_{\boldsymbol{k}+\boldsymbol{q}}) - \Theta(-\epsilon^{s'}_{\boldsymbol{k}+\boldsymbol{q}})\right]$$

Renormalizations Depend on Direction in k-space (short vs long axis)

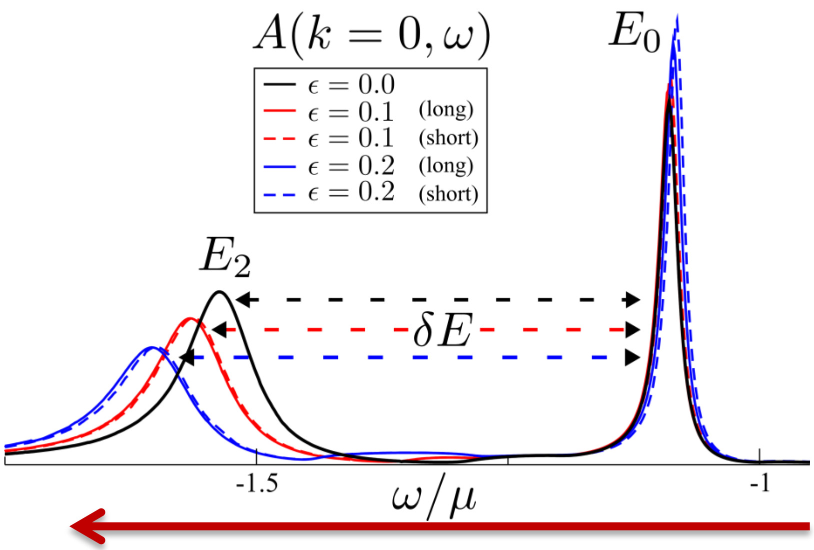


Spectral Function

Two Principle Directions

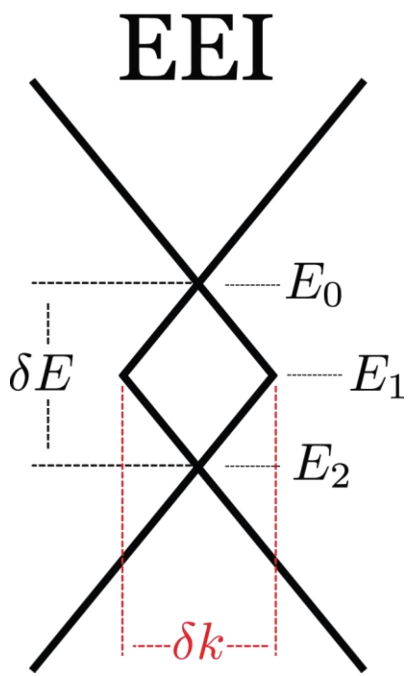


k=0 slice

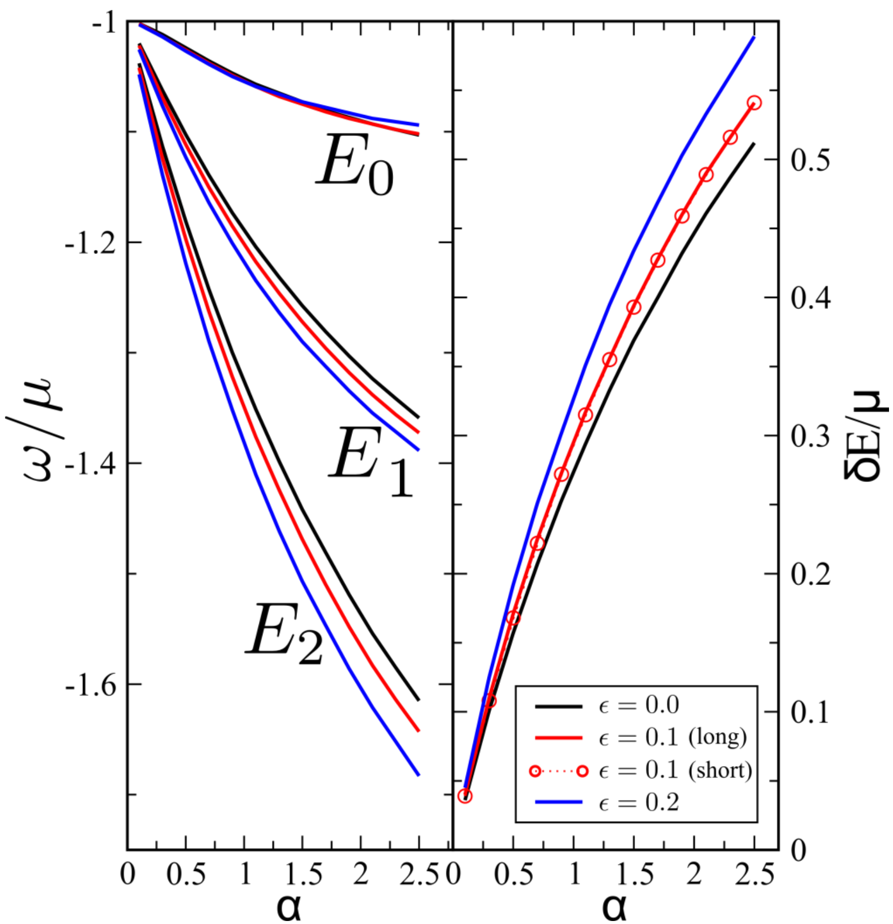


Not a Geometric Effect
Only Correlations

δE shift with correlations



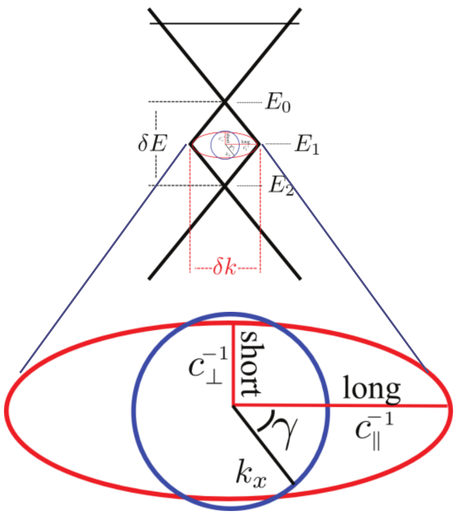
$$E_1 = \frac{E_0 + E_2}{2}$$

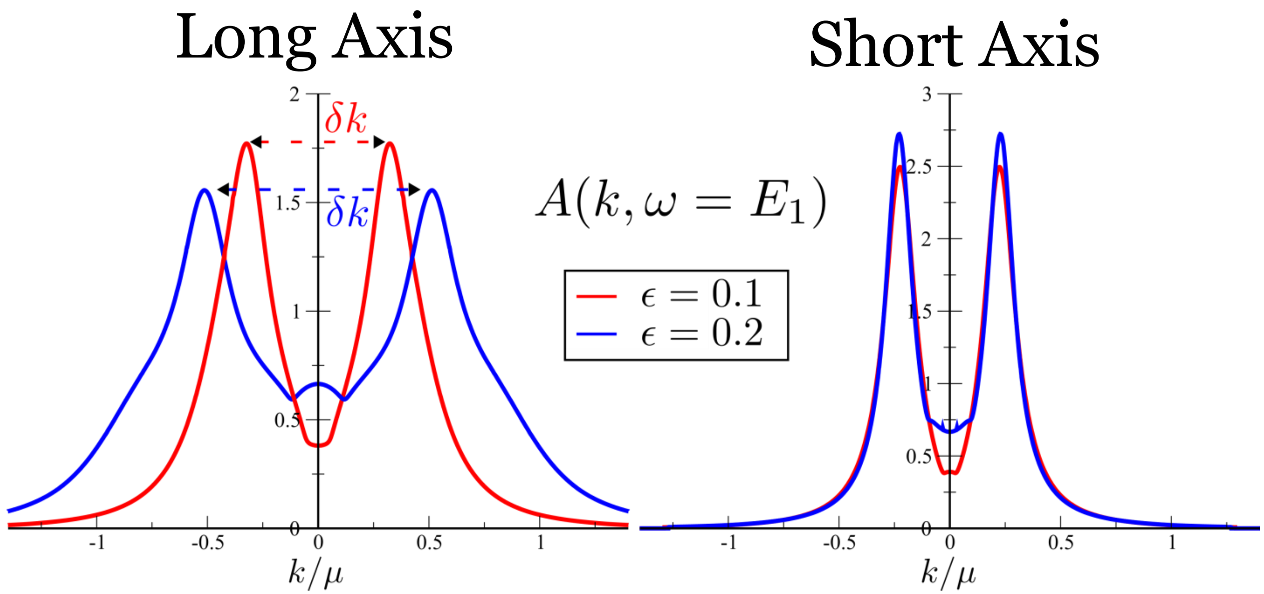


E_0 remains unchanged with Strain

Energy of Plasmaron Ring Shifts in energy

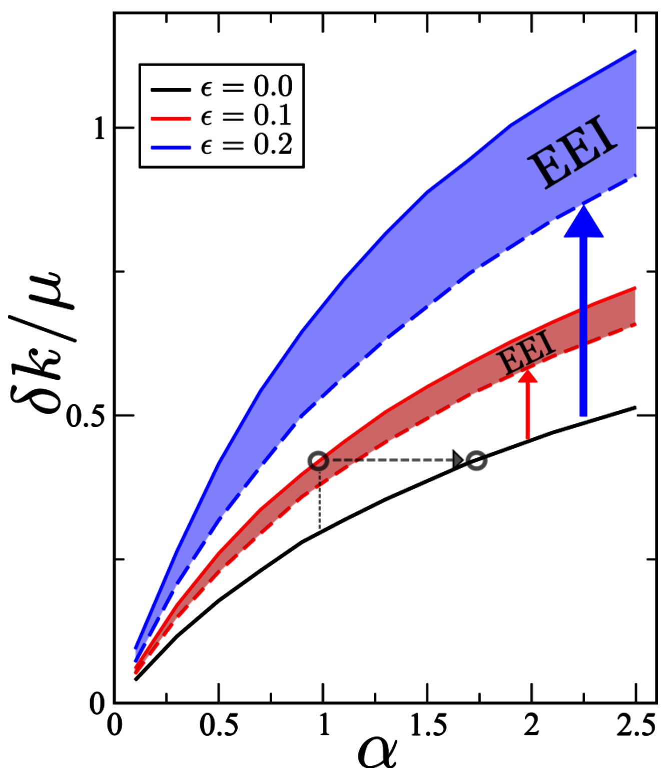
δk shift





How does δk in the long axis deviate from the purely geometric straining which one expects from non-interacting strain?

Long Axis – Variation in Coupling



Start with unstrained EEI Result
(Bostwick et al. Science 382, 999 2010)

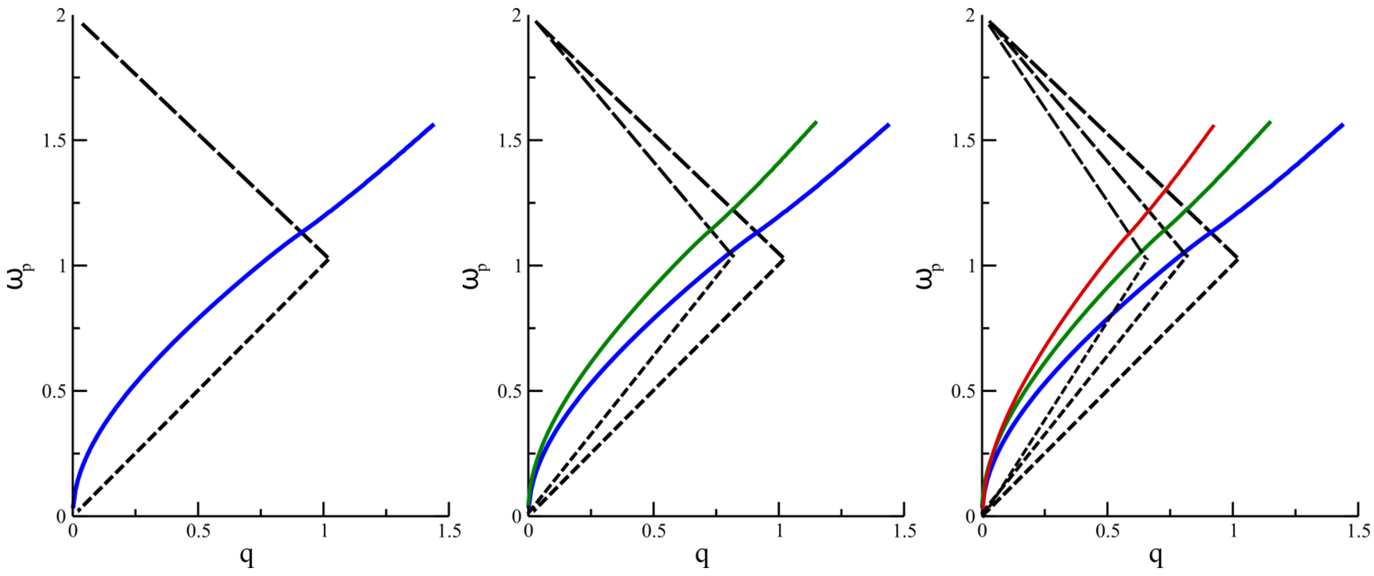
Purely Geometric Strain

Strained G_0W-RPA EEI Enhances This Effect

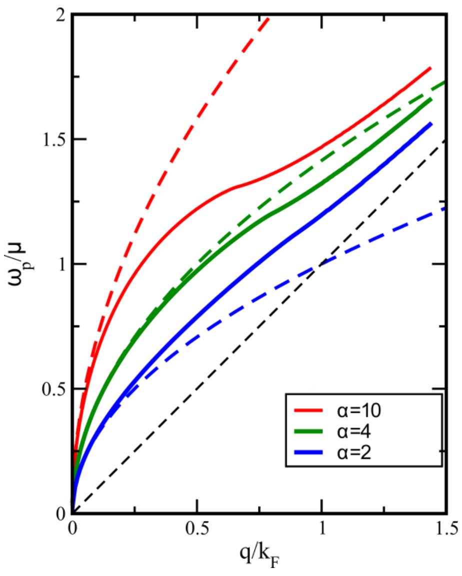
Width of Plasmaron Ring Has Increased Without Changing Substrate (α)

Simple Picture – Low vs High Frequency

Plasmon Dispersion is only Geometrically Modified

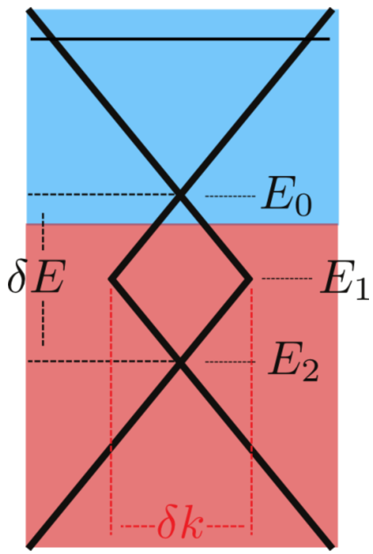


Increasing Strain



Larger α

Recap - Strain

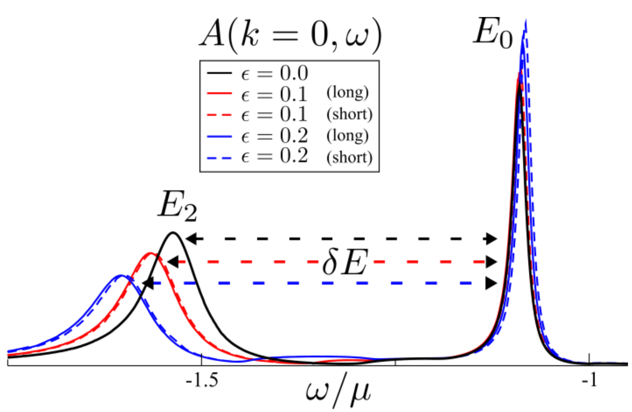


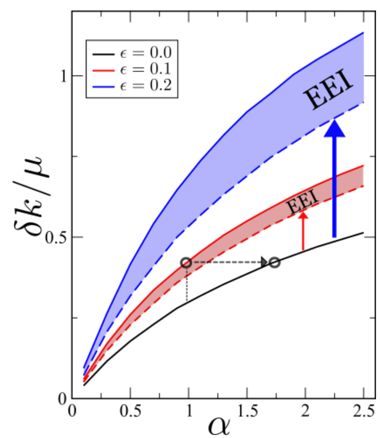
Geometrically Strained Low Frequency

- k_F points don't move due to correlations
- Dirac point at E_0 independent of strain

New Correlations at High Frequencies

- Modified Electron-Plasmon Scattering Peak ('Plasmaron ring')
  - Effectively larger α without changing substrate





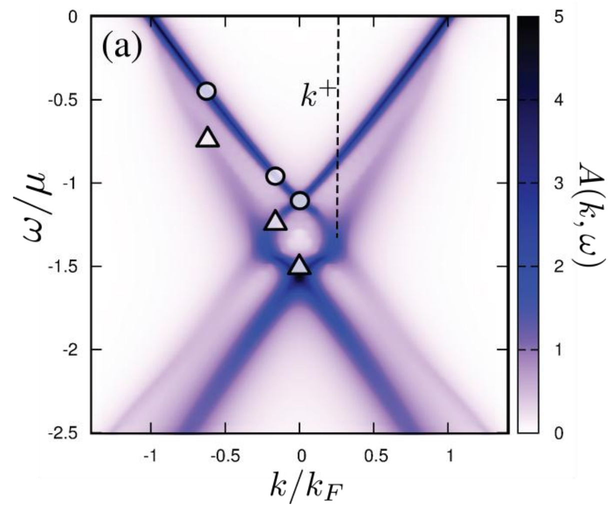
Use strain to tune features of plasmons in ARPES or optics.

Be prepared for extra correlations.

Conclusions

Shown correspondence between quasiparticle spectral peaks and Intraband piece of finite q conductivity

- All Features scale with Chemical Potential
- EPI adds renormalization, λ, factors



$$\frac{\sigma^L(q,\omega)}{\sigma_0} = \frac{8\mu_0}{\pi}\left(\frac{\omega}{\bar{q}}\right)^2 \frac{1}{\sqrt{\bar{q}^2-\omega^2}}\frac{1}{1+\lambda}$$

Application of Strain modifies G_0W-RPA

- Additional non-geometric features
- Possibility to use strain to tune electron-plasmon scattering structures.