Ultrafast dynamics of hot carriers in bulk semiconductors and in accumulation layer: energy loss rate and screening effects.

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Collaborations

Ecole Polytechnique, LSI:	N. Vast,
	R. Sen (post-doc)
	L. Perfetti (ARPES, 2PPE)
Osaka, Japan:	K. Tanimura (ARPES, 2PPE)

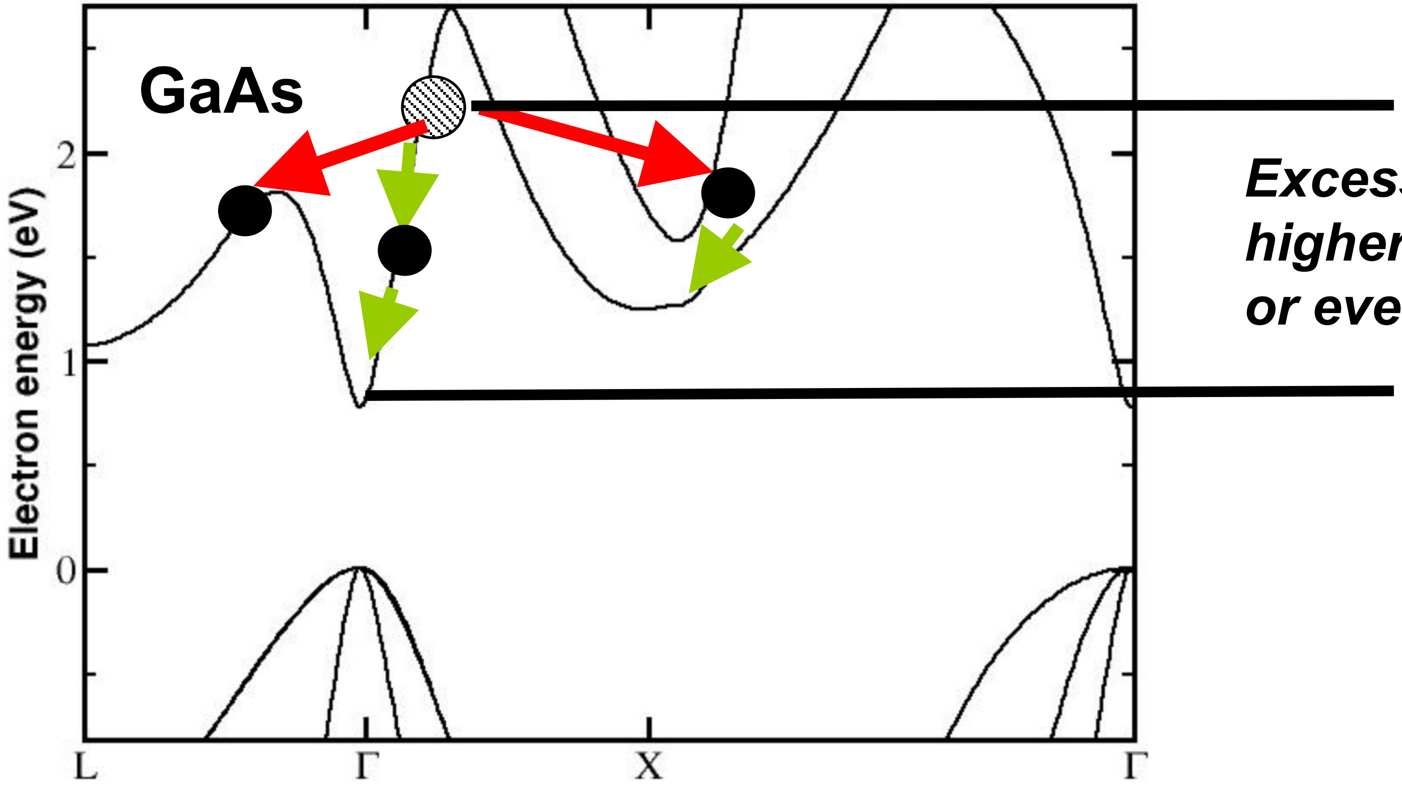
Outline

- ***Electron-phonon scattering for highly excited electrons***

- ***Highly excited electron relaxation in Si***

- ***Photoexcited electron relaxation in InSe***

RELAXATION DYNAMICS OF HIGHLY EXCITED ELECTRONS

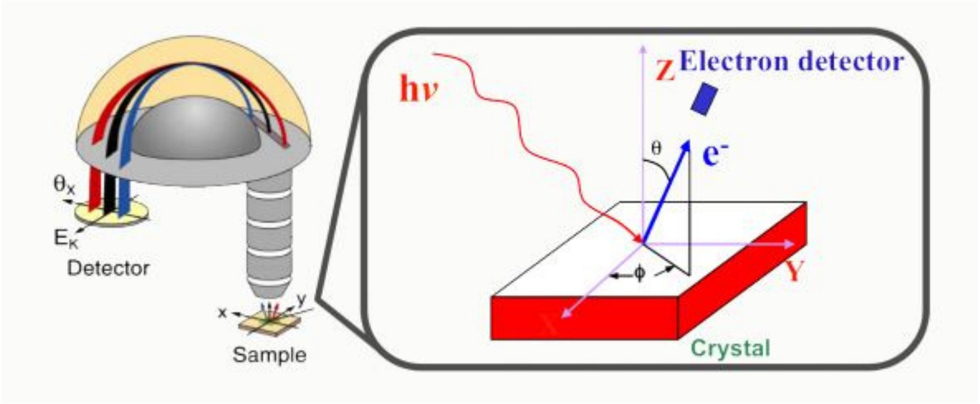


Excess energy:
higher than second
or even third valley minimum

Main scattering mecanism: intervalley electron-phonon scattering

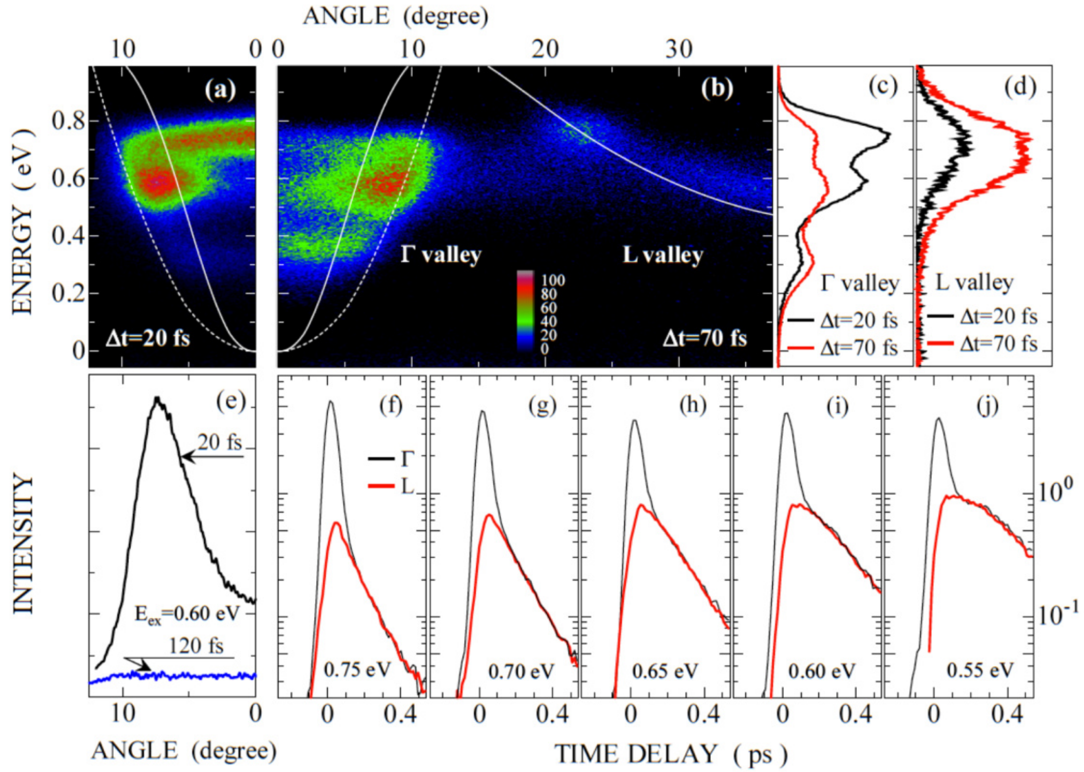
Sjakste, Tanimura, Barbarino, Perfetti, Vast, J. Phys: Cond. Mat. ***30****, 353001 (2018).*

EXPERIMENTS: ARPES



Pump and probe: 2 laser pulses at different time

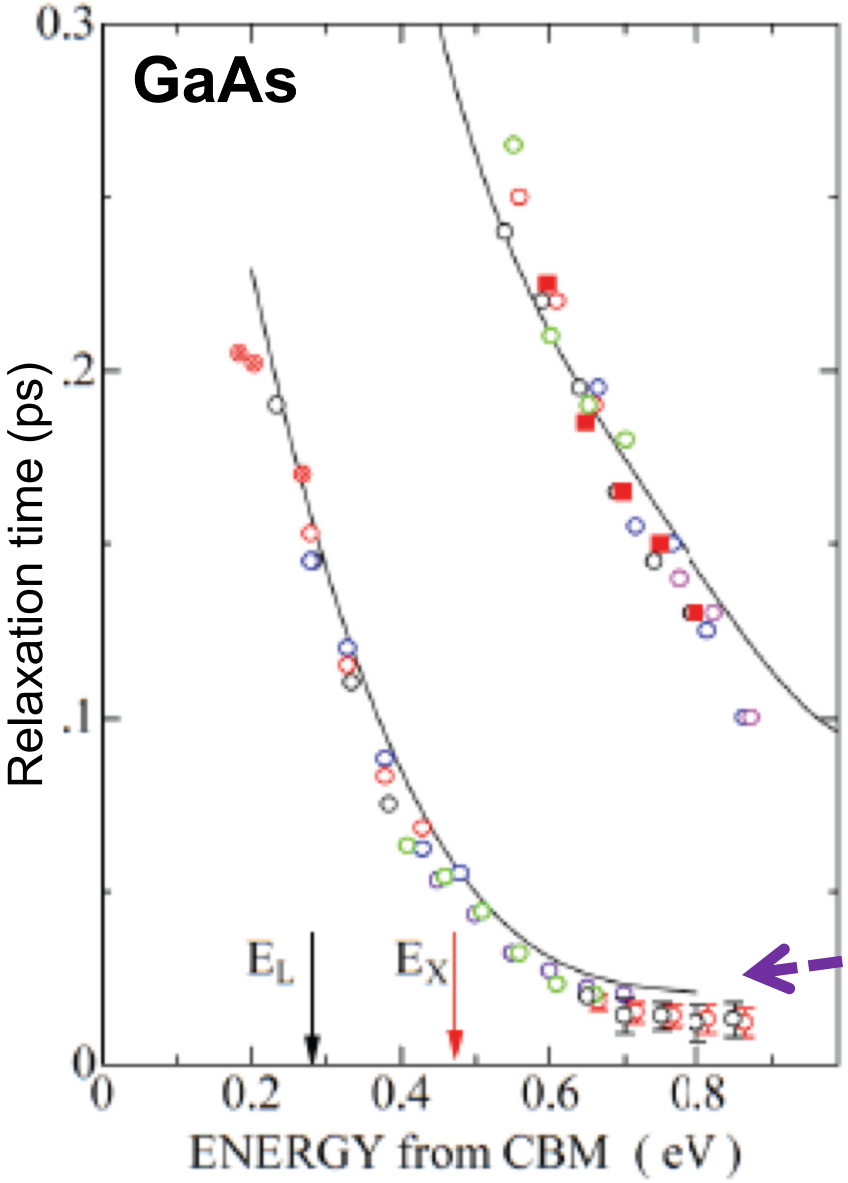
GaAs
300K

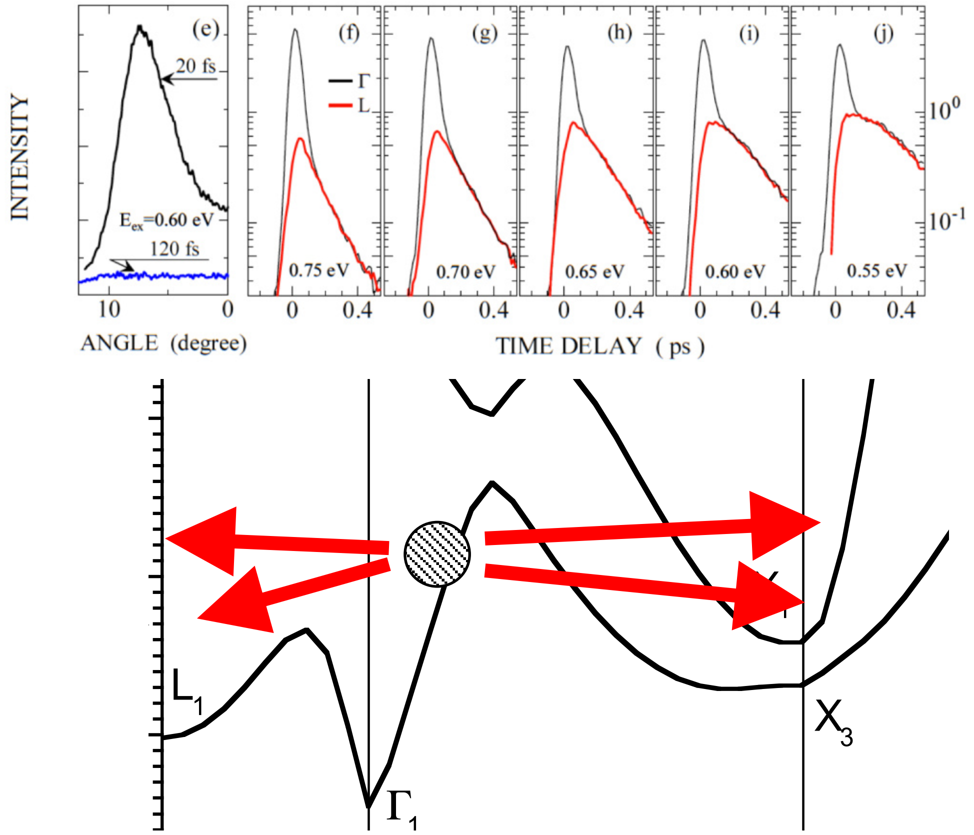


Two distinct relaxation regimes

H. Tanimura, J. Kanasaki, K. Tanimura, J. Sjakste, N. Vast, M. Calandra, F. Mauri, PRB 93 (2016) (R)

HOT ELECTRON ENSEMBLE (HEE)





« Short » time: wavepacket spreads over BZ;
« isomerization »: total scattering rate, mostly intervalley transitions

Tanimura et al, PRB 93 (2016) 161203 (R).

Sjakste et al, J. Phys: Cond. Mat. ***30****, 353001 (2018).*

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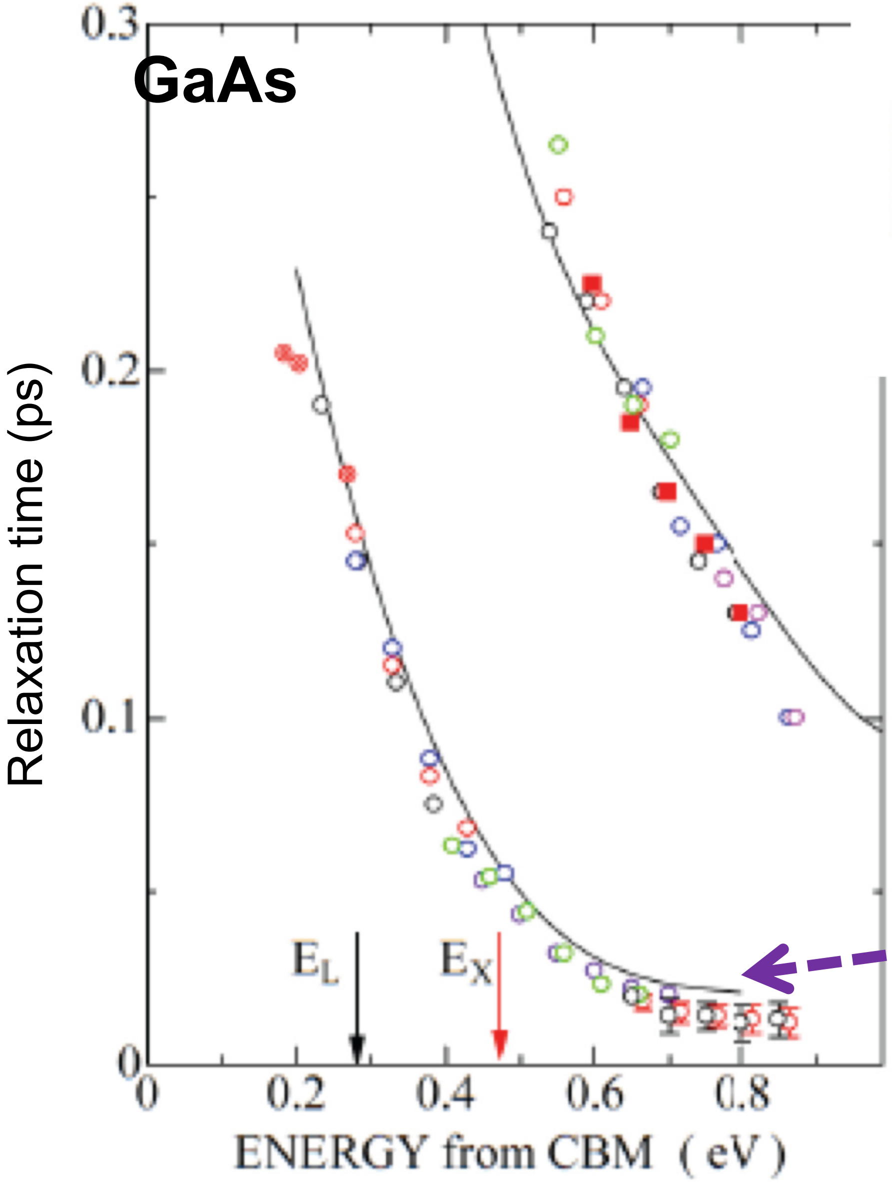

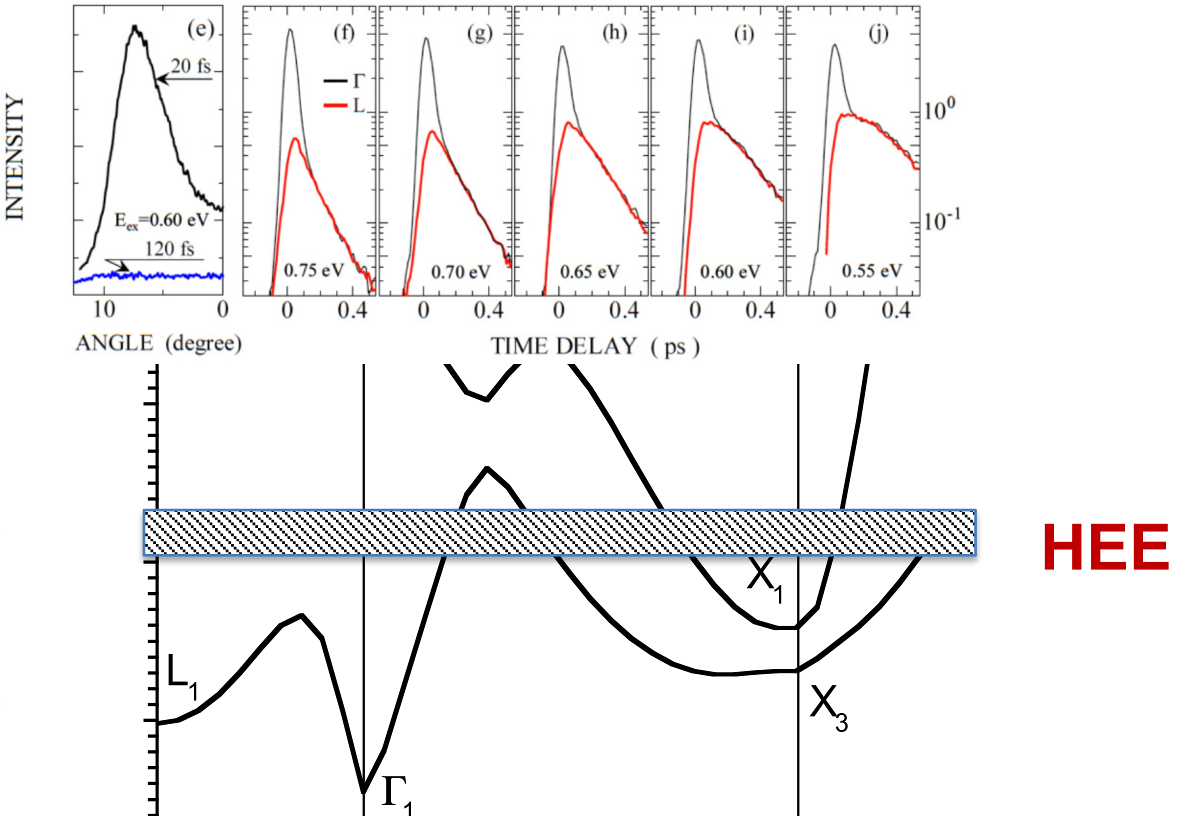

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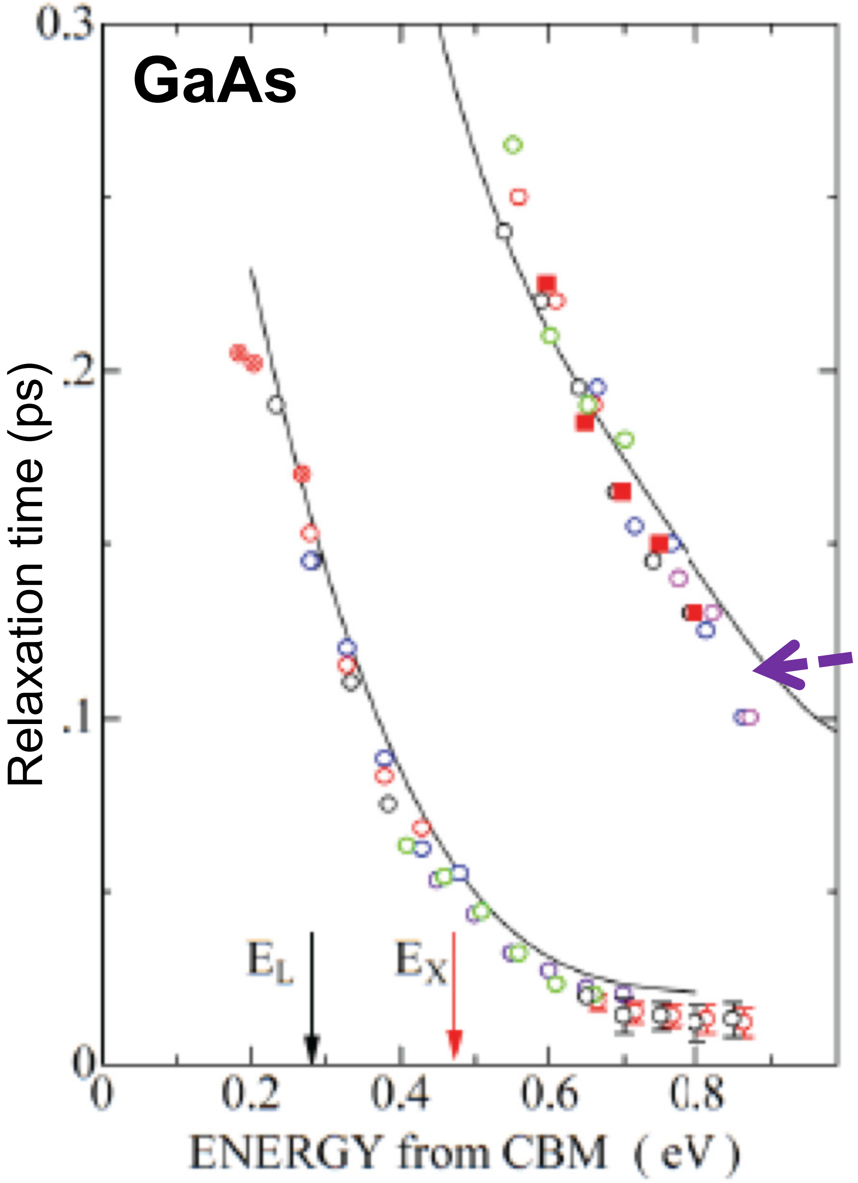
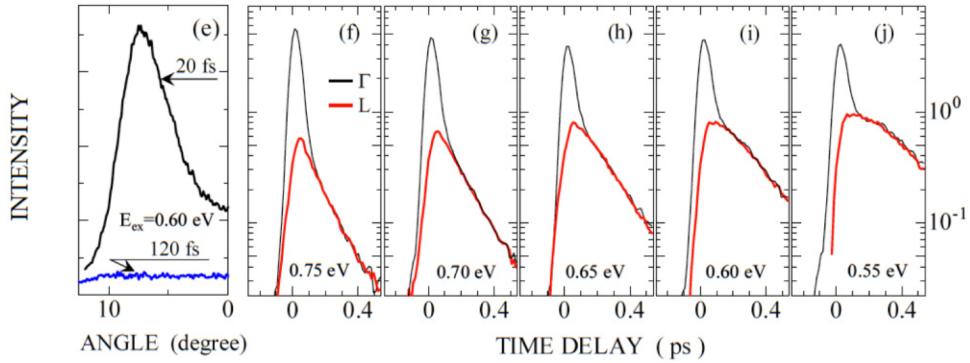
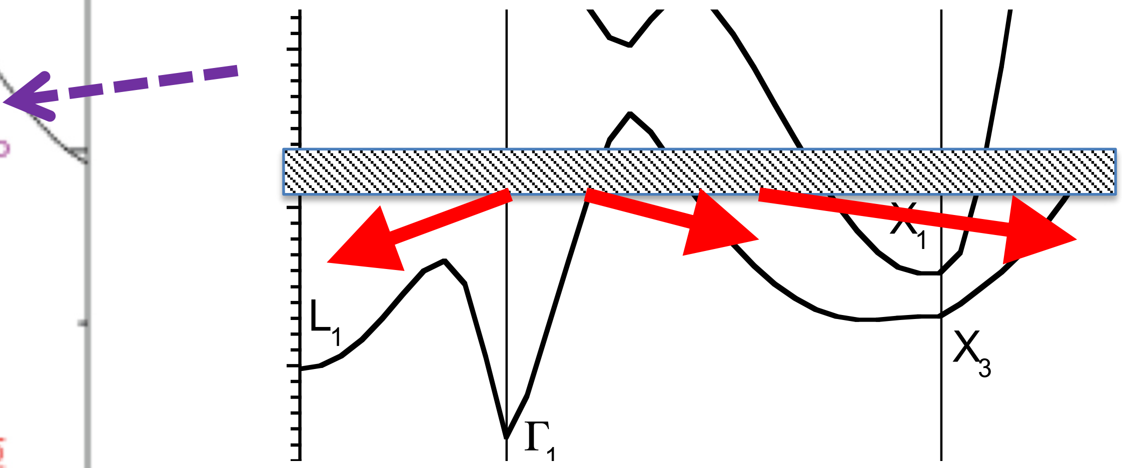




« long » time: energy transfer to phonons



mostly intervalley transitions

Tanimura, Kanasaki, Tanimura, Sjakste, Vast, Calandra, Mauri, PRB 93 (2016) 161203 (R).

CALCULATIONS: DFPT+Wannier

Reciprocal space
Bloch functions
Initial grid

$$\left\langle \Psi_{n,k} \middle| \Delta W_q^{\lambda} \middle| \Psi_{n',k+q} \right\rangle$$ **- *Non-local part (if polar)***

Real space
Maximally localized Wannier functions
Interpolation on dense grid

Reciprocal space
Bloch functions
Dense grid

$$\left\langle \Psi_{n,k} \middle| \Delta W_q^{\lambda} \middle| \Psi_{n',k+q} \right\rangle$$ **+ *Non-local part (if polar)***

J. Sjakste, N. Vast, M. Calandra, F. Mauri, PRB 92 (2015) 054307

C. Verdi, F. Giustino, PRL 115 (2015) 176401

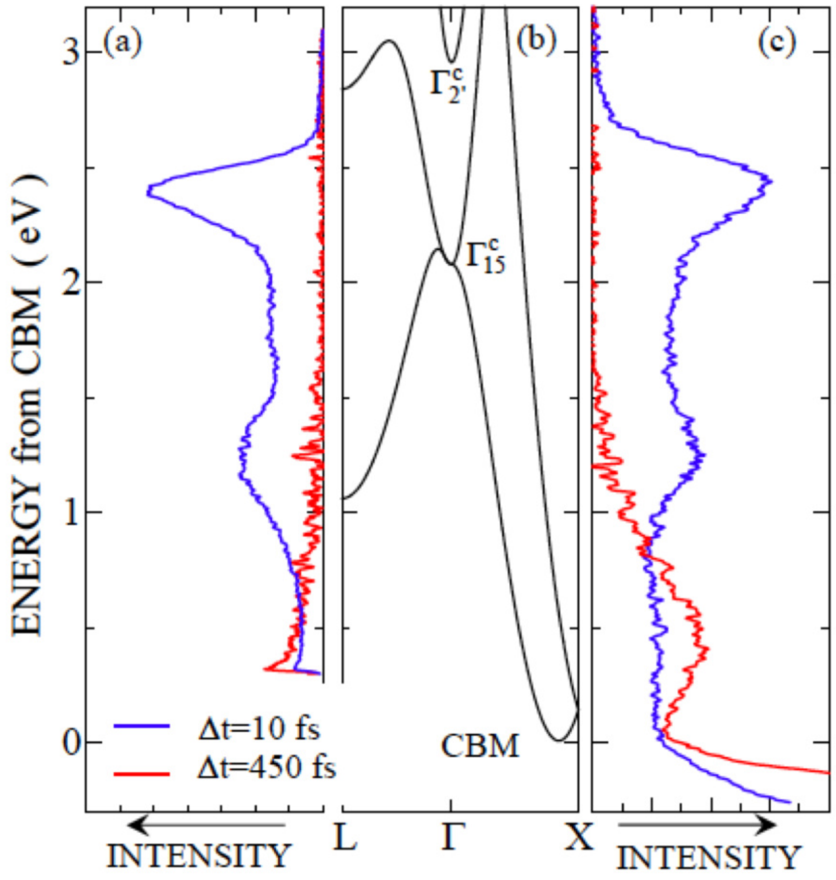
Outline

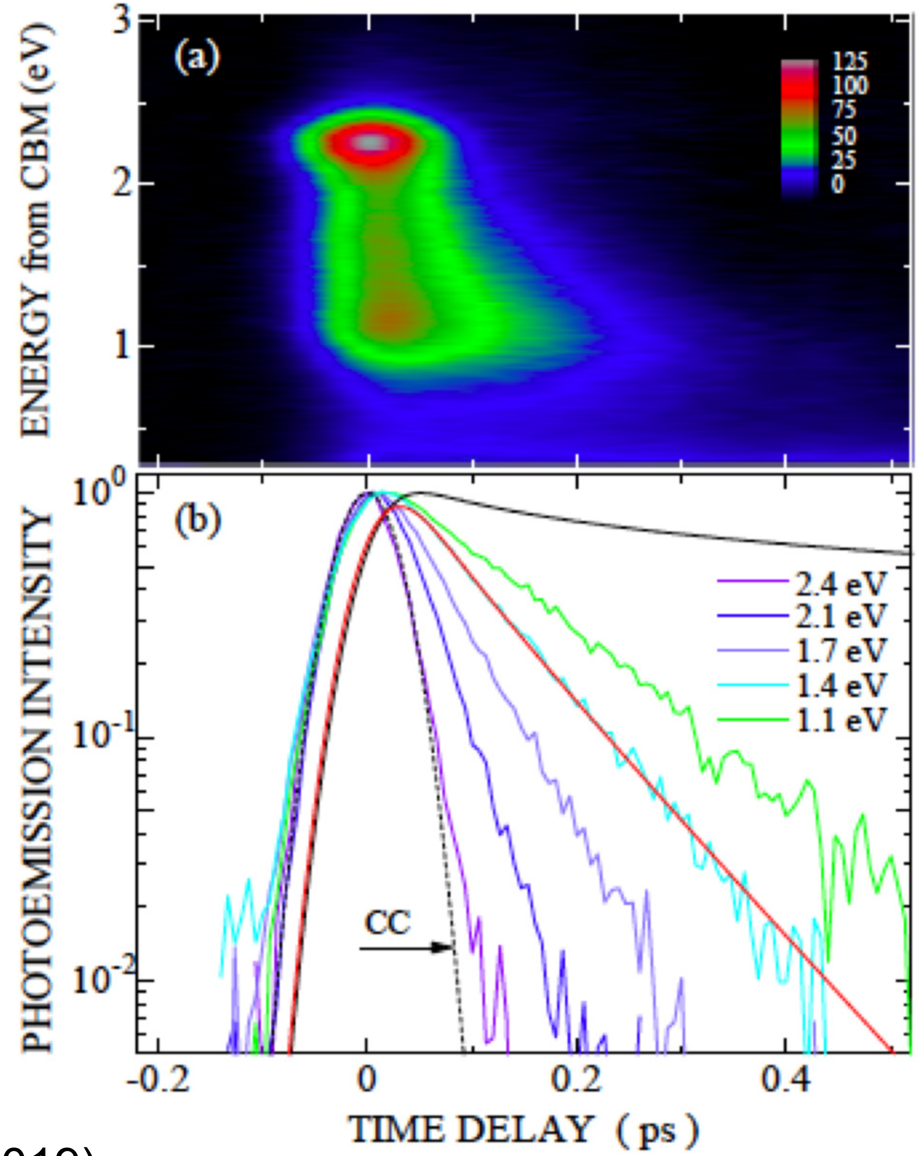
- Electron-phonon scattering for highly excited electrons

- ***Highly excited electron relaxation in Si***

- ***Photoexcited electron relaxation in InSe***

HIGHLY EXCITED ELECTRONS IN SILICON: 2PPE





Excess energies: 1-3 eV above CBM

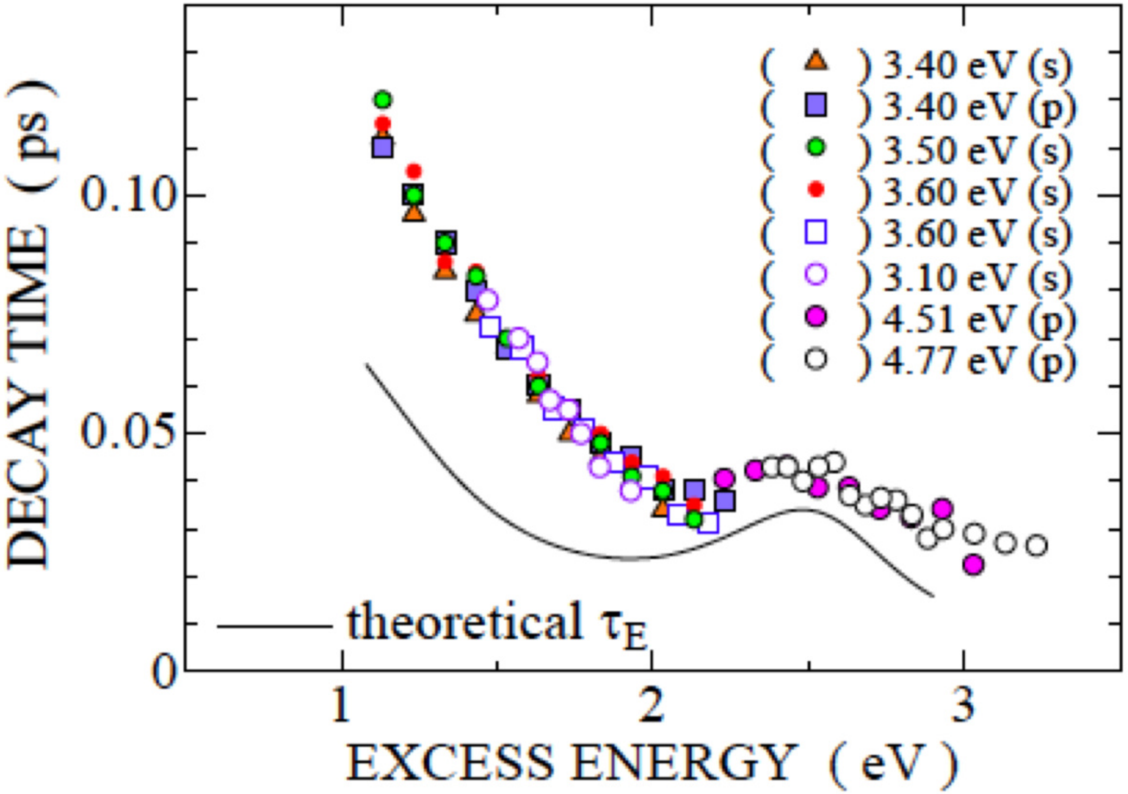
Tanimura et al, PRB **100**, 03520 (2019).

HIGHLY EXCITED ELECTRONS IN SILICON: INTERPRETATION PROBLEM

Previous work: conflict theory/experiment:

Measured relaxation times 10 times longer than calculated ones

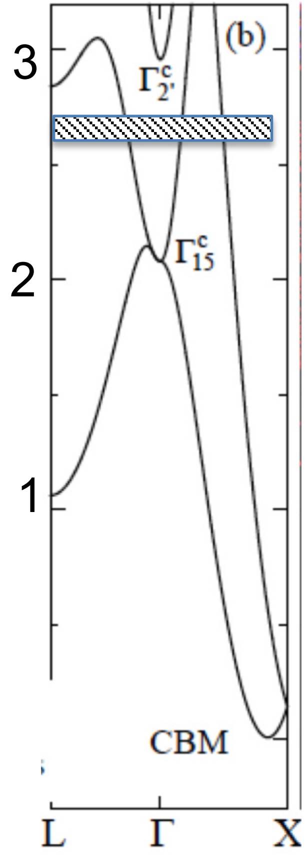
Ichibayashi *et al,* Phys. Rev. B 84, 235210 (2011).



This work: HEE idea

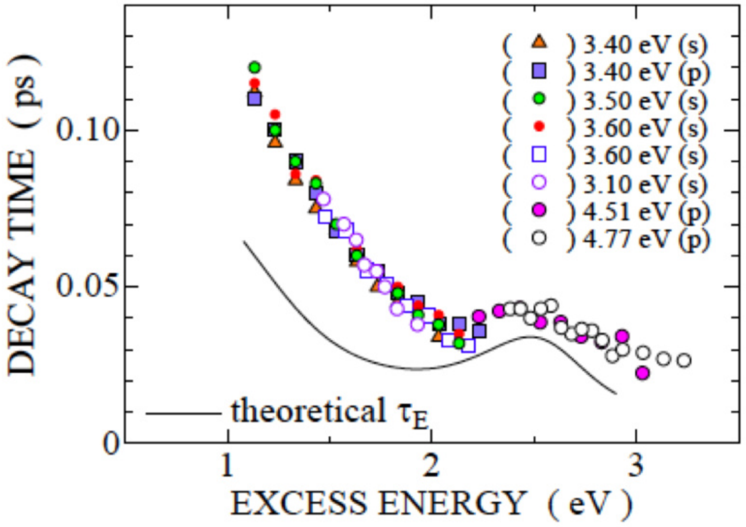
Initial relaxation:
too fast to be measured

Measured relaxation times:
energy loss

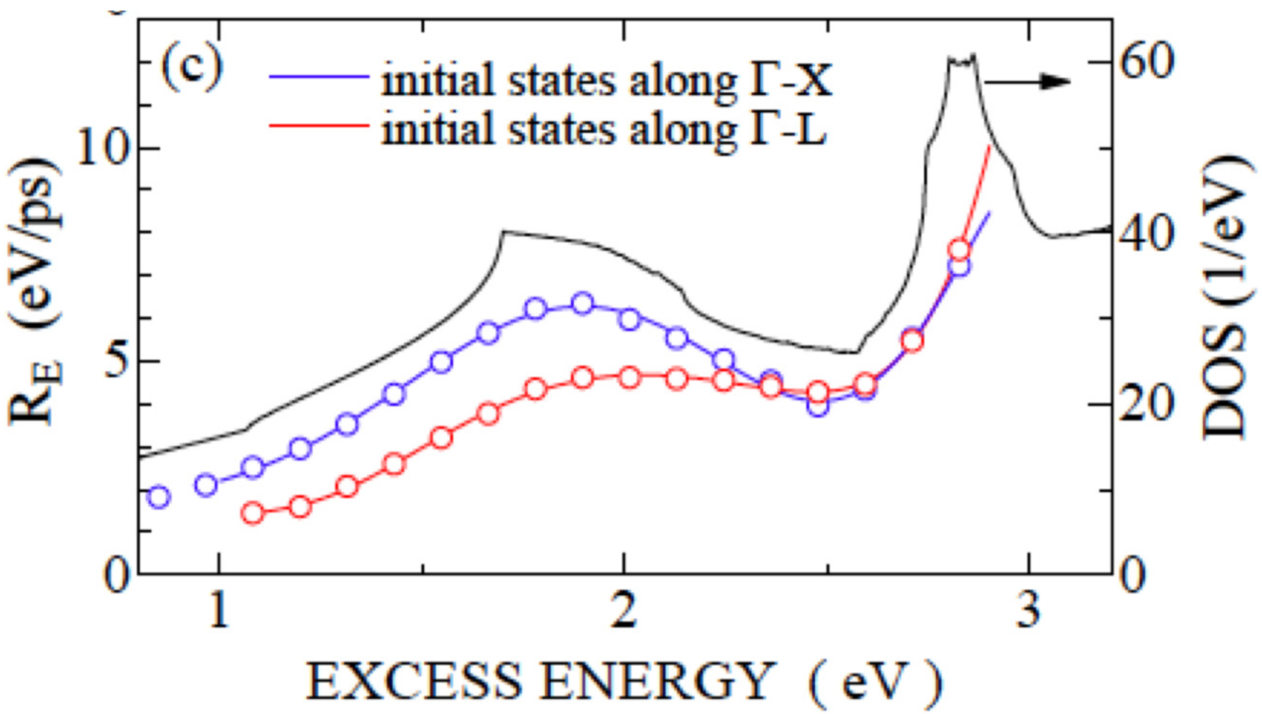


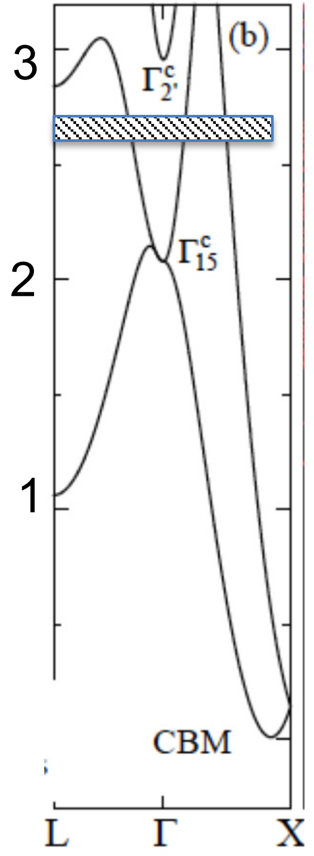
Tanimura, Kanasaki, Tanimura, Sjakste, Vast PRB **100**, 03520 (2019).

HIGHLY EXCITED ELECTRONS IN SILICON: ENERGY LOSS RATE



$$dE / \tau_{\mathrm{E}} = \Gamma_{em}\omega_{em} - \Gamma_{abs}\omega_{abs}$$





Energy loss rate:
Determined by DOS of final electronic states

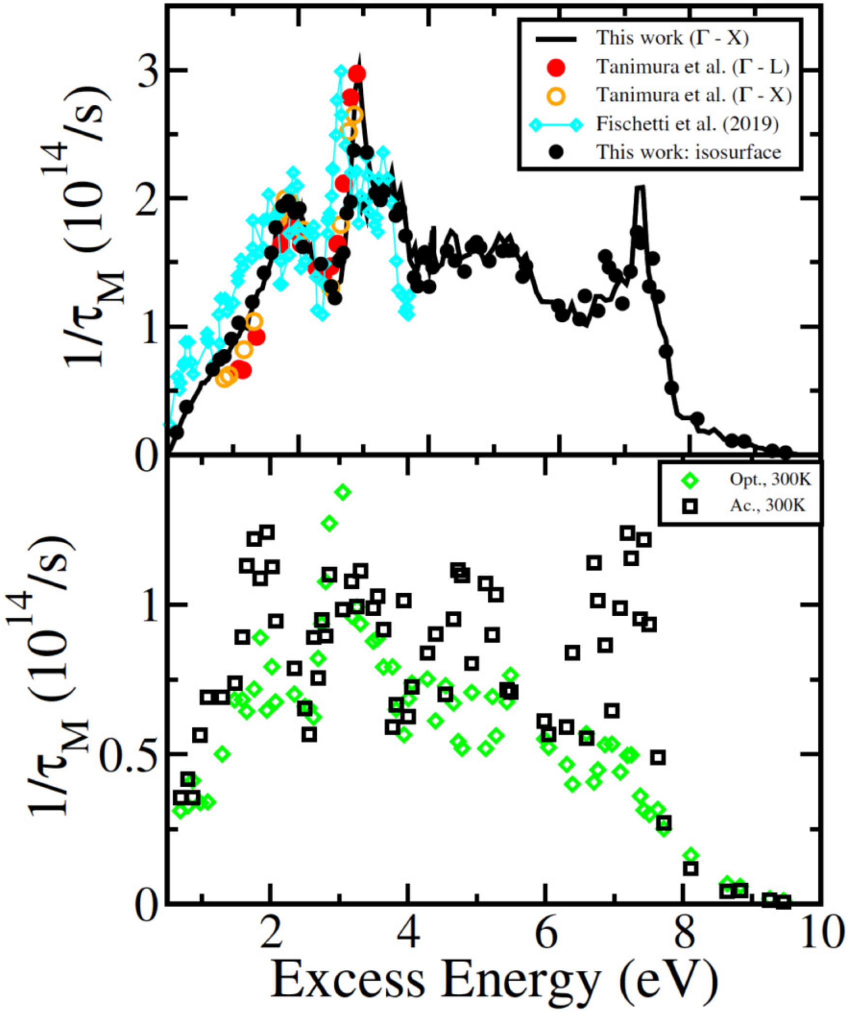
Tanimura, Kanasaki, Tanimura, Sjakste, Vast PRB **100**, 03520 (2019).

MAIN CHANNELS: IMPORTANT FOR MONTE CARLO

Raja Sen

Si, 300 K

Total scattering rate:
acoustical phonons are dominant (at 300K)



EPW code, EPIK code: identical results

Also: *Bernardi at al, PRL (2014)*

MAIN CHANNELS: IMPORTANT FOR MONTE CARLO

Raja Sen

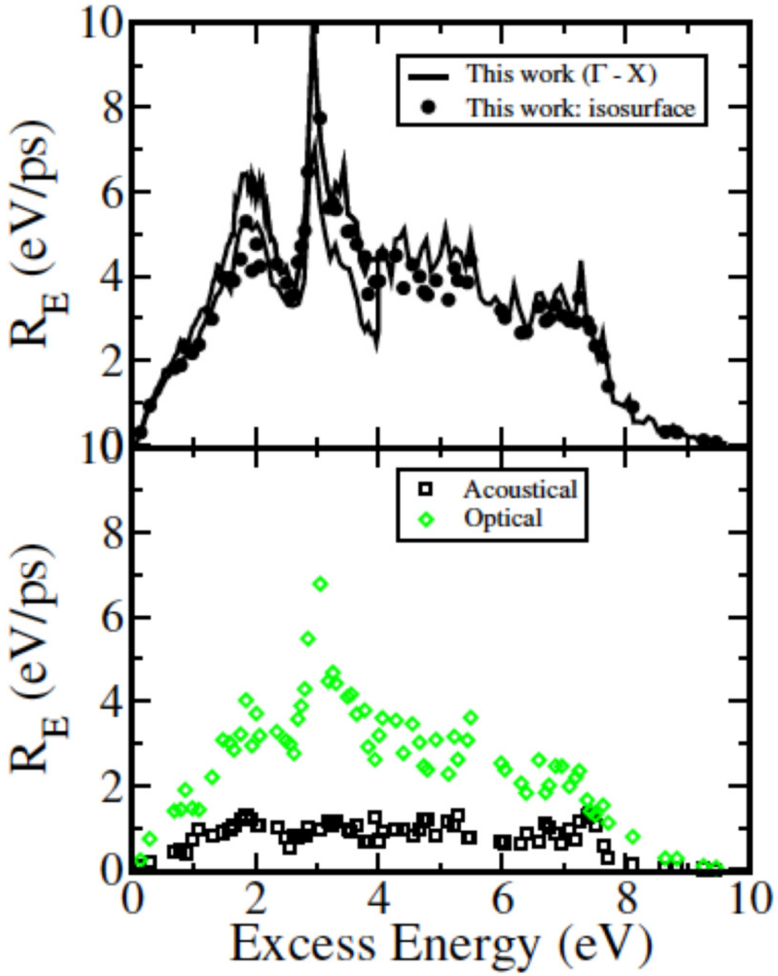
Energy loss rate:
optical phonons are dominant

Not unexpected:
Ahmad et al, Phys. Stat. Sol. 40:631 (1970)

Temperature-dependent contribution of acoustical phonons cancels out of energy loss

EPW code, EPIK code: identical results

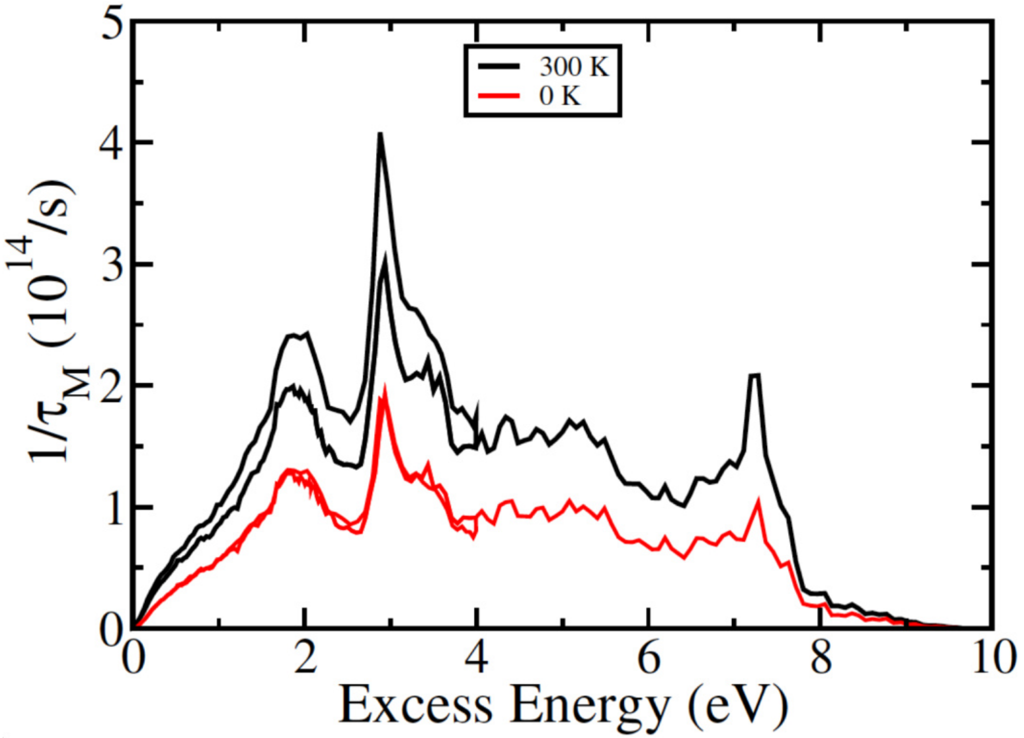
Si, 300 K



TEMPERATURE DEPENDENCE

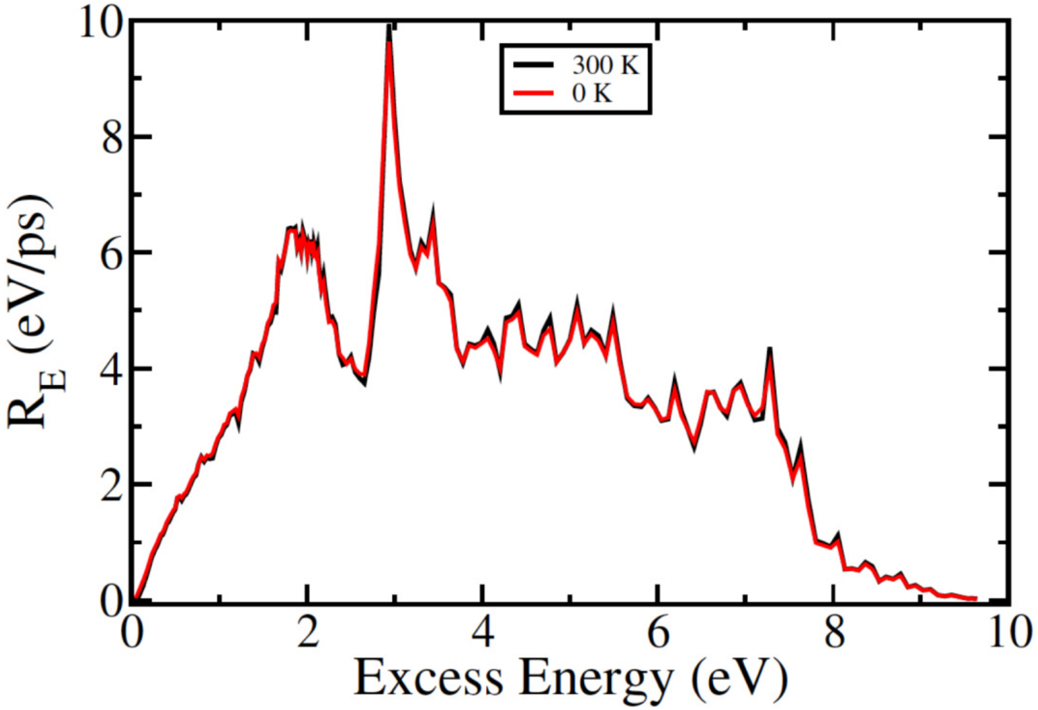
Raja Sen, post-doc

Total scattering rate, Si



Populations of acoustical phonons grow with temperature

Energy loss rate, Si.



Negligible temperature dependence for energy loss rate

GaAs (Theory and experiment): *Sjakste et al, PRB* **97**, 064302 *(2018)*

Outline

- *Electron-phonon scattering: general picture*

- *Electron-phonon scattering for highly excited electrons*

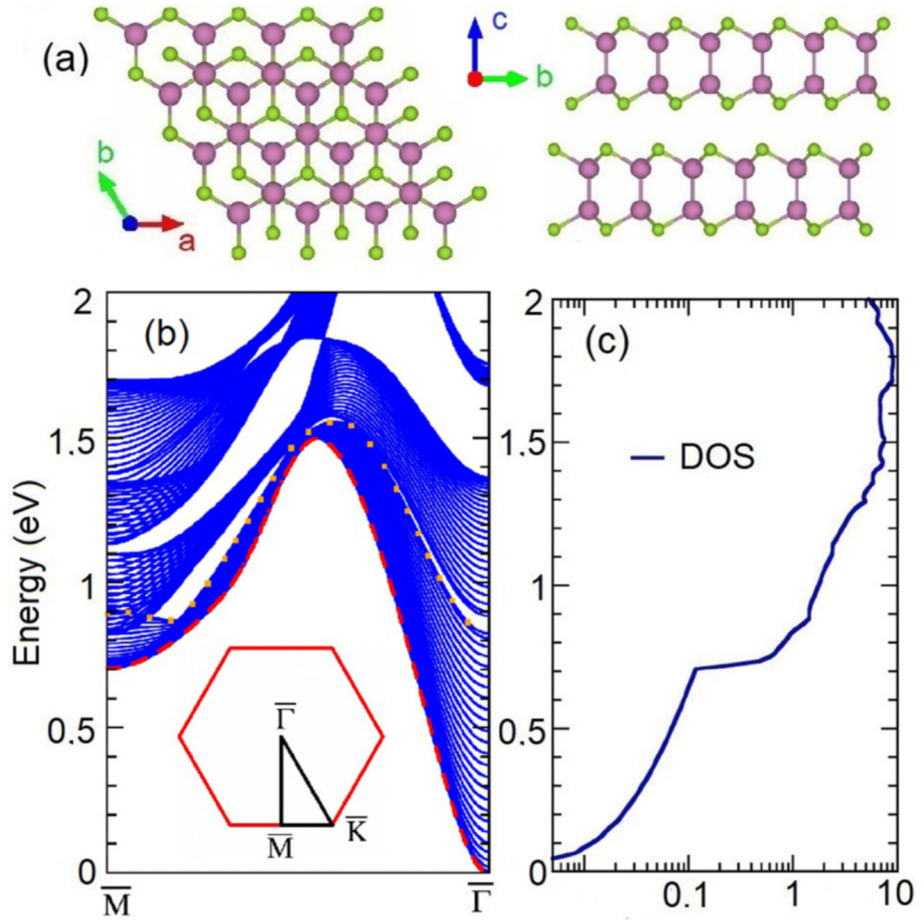
- *Highly excited electron relaxation in Si*

- *Photoexcited electron relaxation in InSe*

Photoexcited electron relaxation in InSe

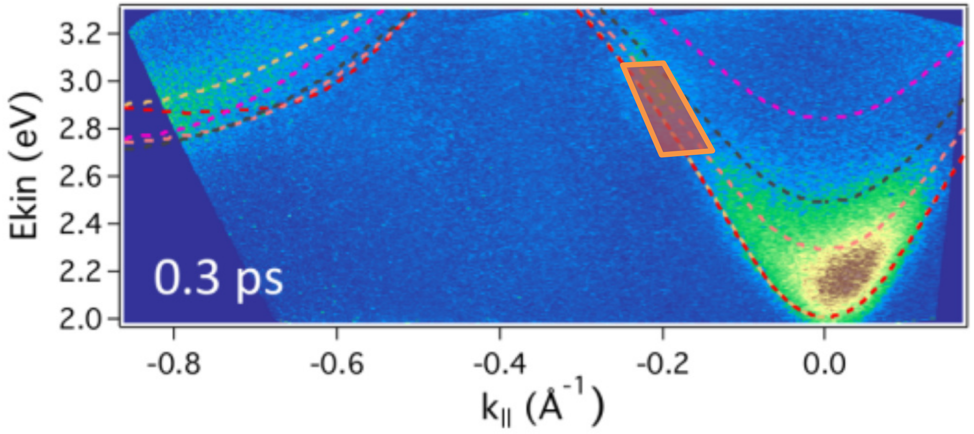
Luca PERFETTI
Zhesheng CHEN
Zailan ZHANG
Raphael CABOUAT
Jelena SJAKSTE
Cristine GIORGETTI
Valerie VENIARD
Abdelkarim Ouerghi
Hugo Henck

Evangelos PAPALAZAROU
Marino MARSI

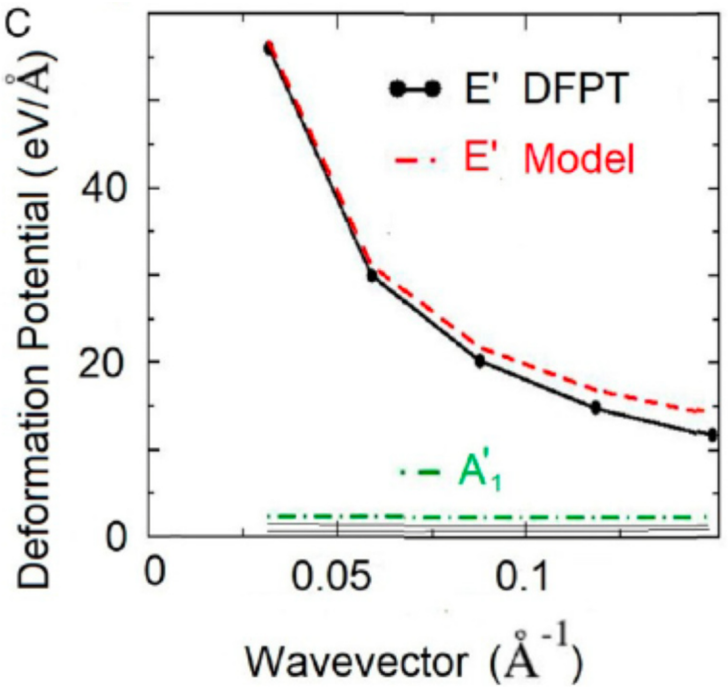


- Layered material
- Very narrow Γ valley
- Large M valley

InSe: energy relaxation in Γ valley



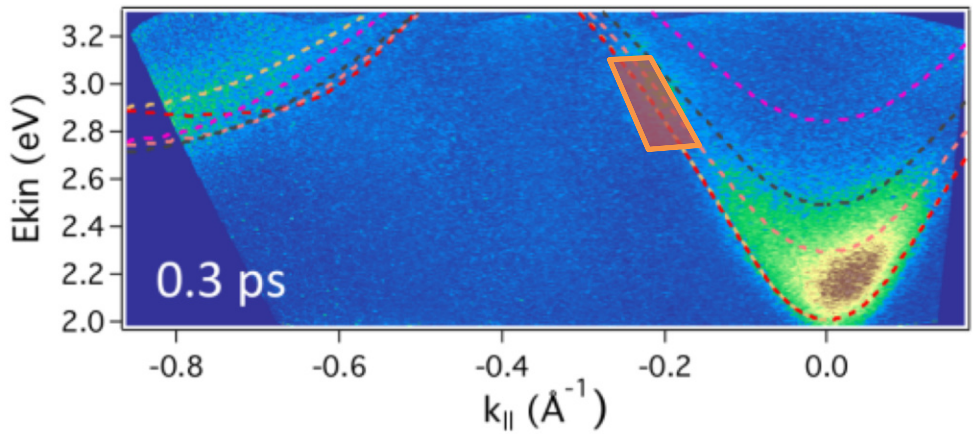
Excess energy below 0.7eV



- **no intervalley scattering**
- **very narrow Γ valley (q<0.2 ang.)**
- **Fröhlich scattering**
  (scatering by polar phonons)

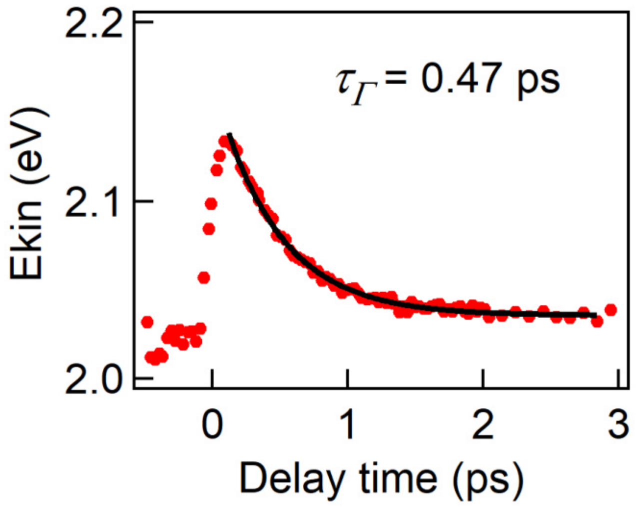
Chen, Giorgetti, Sjakste, Cabouat et al, PRB 97, 241201(R) (2018).

InSe: energy relaxation in Γ valley

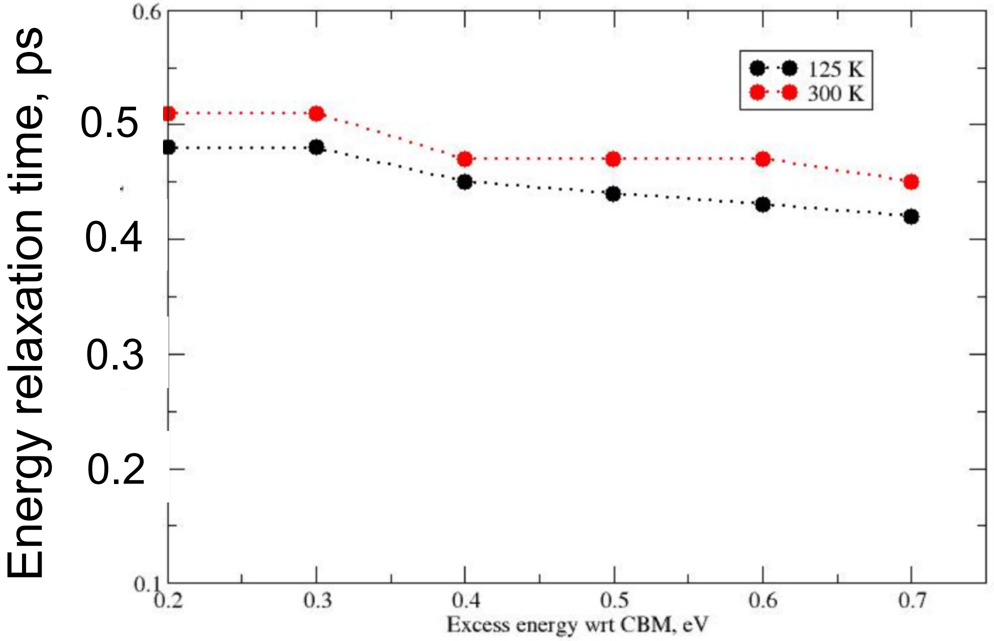


below 0.7 eV

Experiment



Theory: energy transfer due to coupling with polar phonons

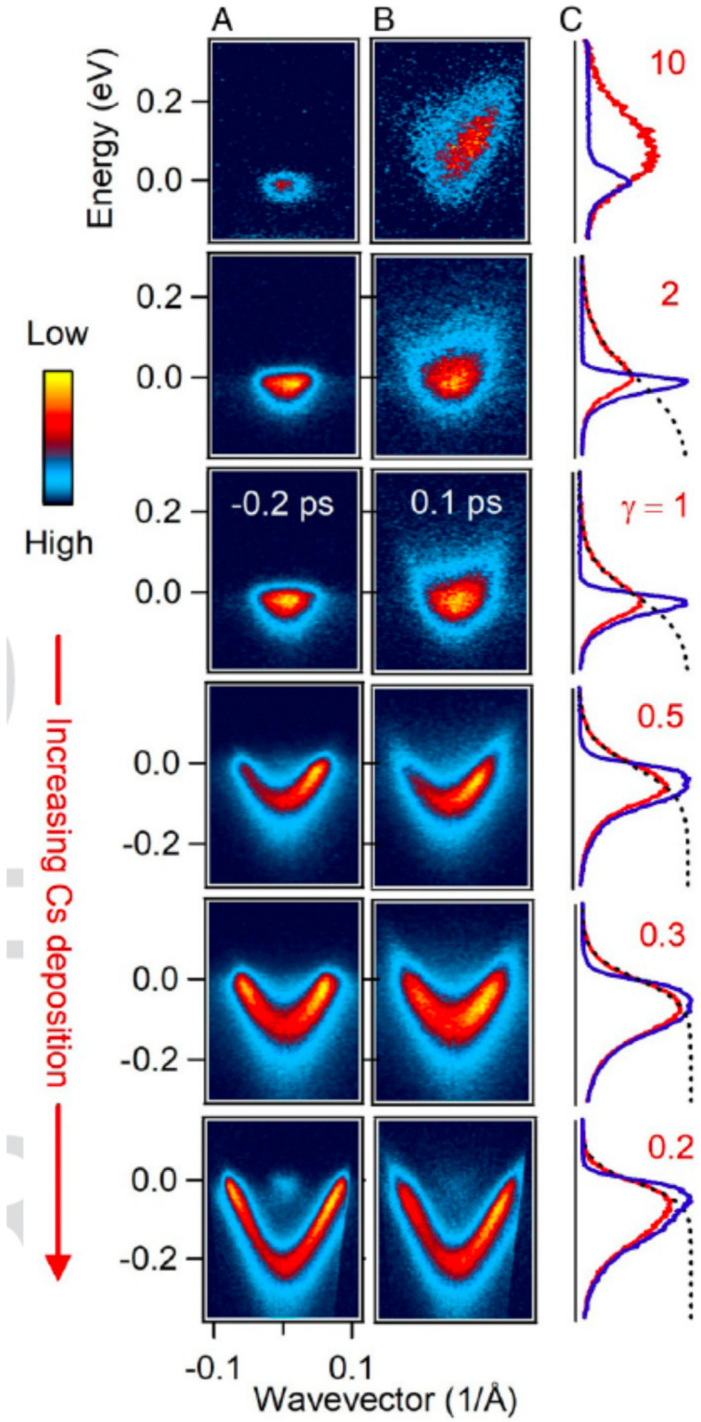


scattering by polar phonons

$$\frac{4\pi ie}{\varepsilon_\infty q^2} q_\mu \sum_\alpha Z_{\mu\lambda}(\alpha) e_\lambda(\alpha \hat{q})$$

Good agreement between theory and experiment.

Chen, Giorgetti, Sjakste, Cabouat et al, PRB 97, 241201(R) (2018).



Quasi-two-dimensional gas on InSe

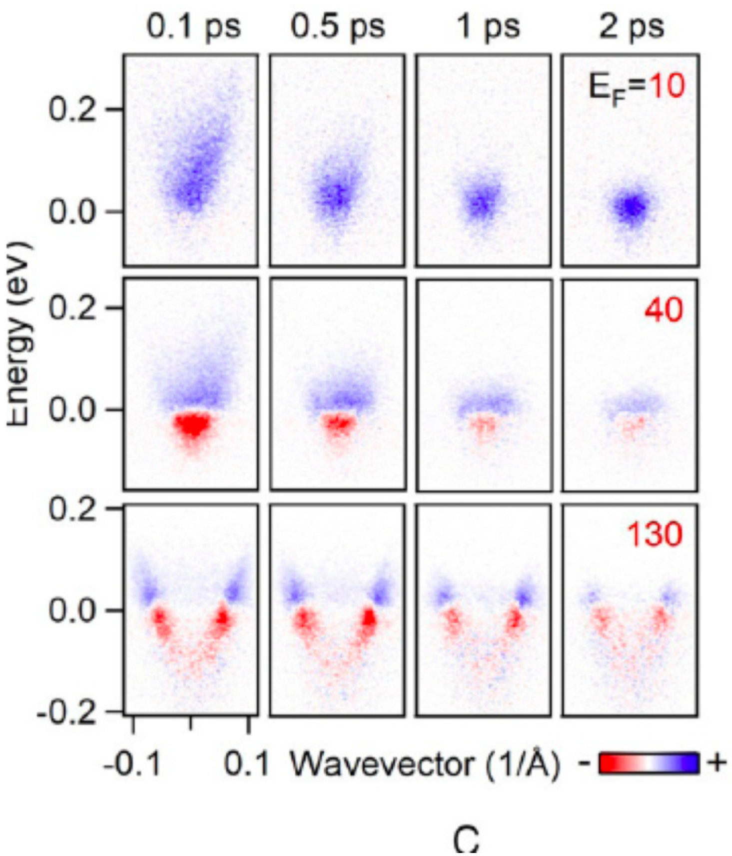
Experiment:

Zhesheng CHEN
Luca PERFETTI

Increasing Cs deposition on InSe surface

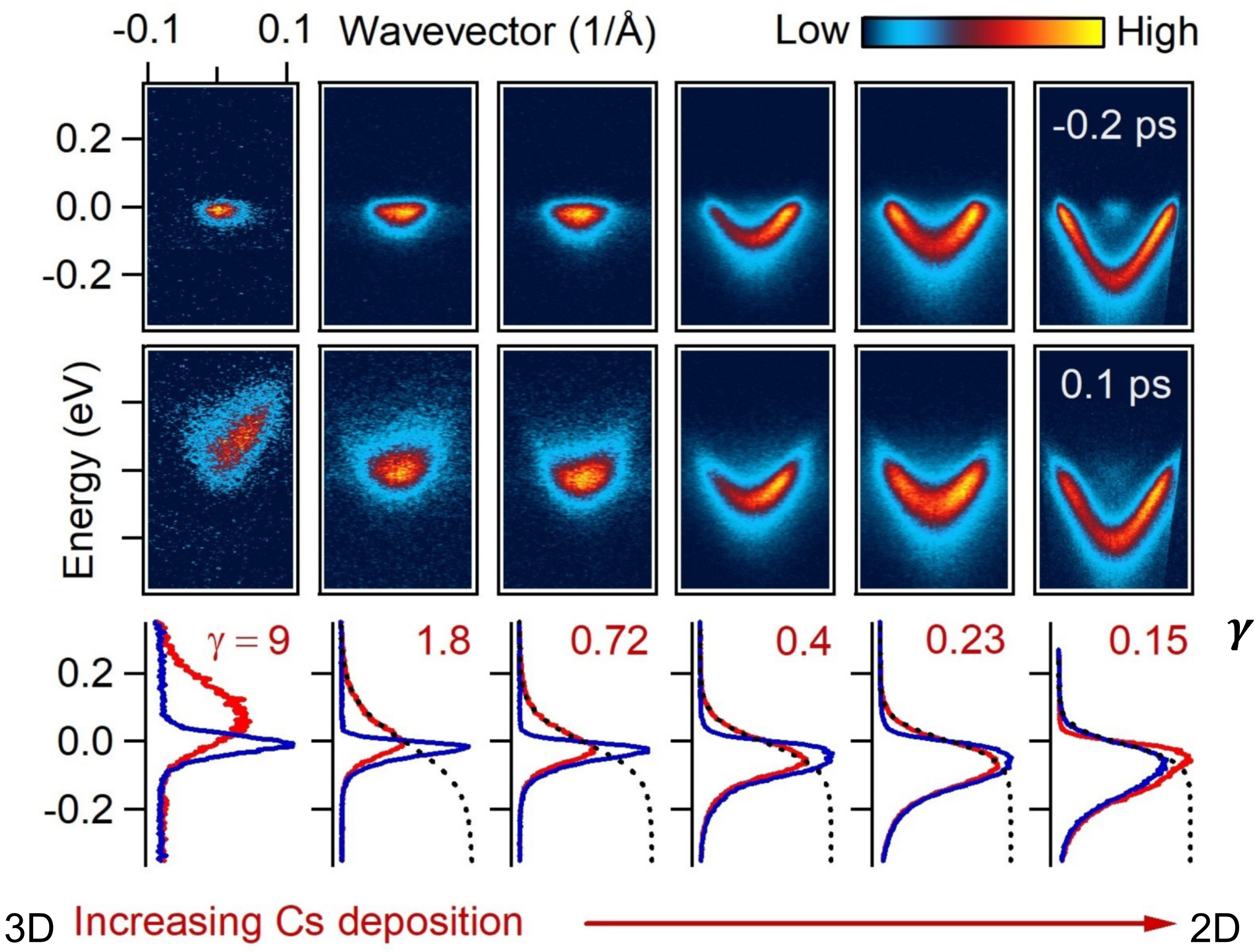
electron gas created on the surface

Dynamics of excited electrons studied by tr-ARPES

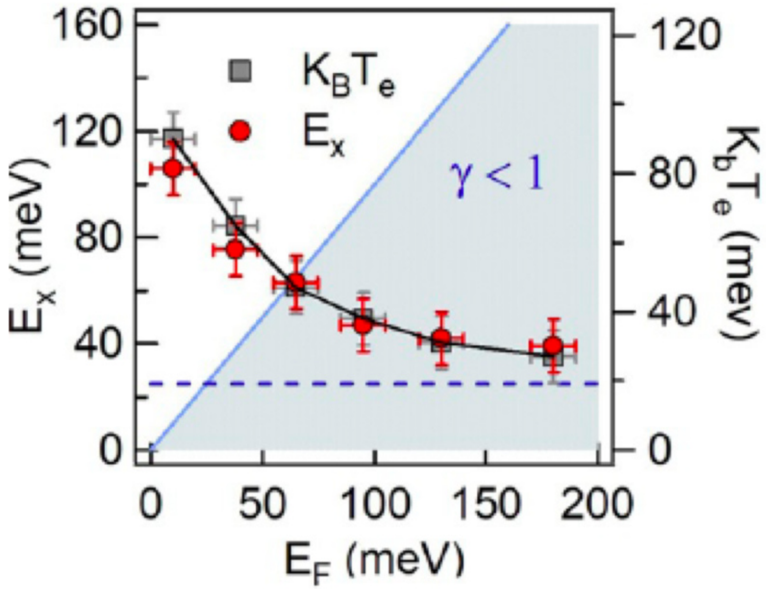


Chen, Sjakste et al, PNAS 117, 21962-21967 (2020)

Photoexcited electrons and 2D gas



$\gamma = T_e/E_F$

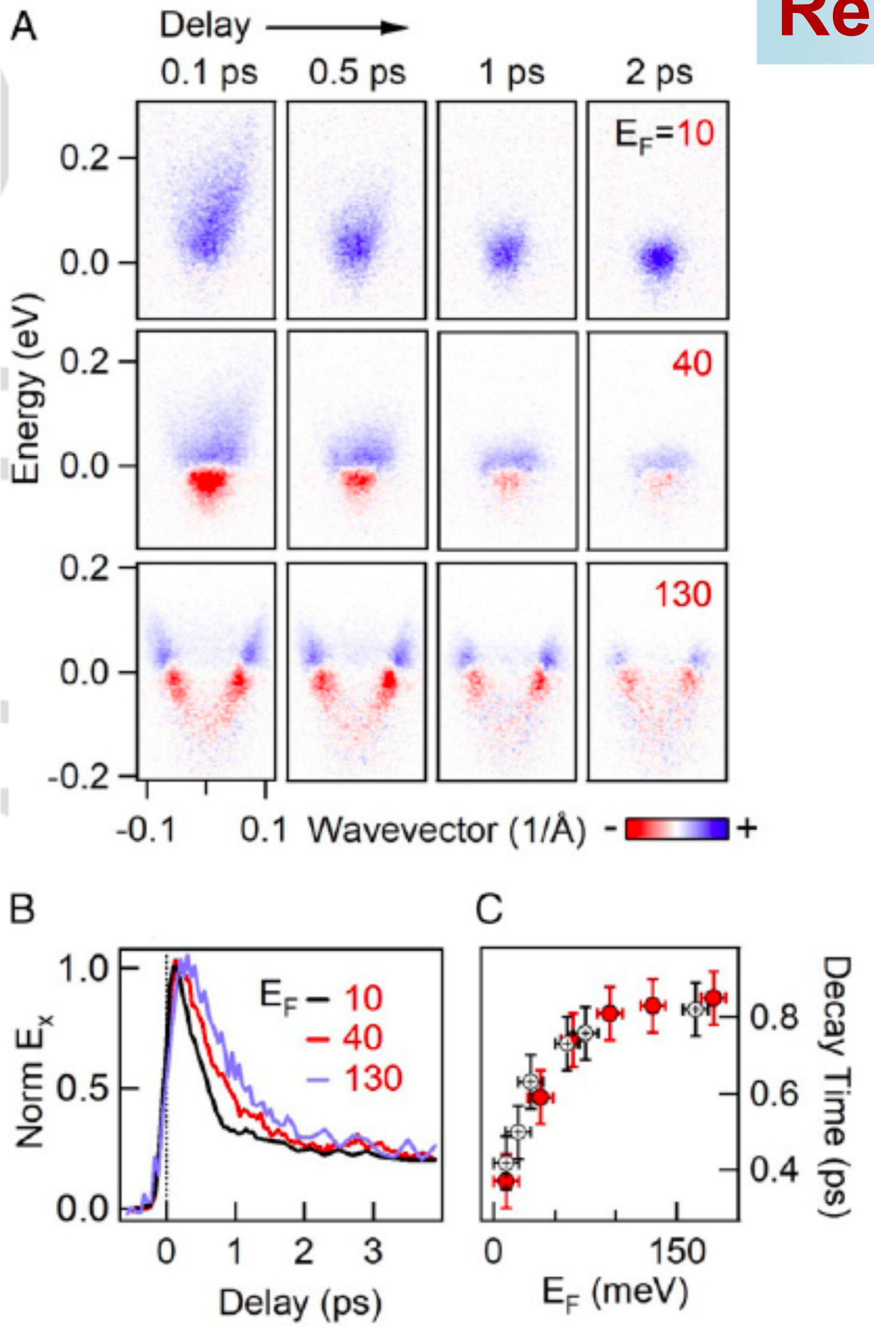


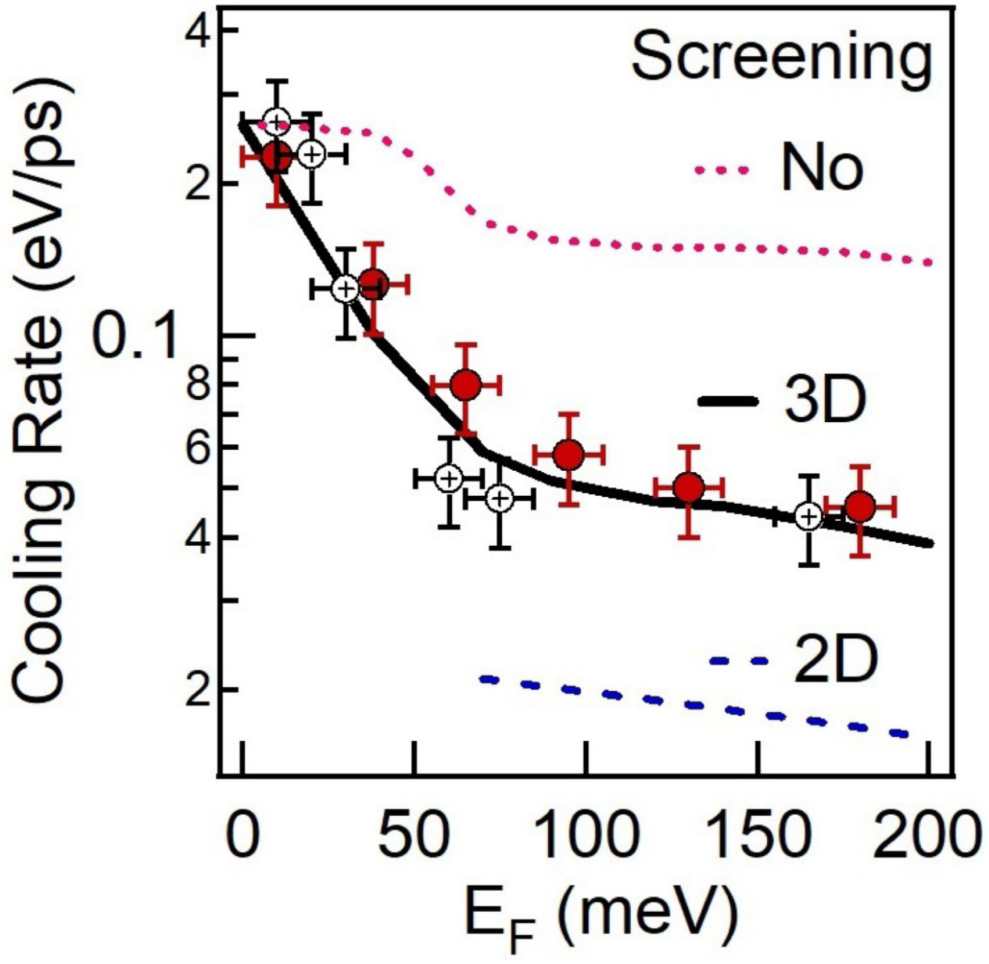
Low deposition: photoexcited electrons are non-thermal

High deposition: photoexcited electrons are thermalized

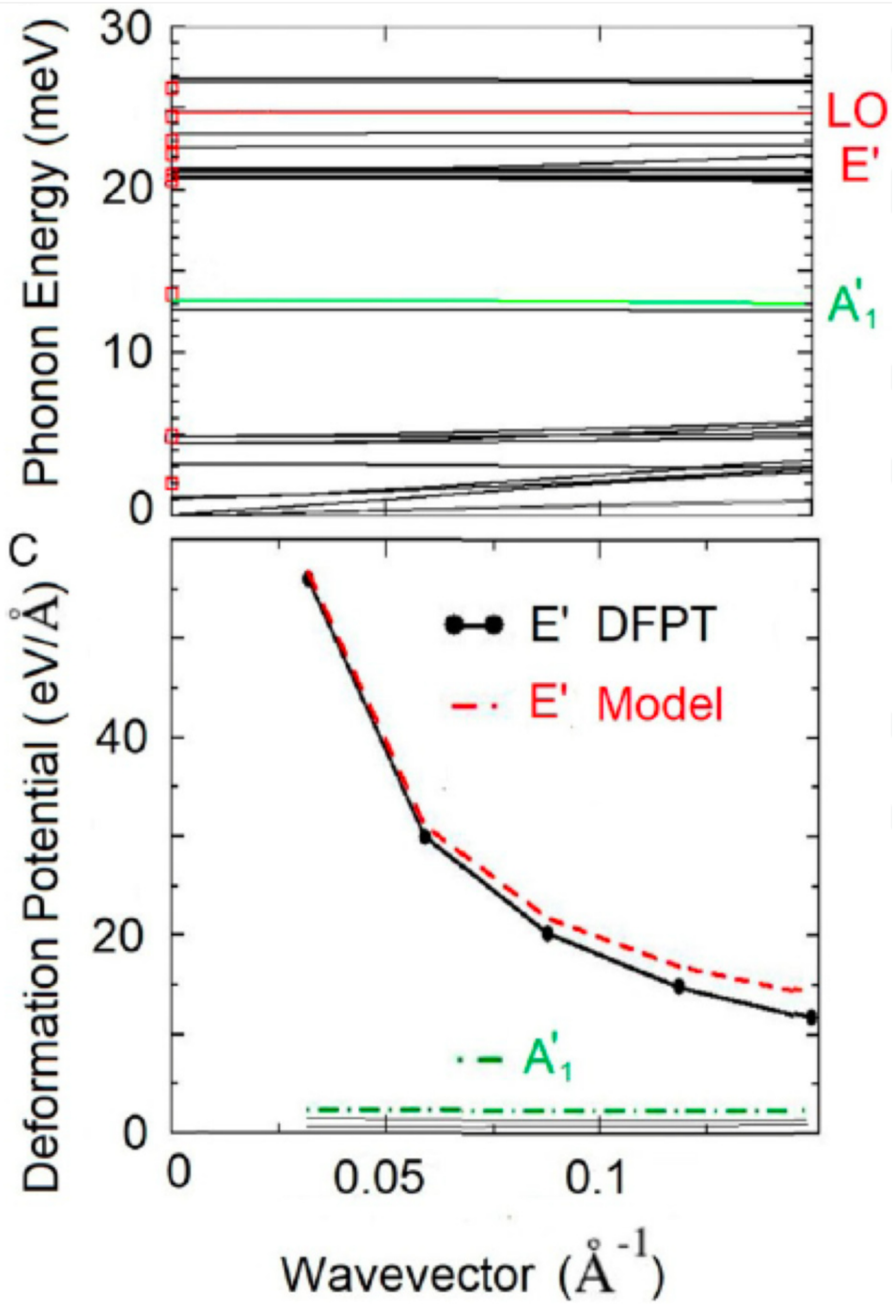
Chen, Sjakste et al, PNAS 117, 21962-21967 (2020)

Relaxation dynamics of photoexcited electrons





Chen, Sjakste et al, PNAS 117, 21962-21967 (2020)



Calculation: screened Fröhlich interaction

3D model:

$$|g^{3D}_{fr}(\mathbf{q})| = \frac{4\pi e^2}{V\epsilon_{bulk}|\mathbf{q}|}\sum_s\sum_{\lambda'}\frac{q_{\lambda'}}{|\mathbf{q}|}Z_{\lambda'\lambda s}\mathbf{e}^s_\lambda(\mathbf{q})/\sqrt{2M_s\omega_\mathbf{q}}$$

Vogl, PRB 13 (1976).

3D screening:

$$\epsilon^{scr}_{bulk} = \epsilon_{bulk}\left(1+\frac{(q_0^{3D})^2}{q^2}\right)$$

Thomas-Fermi

2D model:

$$|g^{2D}_{fr}(\mathbf{q})| = \frac{C_Z}{e^0_{eff}+r_{eff}|\mathbf{q_p}|}$$

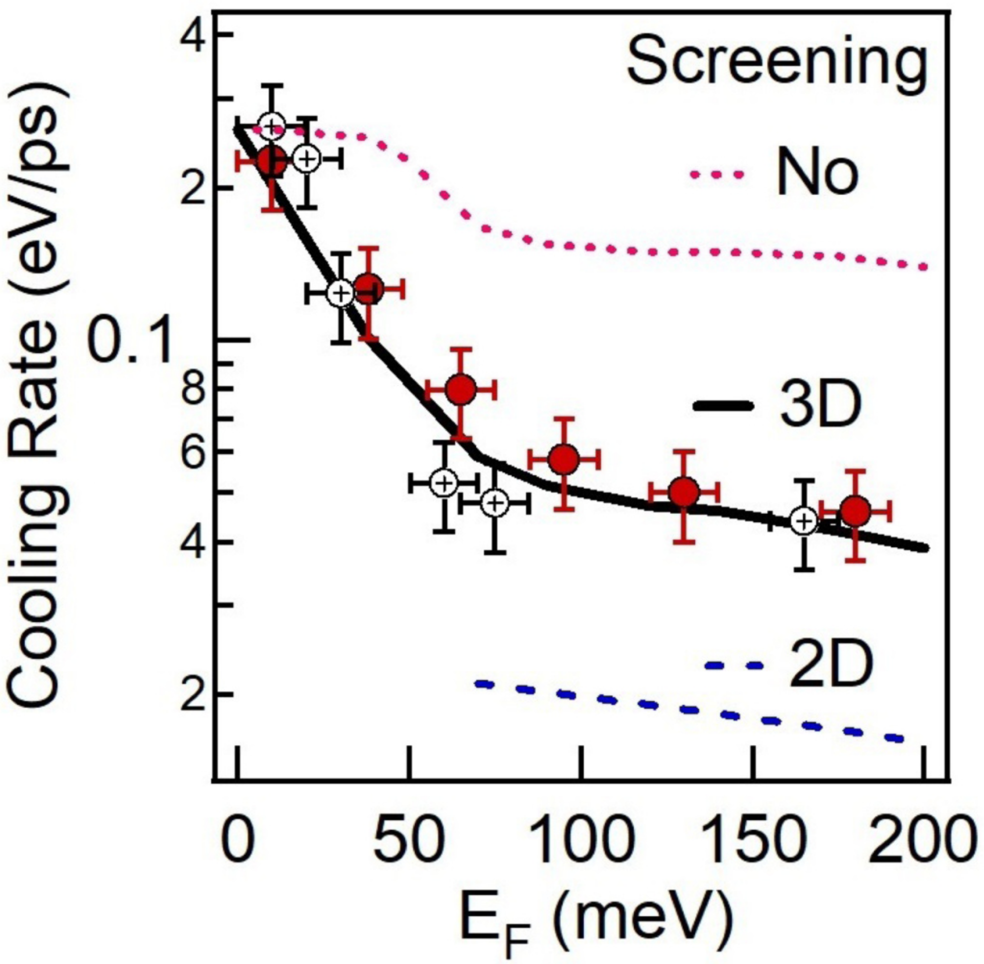
$$C_Z = \frac{2\pi e^2}{A}\times\sum_s\sum_{\lambda'}\frac{q_{\lambda'}}{|\mathbf{q_p}|}Z_{\lambda'\lambda s}\mathbf{e}^s_\lambda(\mathbf{q})/\sqrt{2M_s\omega_\mathbf{q}}$$

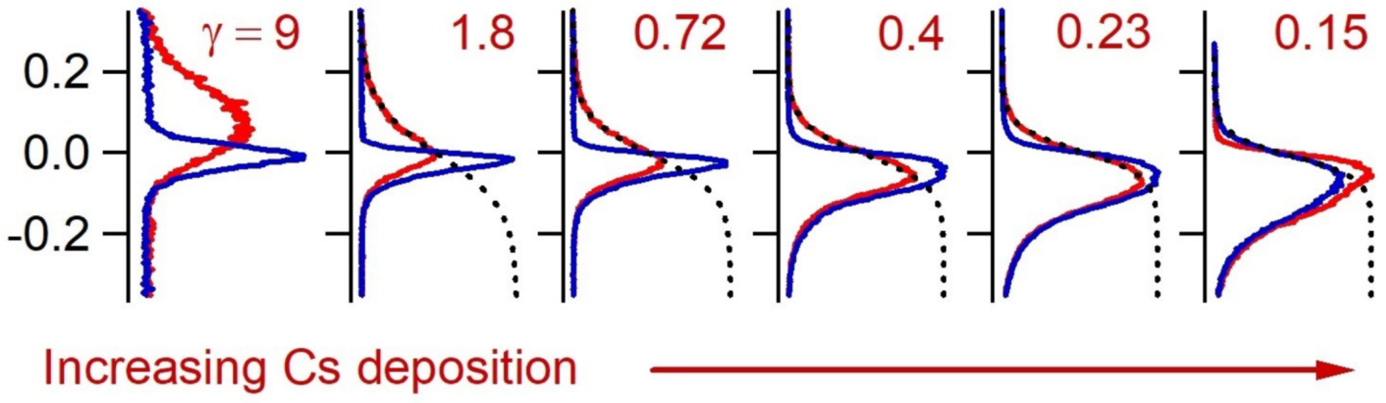
Sohier, Calandra, Mauri, PRB 94 (2016)

2D screening:

Stern, PRL 18, 546 (1967)

Energy transfer in doped InSe





$$\gamma = T_e / E_F$$

Remote coupling of electrons to 3D phonons

Chen, Sjakste et al, PNAS 117, 21962-21967 (2020)

Conclusion

Dynamics of relaxation of highly excited (or « hot »?) electrons in Si and in InSe

- *The concept of hot electron ensemble allowed us to interpret the relaxation times of highly excited electrons in silicon.*
- *In Si, energy loss rate is determined by the DOS of the final electronic states. Negligible temperature dependence.*
- *In InSe, at excess energies below 0.7 eV, energy loss rate is determined by Fröhlich coupling.*
- *Doped InSe:*
  - *screening of polar coupling*
  - *remote coupling to 3D phonons*

Thank you for your attention!