Probing Electron-Phonon Interactions at the Saddle Point in Graphene

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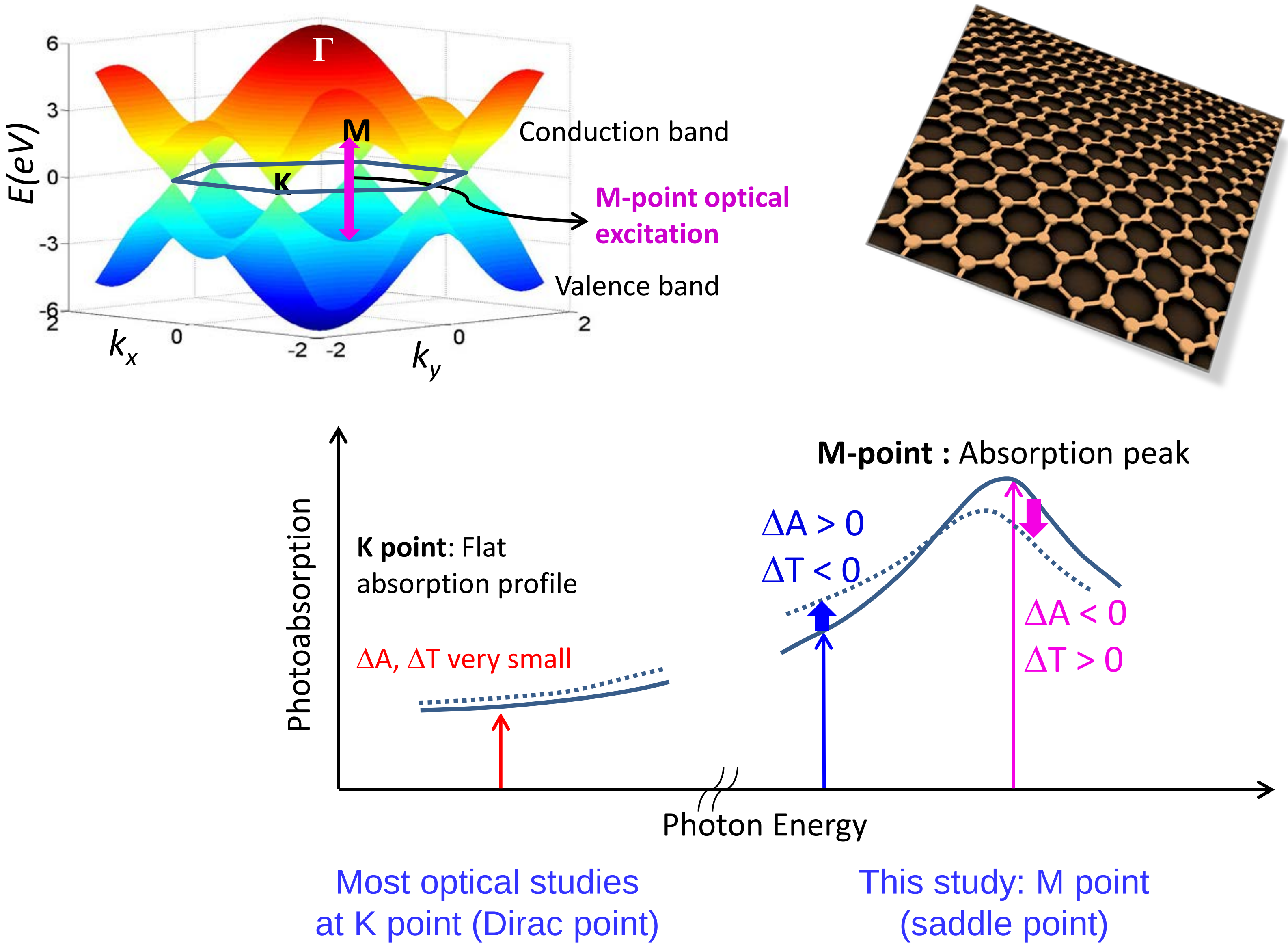
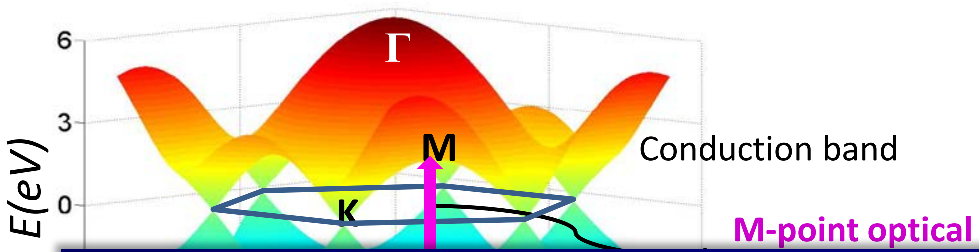
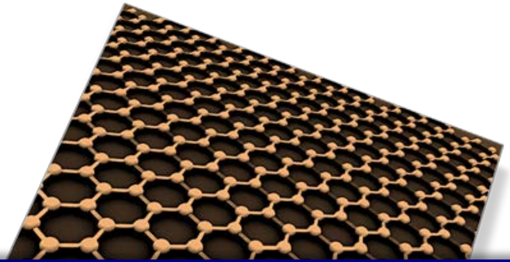


Γ
M
K
E(eV)
6
3
0
-3
-6
2
0
-2
-2
0
2
k_x
k_y
Conduction band
M-point optical excitation
Valence band
M-point : Absorption peak
K point: Flat absorption profile
ΔA, ΔT very small
ΔA > 0
ΔT < 0
ΔA < 0
ΔT > 0
Photoabsorption
Photon Energy
Most optical studies at K point (Dirac point)
This study: M point (saddle point)





Pump-probe spectroscopy at M-point:

- Alternative to well-studied K-point
- Observe many-body effects with sensitive absorption peak renormalization
-
- ...onons

Why interest in acoustic phonons?

Effective acoustic deformation potential

$D^{ac}_{eff} \sim 2.8 - 7\ \mathrm{eV}$ theory

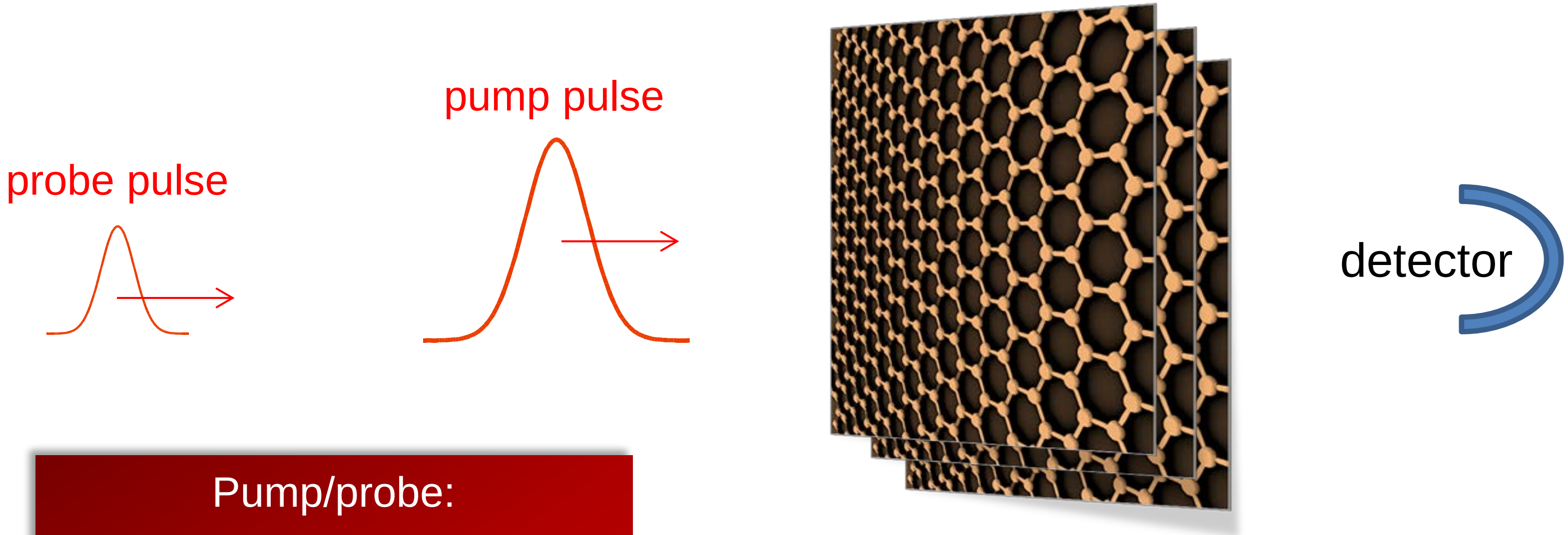
$D^{ac}_{eff} \sim 16 - 50\ \mathrm{eV}$ experiment, mostly electrical transport

Most observables $\sim |D^{ac}_{eff}|^2$

Uncertainty in observables >270

Has implications for optical and electrical transport applications

Deformation potential = fundamental material parameter, determines specific heat, mean free path etc.



Pump/probe:

~100fs duration

up to 500ps delay

degenerate at 1.6, 3.2, 4.8eV

non-degenerate at M-point (probe scanned)

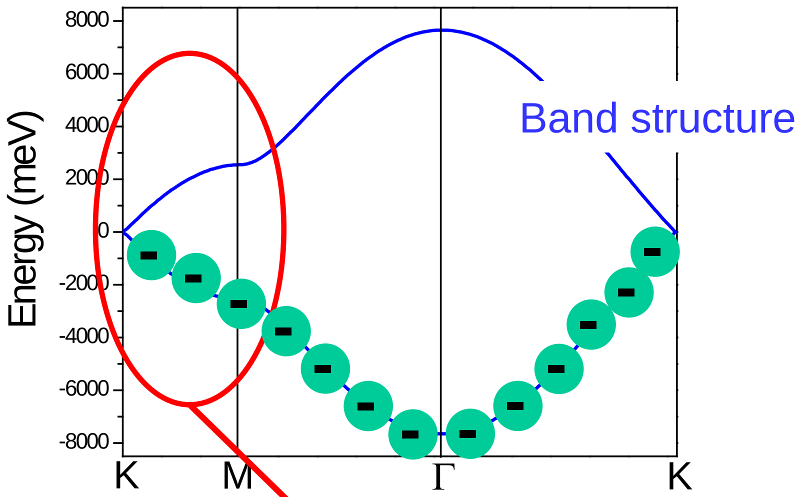
Graphene sample:

CVD-grown layers

10 individually grown layers stacked onto sapphire substrate

Stacking does not influence monolayer behavior (verified through Raman spectroscopy)

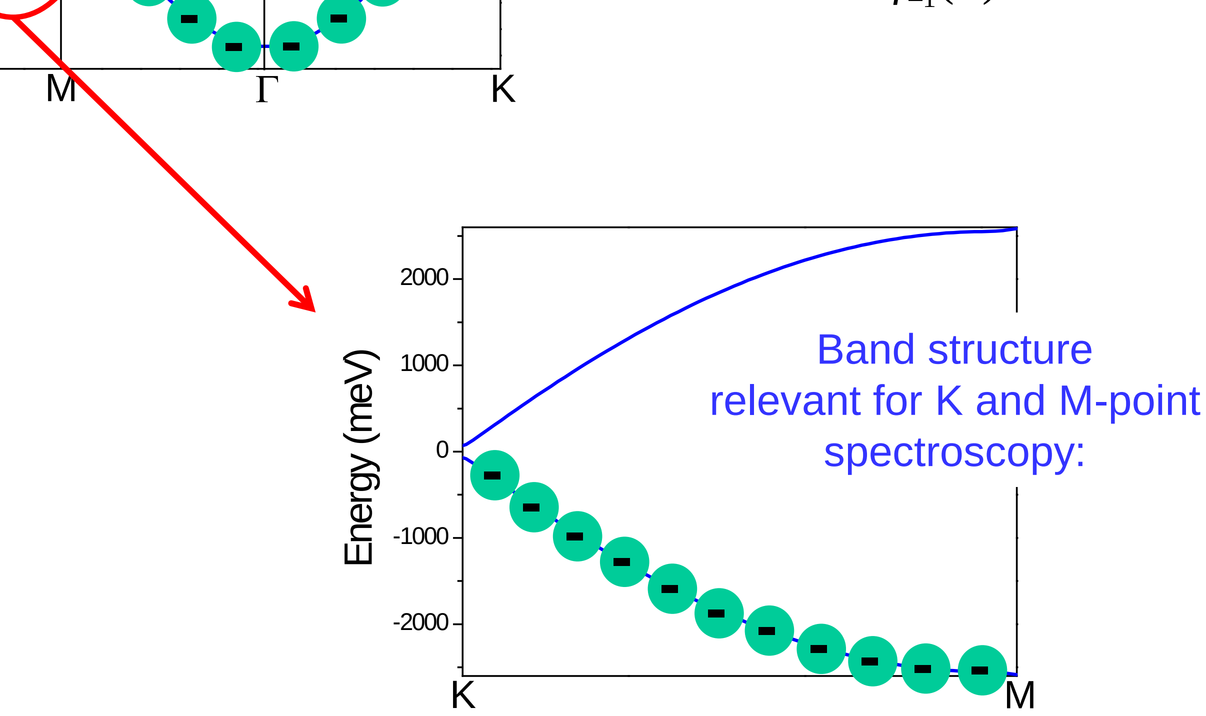
Saddle-point (M-point) spectroscopy



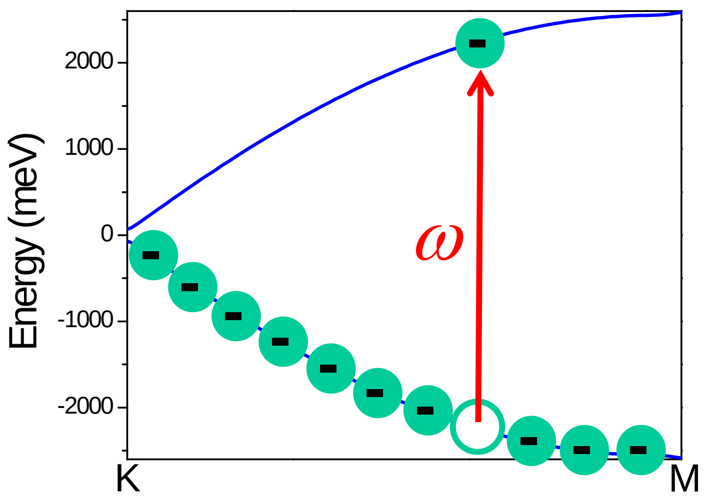
occupation at T=0K

$$f_{+1}(\boldsymbol{k}) = 0$$

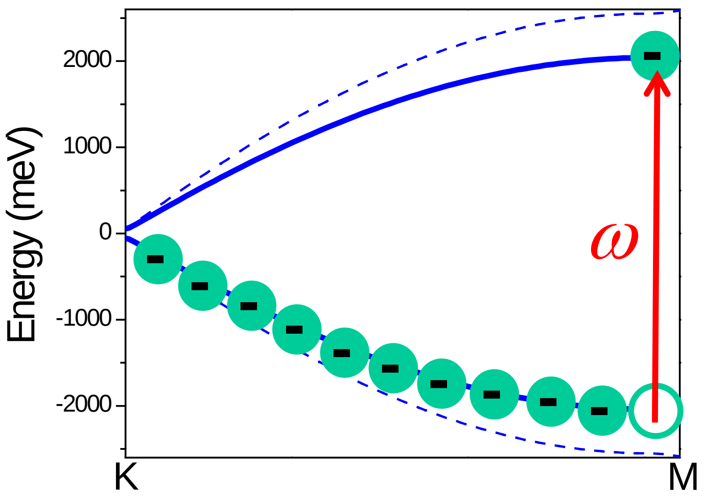
$$f_{-1}(\boldsymbol{k}) = 1$$



Absorption in thermal equilibrium



Absorption in excited system (bands shifted and more broadened)



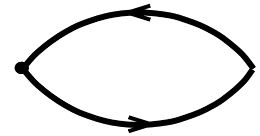
Susceptibility: $\chi(\omega)\big|_{\text{equil}}$ → Transmission: T_0

$\chi(\omega)\big|_{\text{excited}}$ → T

Differential Transmission:

$$\frac{T - T_0}{T_0}$$

Optical spectra



$\chi(\omega)$

$$\chi(\omega) = -\frac{2e^2}{m^2\omega^2} 2 \int_{BZ} \frac{d^2k}{(2\pi)^2} \langle [\boldsymbol{p}(\boldsymbol{k})\cdot\hat{\boldsymbol{e}}]^2 \rangle_\theta \frac{f_{-1}(\boldsymbol{k}) - f_1(\boldsymbol{k})}{\hbar\omega - \Delta E(\boldsymbol{k}) + i\gamma(\omega,\boldsymbol{k})} L_F(\boldsymbol{k})$$

average over angles between flake orientation and opt. polarization

phenomenological Fano shape factor (exciton effect)

- Transition energies:

$$\Delta E(\boldsymbol{k}) = E_1(\boldsymbol{k}) - E_{-1}(\boldsymbol{k}) + \mathrm{Re}\,\Delta\Sigma_1(\boldsymbol{k}) - \mathrm{Re}\,\Delta\Sigma_{-1}(\boldsymbol{k})$$

$$\gamma(\omega,\boldsymbol{k}) = \gamma_0(\omega) - \mathrm{Im}\,\Delta\Sigma_1(\boldsymbol{k}) - \mathrm{Im}\,\Delta\Sigma_{-1}(\boldsymbol{k})$$

e-ph scattering w/o excitation (adjusted to match exp. linear absorption)

pump-induced changes

- Transmission calculated from optical transfer matrix approach

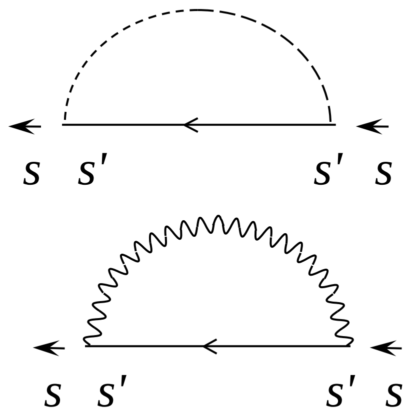
- General remarks on optical matrix elements and $\hat{\mathbf{p}} = \frac{im}{\hbar}[\mathrm{H}_0, \mathbf{r}]_-$ see Gu et al. 2013

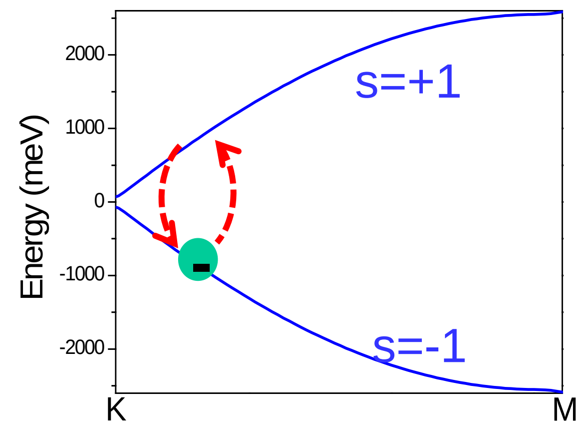
Fano lineshape factor L_F from: Mak, Shan, Heinz, PRL 106, 046106 (2011)
Optical matrix elements: Gu, Kwong, Binder, PRB 87, 125301 (2013)

Renormalization

Σ^{e-ph}

Σ^{e-e}





$$\Sigma^{\text{e-ph}} = \Sigma^{\text{e-ph}}_{\text{equil}} + \Delta\Sigma^{\text{e-ph}}$$

changes due to modified phonon populations (temperature change) and modified electron populations

does not require thermal phonon population, optical phonons dominate

requires thermal phonon population, favors acoustic phonons

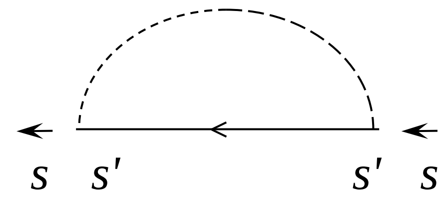
$$\text{Signal} = \frac{T - T_0}{T_0} \sim \Delta\Sigma^{\text{e-ph}}$$

optical signal sensitive to acoustic phonons

Renormalization due to electron-phonon interaction

$$C^{\mathrm{ac}}_{s,s'}(\boldsymbol{k},\boldsymbol{k}') = \frac{\hbar A_0}{4m\omega^{\mathrm{LA}}_{\boldsymbol{k}-\boldsymbol{k}'}} \left| D^{\mathrm{ac}}_{\mathrm{eff}} \, |\boldsymbol{k}-\boldsymbol{k}'|_{\text{reduced to BZ}} \, \langle u_{\boldsymbol{k},s} | u_{\boldsymbol{k}',s'} \rangle \right|^2$$

$$C^{\mathrm{op}}_{s,s'}(\boldsymbol{k},\boldsymbol{k}') = \frac{\hbar A_0}{4m\omega^{\mathrm{LO}}_{\boldsymbol{k}-\boldsymbol{k}'}} \left| D^{\mathrm{ac}}_{\mathrm{eff}} \langle u_{\boldsymbol{k},s} | u_{\boldsymbol{k}',s'} \rangle \right|^2$$



Emission possible without thermal excitation, optical phonons dominate

$$\Sigma^{\text{e-ph}}_s(\boldsymbol{k}) = \int \frac{d^2k'}{(2\pi)^2} \sum_{\mu,s'} C^{\mu}_{s,s'}(\boldsymbol{k},\boldsymbol{k}') \left[\frac{1 - f_{s'}(\boldsymbol{k}') + n^{\mathrm{ph}}_{\mu}(\boldsymbol{k}-\boldsymbol{k}')}{E^{(0)}_s(\boldsymbol{k}) - E^{(0)}_{s'}(\boldsymbol{k}') - \hbar\omega^{\mu}_{\boldsymbol{k}-\boldsymbol{k}'} + i\eta} + \frac{f_{s'}(\boldsymbol{k}') + n^{\mathrm{ph}}_{\mu}(\boldsymbol{k}-\boldsymbol{k}')}{E^{(0)}_s(\boldsymbol{k}) - E^{(0)}_{s'}(\boldsymbol{k}') + \hbar\omega^{\mu}_{\boldsymbol{k}-\boldsymbol{k}'} + i\eta} \right]$$

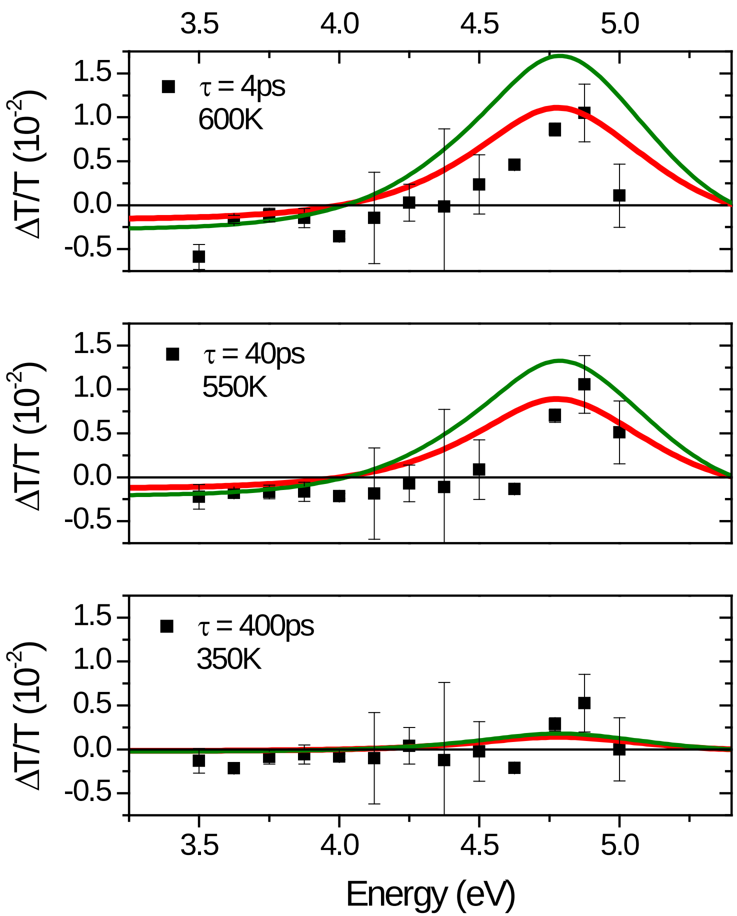
phonon emission — phonon absorption

Phonon emission contribution requires thermal occupation! Favors acoustic phonons!

$$\Delta\Sigma^{\text{e-ph}}_s(\boldsymbol{k}) = \int \frac{d^2k'}{(2\pi)^2} \sum_{\mu,s'} C^{\mu}_{s,s'}(\boldsymbol{k},\boldsymbol{k}') \left[\frac{-\Delta f_{s'}(\boldsymbol{k}') + \Delta n^{\mathrm{ph}}_{\mu}(\boldsymbol{k}-\boldsymbol{k}')}{E^{(0)}_s(\boldsymbol{k}) - E^{(0)}_{s'}(\boldsymbol{k}') - \hbar\omega^{\mu}_{\boldsymbol{k}-\boldsymbol{k}'} + i\eta} + \frac{\Delta f_{s'}(\boldsymbol{k}') + \Delta n^{\mathrm{ph}}_{\mu}(\boldsymbol{k}-\boldsymbol{k}')}{E^{(0)}_s(\boldsymbol{k}) - E^{(0)}_{s'}(\boldsymbol{k}') + \hbar\omega^{\mu}_{\boldsymbol{k}-\boldsymbol{k}'} + i\eta} \right]$$

$\Delta f_s(\boldsymbol{k},\tau) = f_s(\boldsymbol{k},\tau) - f^{\mathrm{equi}}_s(\boldsymbol{k})$, $\quad \Delta n^{\mathrm{ph}}_{\mu}(\boldsymbol{q}) = n^{\mathrm{ph}}_{\mu}(\boldsymbol{q},T_{\mu}) - n^{\mathrm{ph}}_{\mu}(\boldsymbol{q},T_{\mathrm{equi}})$, $\quad \langle u_{\boldsymbol{k},s} | u_{\boldsymbol{k}',s'} \rangle = 1/2\left(1 + ss' e^{i(\phi(\boldsymbol{k})-\phi(\boldsymbol{k}'))}\right)$

Saddle-point (M-point) spectroscopy: Results



Experiment:

Zero-crossing at ~4.1eV

Similar shape for all delay times (suggesting similar physical origin)

Theory reproduces

✓ order of magnitude

✓ shape

✓ zero-crossing

$D_{\text{eff}}^{ac} = 5.3\,\text{eV}$ Kaasbjerg et al., PRB 85,165440 (2012)

$D_{\text{eff}}^{op} = 11\,\text{eV/A}$ Low et al. PRB 86, 045413 (2012)

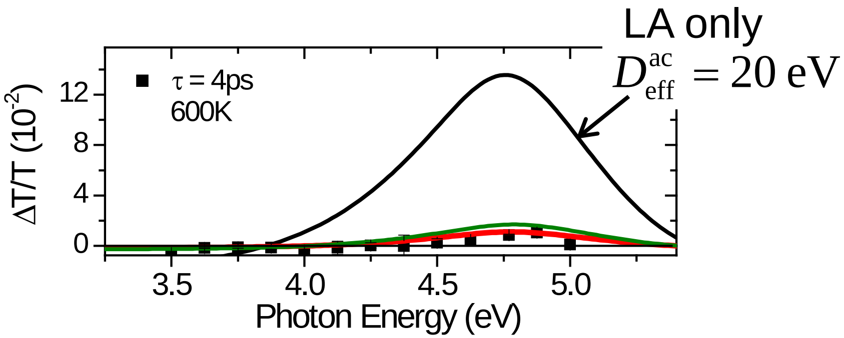
Red line: LA

Green line: LA+LO

Roberts, Binder, Kwong, Golla, Cormode, LeRoy, Everitt, Sandhu, Phys. Rev. Lett. 112, 187401 (2014)

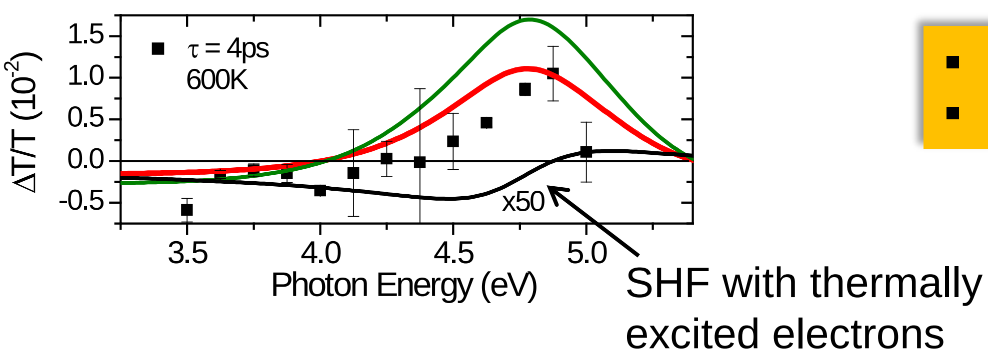
Saddle-point (M-point) spectroscopy: Results

$D_{eff}^{ac} = 20$ eV:



- Lineshape wrong
  (ratio positive vs negative ΔT/T)
- Zero-crossing too low
- Order of magnitude wrong

Electron-electron interaction (screened Hartree-Fock):



- Lineshape completely wrong
- Order of magnitude wrong

Both do not reproduce experiment

Roberts, Binder, Kwong, Golla, Cormode, LeRoy, Everitt, Sandhu, Phys. Rev. Lett. 112, 187401 (2014)

Summary

- Presented differential optical transmission spectra from transitions close to M-point
- Observed π-π* band renormalization
- For time delays > 4 ps, electron-phonon interaction dominates
- Differential transmission geometry favors acoustic phonons as source of ΔT/T
- Studied electron-LA deformation potential (range of literature values ~2-50 eV)
- Initial (at 4 ps) temperature determined by phonon energy and deposited optical energy
- Temperature for 40ps and 400ps obtained from time-dependent data with 190 ps rate
- Spectra show negative and positive signal with crossing at ~4.1eV
- Good theory-experiment agreement with effective acoustic deformation potential ~5eV
- Electron-electron effects do not reproduce experiment

Reference: Roberts, Binder, Kwong, Golla, Cormode, LeRoy, Everitt, Sandhu, Phys. Rev. Lett. 112, 187401 (2014)

Future improvements may include:

- Substrate dependence of deformation potentials
- Excitonic effects in probe absorption
- Excitonic effects in electron-phonon coupling
- More detailed electron-phonon coupling models (def. potential plus gauge field coupling)
- Substrate dependence of gauge field coupling