First-principles calculation of many-body effects in graphene

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Acknowledgments

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- Prof. Sohrab Ismail-Beigi (Yale University)
- Dr. Jay Deep Sau (University of Maryland)
- Dr. Eli Rotenberg (ALS)
- Dr. Jessica L. McChesney (ALS)
- Dr. Aaron Bostwick (ALS)
- Dr. Taisuke Ohta (ALS)
- Dr. Euyheon Hwang (University of Maryland)
- Dr. Emmanouil Kioupakis (UC Santa Barbara)
- Cohen Louie group members

Financial support: NSF and DOE
Computational resources: TeraGrid and NERSC
- Preliminaries
  - Graphene
  - Electron self energy / ARPES
  - Migdal approximation
  - \( GW \) approximation

- Electron self energy in graphene
  - Contribution from electron-phonon (e-ph) interactions
  - Contribution from electron-electron (e-e) interactions
  - Combining the effects of e-ph and e-e interactions
Preliminaries
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Charge carriers in graphene are 2D massless Dirac fermions

Figure: Geim and McDonald, Phys. Today (2008)
Klein tunneling in graphene

The most spectacular phenomenon of 2D massless Dirac fermions

Isolation of single-layer graphene and observation of new integer quantum Hall effect

A. K. Geim Group at Manchester

P. Kim Group at Columbia
Epitaxial growth of graphene layers on SiC

Thermal decomposition of SiC at high temperature (>1400°C)

Sublimation of Si


By controlling temperature, growth of 1 to ~100 graphene layers (tens of microns)

(Slide courtesy of Walter de Heer)
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Electron self energy due to many-body effects

Many-body effects:
- e-e interactions (e-h pair generation, plasmon emission)
- e-ph interactions
- impurity scattering
- ...

\[ \varepsilon_k = \varepsilon_k^0 + \text{Re} \Sigma_k \]  
Quasiparticle energy

\[ \tau_k = \frac{\hbar}{2 \text{Im} \Sigma_k} \]  
Quasiparticle lifetime
Angle-resolved photoemission spectroscopy (ARPES)

Angle-resolved photoemission spectroscopy (ARPES)

\[ I(k, \omega, \epsilon', h\nu) = I_0(k, \omega, \epsilon', h\nu) f_{FD}(\omega) A(k, \omega) \]

\[ A(k, \omega) = \frac{2}{\pi} \frac{- \text{Im} \Sigma(k, \omega)}{[\omega - \epsilon_k - \text{Re} \Sigma(k, \omega)]^2 + [\text{Im} \Sigma(k, \omega)]^2} \]

ARPES \rightarrow electron self energy
Width of the momentum distribution curve (MDC)

$$\varepsilon_k = \hbar v_F k \quad \text{(graphene or doped cuprates)}$$

$$A(k, \omega) = \frac{2}{\pi} \frac{-\text{Im} \Sigma(k, \omega)}{\left[ \omega - \hbar v_k k - \text{Re} \Sigma(k, \omega) \right]^2 + [\text{Im} \Sigma(k, \omega)]^2}$$

$$\approx \frac{2}{\pi} \frac{-\text{Im} \Sigma(\omega)}{\left[ \omega - \hbar v_k k - \text{Re} \Sigma(\omega) \right]^2 + [\text{Im} \Sigma(\omega)]^2}$$

Lorentzian of $k$

$$\Delta k(\omega) = \text{FWHM} \approx 2 \times \frac{|\text{Im} \Sigma(\omega)|}{\hbar v_F}$$

Imaginary part of the self energy is proportional to the MDC width in graphene, to a good approximation.
Angle-resolved photoemission spectroscopy (ARPES)

- Energy distribution curve (EDC)
  - Quasiparticle bandstructure
  - Real part of the self energy

- Momentum distribution curve (MDC)
  - Width gives lifetime
  - Imaginary part of the self energy

Motivation

ARPES of epitaxial graphene on (0001) surface of SiC

1) A. Bostwick, et. al., Nature Physics 3, 36 (2007)
Motivation

Motivation

Substrate induced band gap

Important issue for applications

Interpretation of the ARPES spectra of graphene

Q: Self-energy effects$^1$ or substrate induced band gap$^2$?

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Migdal approximation

\[ \Sigma^{e-ph} = i g^2 \, D \, G_0 \]

Electron self energy arising from e-ph interactions
Electron self energy arising from e-ph interactions

• Step 1. DFT wavefunctions (planewave basis) and energy eigenvalues within LDA using \textit{ab initio} pseudopotentials.\textsuperscript{1}

\[
\left[-\frac{\nabla^2}{2} + V_{\text{ion}} + V_{\text{Hartree}} + V_{\text{xc}}^{\text{LDA}}\right] \psi_{nk} = E_{nk}^{\text{LDA}} \psi_{nk}
\]

• Step 2. Phonon eigenmodes / frequencies / electron-phonon coupling within DFPT \textsuperscript{2}

\[
g_{n,n',\lambda} (k, q) = \sqrt{\frac{\hbar}{2M\omega_{\lambda,q}}} \langle n,k | \Delta V_{\text{SCF}} (\lambda, q) | n', k + q \rangle
\]

Electron self energy arising from e-ph interactions

- **Step 3.** Interpolation of the above quantities for q points in a fine grid (500 x 500 irreducible points) using maximally localized Wannier functions.\textsuperscript{1-3}

- **Step 4.** Self-energy of electron within Migdal formalism.\textsuperscript{4}

\[
\Sigma_n(k, E) = \sum_{n', \lambda} \int \frac{d^2 q}{A_{BZ}} g_{n,n',\lambda}(k,q)^2 \left[ \frac{n_B(\omega_{\lambda,q}) + 1 - n_F(\xi_{n',k+q})}{E - \xi_{n',k+q} - \omega_{\lambda,q} + i\delta} + \frac{n_B(\omega_{\lambda,q}) + n_F(\xi_{n',k+q})}{E - \xi_{n',k+q} + \omega_{\lambda,q} + i\delta} \right]
\]

4) J.R. Schrieffer, Theory of Superconductivity (W.A.Benjamin,Inc., 1964)
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Electron self energy arising from e-e interactions

\[ \Sigma^{e-e} = i G_0 W_0 \]
Electron self energy arising from e-e interactions

- **Step 1. Ground state calculation**
  - DFT wavefunctions and eigenvalues within **LDA** using *ab initio* pseudopotentials. \(^1\)

\[
\left[ -\frac{\nabla^2}{2} + V_{\text{ion}} + V_{\text{Hartree}} + V_{xc}^{\text{LDA}} \right] \psi_{nk} = E_{nk}^{\text{LDA}} \psi_{nk}
\]

- **Step 2. QP self-energy within **GW** Approximation \(^2\)

\[
\left[ -\frac{\nabla^2}{2} + V_{\text{ion}} + V_{\text{Hartree}} + \sum(E_{nk}) \right] \psi_{nk} = E_{nk} \psi_{nk} \quad (\sum = iG_0W_0)
\]

Electron self energy arising from e-e interactions

- *GW* calculations performed with BerkeleyGW code\textsuperscript{1}.
- Full dynamical dielectric screening of graphene was calculated\textsuperscript{2}.
- Substrate screening was modeled by\textsuperscript{3-4}

\[
\varepsilon_q(\omega) = \varepsilon_{\text{eff}} - \nu_q P_q(\omega)
\]

\[
\varepsilon_{\text{eff}} = \frac{(1 + \varepsilon_{\text{sub}}^\infty)}{2}
\]

- \(\nu_q\) bare Coulomb interaction
- \(P_q(\omega)\) polarizability of graphene

1) J. R. Deslippe et al., computer code BerkeleyGW (2009)
4) M. Polini et al., PRB 77, 081411 (2008)
Outline

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Imaginary part of the electron self energy (e-ph interactions)

Conventional metal:

$$\text{Im} \Sigma^{e-ph}(\varepsilon)$$

1. Constant DOS near the Fermi level
2. One effective optical phonon branch (Einstein phonon spectrum)
Imaginary part of the electron self energy (e-ph interactions) reflects linear density of states in graphene. Highest optical phonon branches (~ 0.2 eV) are dominant.

Park, Giustino, Cohen, and Louie, PRL (2007)
The real part gives rise to 4%~8% reduction in the fermi velocity.

Park, Giustino, Cohen, and Louie, PRL (2007)
Comparison with experiment (MDC width)


Park, Giustino, Cohen, and Louie, PRL (2007)
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Imaginary part of the self energy (decay rate)

Decay rate of a hole via e-e interactions?
Electron energy loss function

\[
\text{Im} \left[ \frac{1}{\varepsilon(q, \omega)} \right]
\]

is non-zero when

\[
\text{Im} \varepsilon(q, \omega) \neq 0 \quad \text{Electron-hole pair generation}
\]

\[
\begin{cases}
\text{Re} \varepsilon(q, \omega) = 0 \\
\text{Im} \varepsilon(q, \omega) = 0 \\
\end{cases}
\quad \text{Plasmon emission}
\]
Energy loss function of $n$-doped graphene

$$-\text{Im} \varepsilon(q, \omega)$$

$$\text{Im} \left[ \frac{1}{\varepsilon(q, \omega)} \right]$$

Figure taken from M. Polini et al., PRB 77, 081411 (2008)
Plasmons (oscillation w/o external field) in 3D & 2D

\[ \omega_{\text{plasmon}} (q = 0) \]

3D: finite restoring force

\[ \omega^{3D}_{\text{plasmon}} (q = 0) \neq 0 \]

2D: zero restoring force

\[ \omega^{2D}_{\text{plasmon}} (q = 0) = 0 \]
Imaginary part of the self energy (decay rate)

Decay rate of a hole via e-e interactions?

\[ \text{Im} \left[ \frac{1}{\varepsilon(q, \omega)} \right] \]
• In $n$-doped graphene, a hole above the Dirac point energy do not decay by plasmon emission.
• In \( n \)-doped graphene, a hole below the Dirac point energy can decay by plasmon emission (transition into upper band).
• Transition into lower band does not involve plasmons.
An optimal energy exists for transitions into upper band having the largest plasmon contribution.
Imaginary part of the electron self energy (e-e interactions)

\[ \text{Im} \Sigma_{e-k}^{e-e} (\varepsilon_k) \]

- Total decay rate
- Decay into upper band
- Decay into lower band

In agreement with previous argument
Strong anisotropy: van Hove singularity

Park, Giustino, Spataru, Cohen, and Louie, PRL (2009)
Substrate dielectric screening reduces the decay rate. Also, the plasmon induced peak position is shifted.

Park, Giustino, Spataru, Cohen, and Louie, PRL (2009)
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Electron self energy arising from e-e and e-ph interactions

\[ \text{Im} \Sigma_k (\varepsilon_k) \]

The two interactions are comparable near the Fermi energy.

Park, Giustino, Spataru, Cohen, and Louie, PRL (2009)
Reasonable agreement between theory (substrate screening) and experiment.
Lower than measurements near the Dirac point energy: midgap states?

Park, Giustino, Spataru, Cohen, and Louie, PRL (2009)
Q: Self-energy effects\textsuperscript{1} or substrate induced band gap\textsuperscript{2}?  

A: Both are important.

First-principles calculations

\[ D \]
\[ G_0 \]
\[ g \]
\[ g \]

\[ W_0 \]
\[ G_0 \]

→ Insights to interpret ARPES spectra of graphene