Nonequilibrium dynamics of crystal lattices

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Christian-Albrechts-Universität zu Kiel

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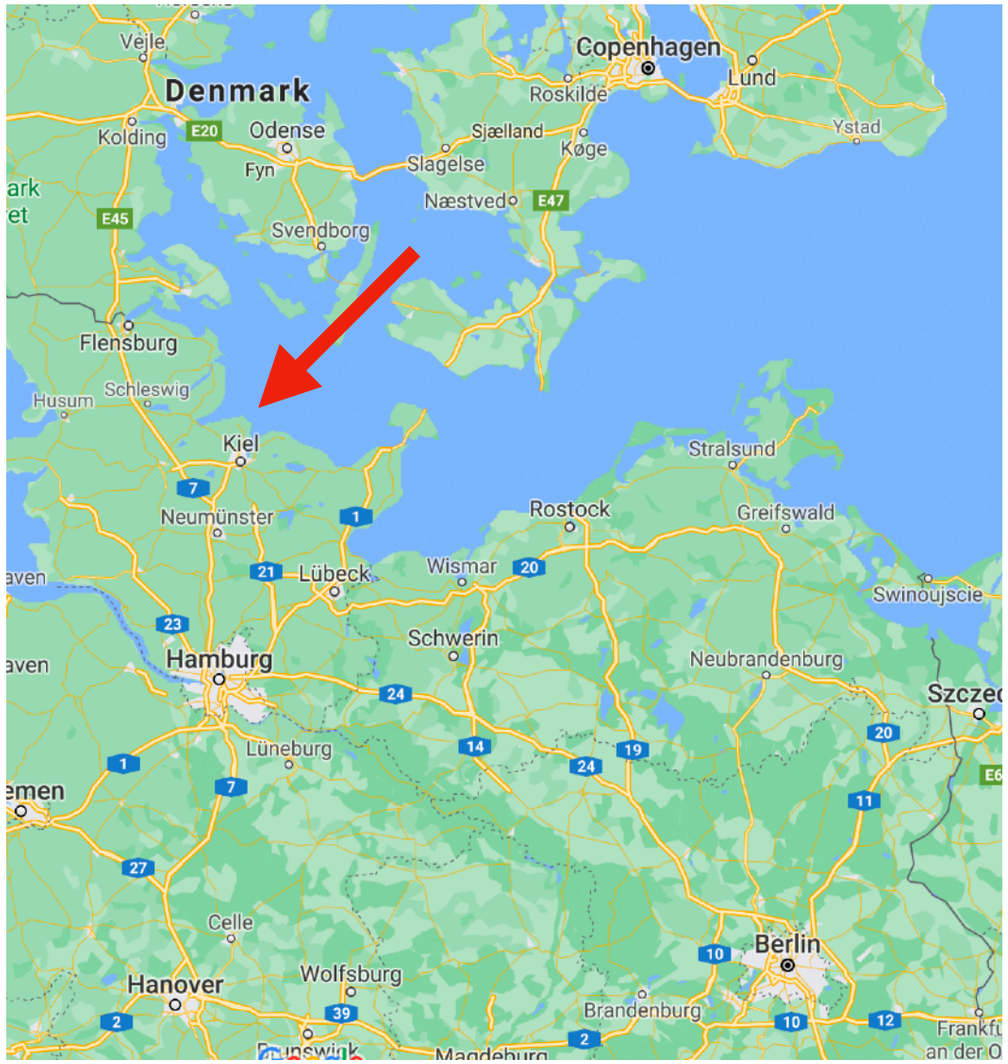
The University of Kiel (founded in 1665)

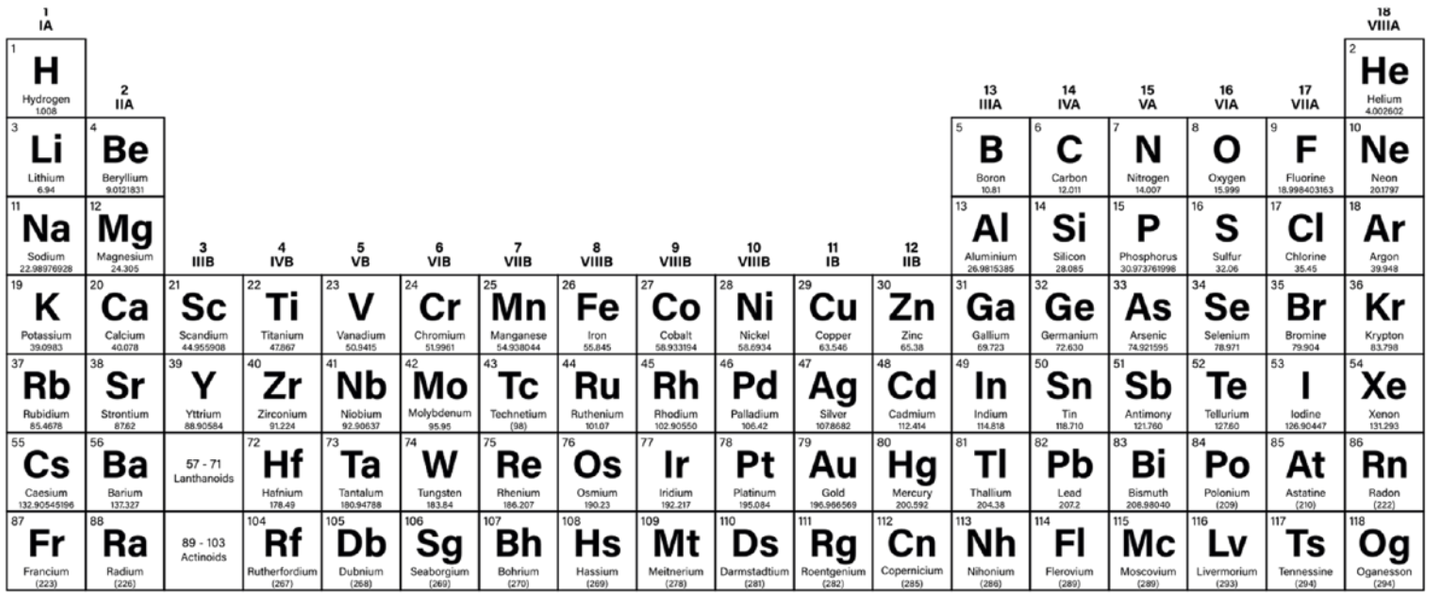


The paradigm of first-principles electronic-structure theory

The Schrödinger equation:

$$\hat{\mathcal{H}}\,\Psi_s(r,R) = E_s\,\Psi_s(r,R)$$

…a single equation for the whole of matter



Molecules

The chemical universe database (GDB-17) [1]

10^{11} entries

(Inorganic) Crystalline solids

The Karlsruhe Inorganic crystal-structure database (ICSD) [2]

200.000 entries

[1] J. Chem. Inf. Model. 52, 2864 (2012)

[2] icsd.fiz-karlsruhe.de

The paradigm of <u>first-principles</u> electronic-structure theory

electrons + nuclei

The Schrödinger equation:

$$\hat{\mathcal{H}}\,\Psi_s(r,R) = E_s\,\Psi_s(r,R)$$

…a single equation for the whole of matter

Hamiltonian operator for coupled electrons and nuclei (a differential operator)

Energy of the system in its s-th state (a scalar)

Wave function of electrons and nuclei (a function of coordinates)

$\Psi_s(r,R)$ **encodes all information (and all properties) about of a system of electrons and nuclei**

Example: the H atom

$$\psi_{n\ell m}(r,\theta,\phi) = \sqrt{\left(\frac{2}{na_0^*}\right)^3 \frac{(n-\ell-1)!}{2n(n+\ell)!}} e^{-\rho/2}\rho^\ell L_{n-\ell-1}^{2\ell+1}(\rho) Y_\ell^m(\theta,\phi)$$

Any other material: too complex! → **Approximations are needed!**

The paradigm of <u>first-principles</u> electronic-structure theory

The Born-Oppenheimer approximation:

$$\Psi_s(r, R) \simeq \psi_\nu(r; R)\chi_{\nu s}(R)$$

electrons (fixed nuclei)

$$\hat{H}^{\mathrm{el}}\,\psi_\nu(r) = E^{\mathrm{el}}_\nu \psi_\nu(r)$$

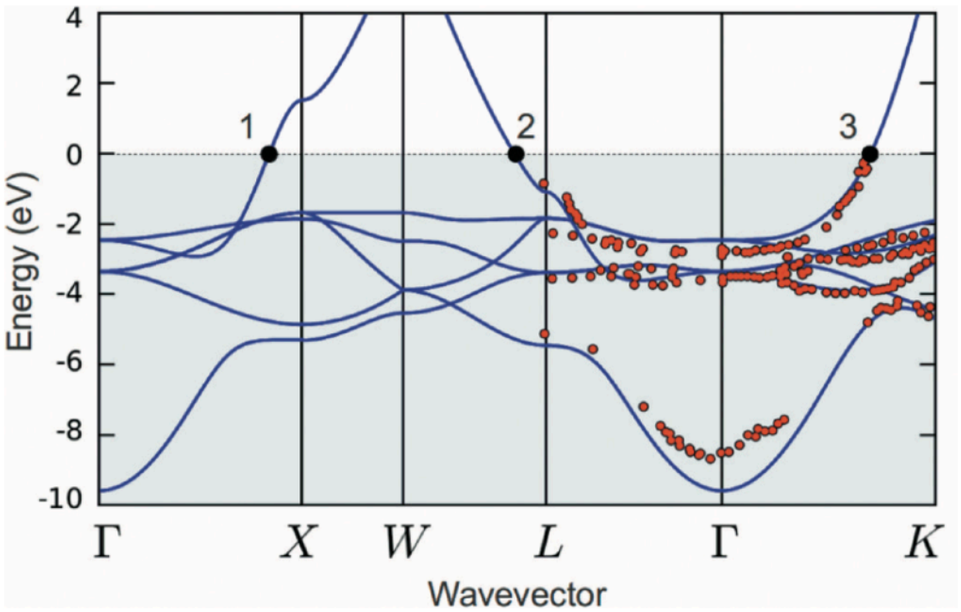
nuclei (fixed electrons)

$$\hat{H}^{\mathrm{nuc}}\,\chi_{\nu s}(R) = E_s\,\chi_{\nu s}(R)$$

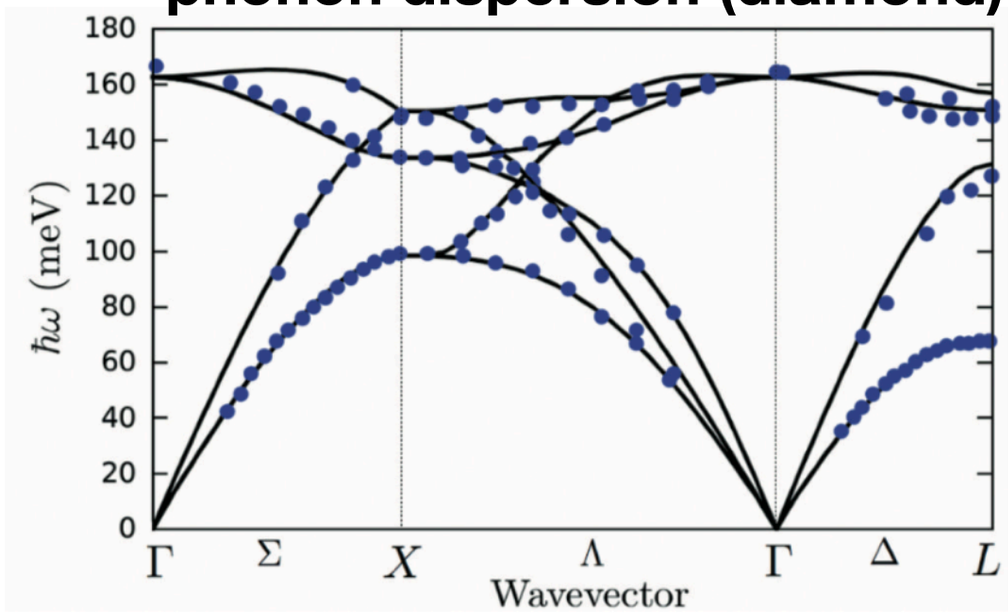
Density functional theory (DFT): $n(r) \Leftrightarrow \Psi$

Hohenberg and Kohn, Phys. Rev. (1964)
Kohn and Sham, Phys. Rev. (1965)

Electron band structure (Copper)



phonon dispersion (diamond)



Figs.: Giustino, Materials Modelling using Density Functional Theory (2014)

Exp.: [1] Courths, and Hüfner, Phys. Rep. 112, 53 (1984)
[2] Warren et al., Phys. Rev. 158, 805 (1967)

Where do electron-phonon interactions become important?

Charge transport:

- Ohm's law: dissipation of electronic currents
- Conventional superconductivity: dissipationless conduction

Energy conversion:

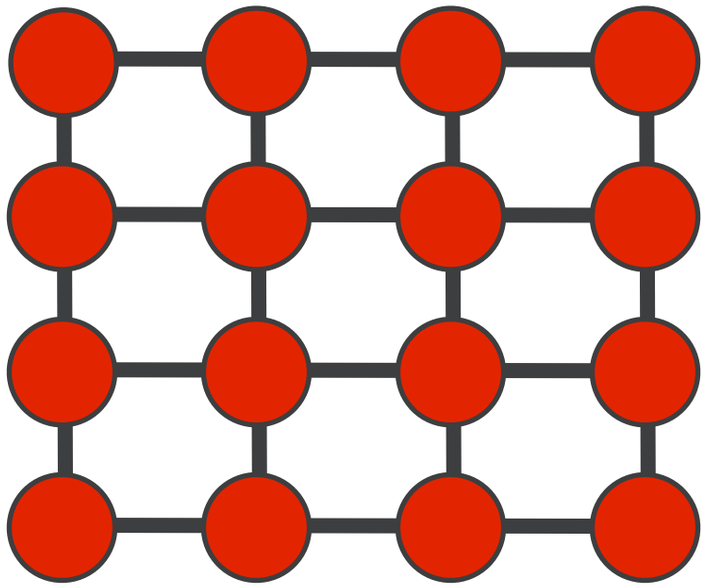
- Phonon assisted optical absorption (silicon photovoltaic)
- Time-scales of hot-carrier dynamics and recombination
- Thermoelectricity (Thermal conductivity)

Designing and understanding new functions of matter

- Electron-phonon quasiparticles (polarons)
- Ultrafast dynamics of electrons and phonons

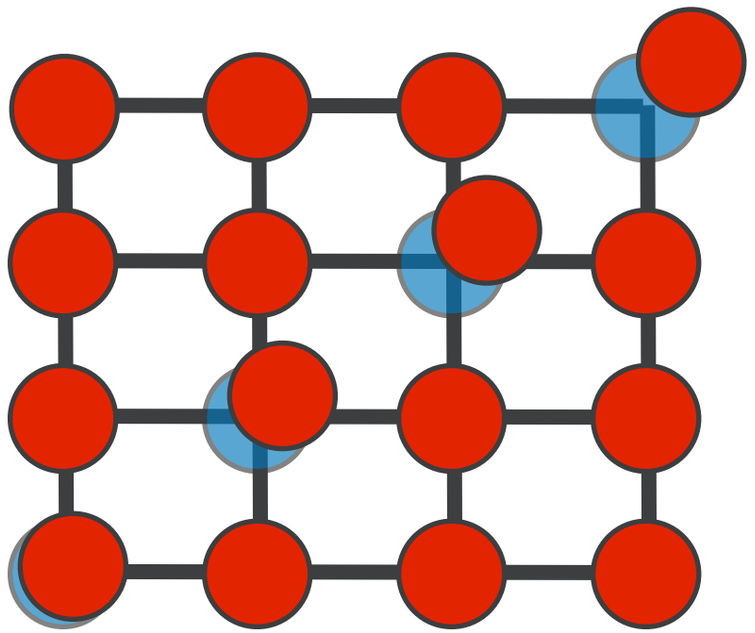
Electron-phonon interactions in solids

Nuclei at equilibrium



Potential: V

Atomic displacement due to a phonon



Perturbed potential: $V + \Delta_{\mathbf{q}\nu} V$ (t)

Electron-phonon coupling matrix elements:

$$g^{\mathrm{e-ph}}_{mn\nu}(\mathbf{k},\mathbf{q}) = \langle \psi_{m\mathbf{k}+\mathbf{q}} | \Delta_{\mathbf{q}\nu} V_{\mathrm{KS}} | \psi_{n\mathbf{k}} \rangle$$

Transition probability amplitude

final electronic state

phonon perturbation

initial electronic state

Modern theory of the electron-phonon interaction

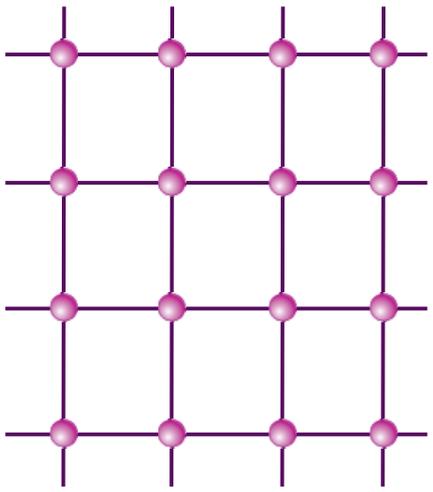
Many-body perturbation theory: $V + \Delta_{\mathbf{q}\nu} V$ (t)

electronic potential — phonon perturbation

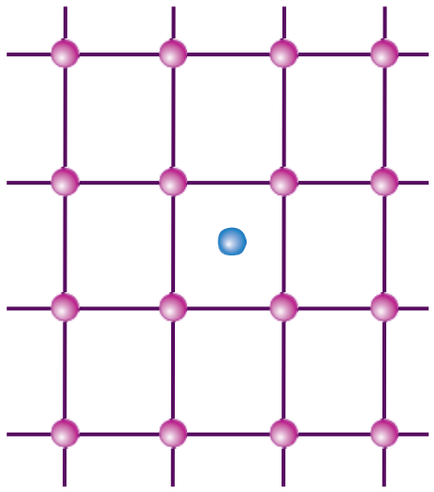
Description	Expression
Electronic charge density	$\langle \hat{n}_{\mathrm{e}}(1) \rangle = -i\hbar \sum_{\sigma_1} G(11^+)$
Nuclear charge density	$\langle \hat{n}_{\mathrm{n}}(\mathbf{r}t) \rangle = n^0_{\mathrm{n}}(\mathbf{r}) - (i\hbar/2) \sum_{\kappa p, \alpha\alpha'} Z_\kappa \partial^2 \delta(\mathbf{r} - \boldsymbol{\tau}^0_{\kappa p}) / \partial r_\alpha \partial r_{\alpha'} D_{\kappa\alpha p, \kappa\alpha' p}(t^+ t)$
Total electrostatic potential	$V_{\mathrm{tot}}(1) = \int d2 v(12) [\langle \hat{n}_{\mathrm{e}}(2) \rangle + \langle \hat{n}_{\mathrm{n}}(2) \rangle]$
Equation of motion, electrons	$[i\hbar\partial/\partial t_1 + (\hbar^2/2m_{\mathrm{e}})\nabla^2(1) - V_{\mathrm{tot}}(1)] G(12) - \int d3 \Sigma(13) G(32) = \delta(12)$
Equation of motion, nuclei	$\sum_{\kappa'' \alpha'' p''} [M_\kappa \omega^2 \delta_{\kappa\alpha p, \kappa'' \alpha'' p''} - \Pi_{\kappa\alpha p, \kappa'' \alpha'' p''}(\omega)] D_{\kappa'' \alpha'' p'', \kappa' \alpha' p'}(\omega) = \delta_{\kappa\alpha p, \kappa' \alpha' p'}$
Electron self-energy	$\Sigma(12) = i\hbar \int d(34) G(13) \Gamma(324) [W_{\mathrm{e}}(41^+) + W_{\mathrm{ph}}(41^+)]$
Screened Coulomb, electrons	$W_{\mathrm{e}}(12) = v(12) + \int d(34) v(13) P_{\mathrm{e}}(34) W_{\mathrm{e}}(42)$
Electronic polarization	$P_{\mathrm{e}}(12) = -i\hbar \sum_{\sigma_1} \int d(34) G(13) G(41^+) \Gamma(342)$
Electronic dielectric matrix	$\epsilon_{\mathrm{e}}(12) = \delta(12) - \int d(3) v(13) P_{\mathrm{e}}(32)$
Vertex	$\Gamma(123) = \delta(12)\delta(13) + \int d(4567) [\delta\Sigma(12)/\delta G(45)] G(46) G(75) \Gamma(673)$
Screened Coulomb, nuclei	$W_{\mathrm{ph}}(12) = \sum_{\kappa\alpha p, \kappa' \alpha' p'} \int d(34) \epsilon^{-1}_{\mathrm{e}}(13) \nabla_{3,\alpha} V_\kappa(\mathbf{r}_3 - \boldsymbol{\tau}^0_{\kappa p}) \times D_{\kappa\alpha p, \kappa' \alpha' p'}(t_3 t_4) \epsilon^{-1}_{\mathrm{e}}(24) \nabla_{4,\alpha'} V_{\kappa'}(\mathbf{r}_4 - \boldsymbol{\tau}^0_{\kappa' p'})$
Phonon self-energy	$\Pi_{\kappa\alpha p, \kappa' \alpha' p'}(\omega) = \sum_{\kappa'' p''} Z_\kappa Z_{\kappa''} (\partial^2 / \partial r_\alpha \partial r'_{\alpha'}) \times [\delta_{\kappa' p', \kappa'' p''} W_{\mathrm{e}}(\mathbf{r}, \mathbf{r}', \omega) - \delta_{\kappa p, \kappa' p'} W_{\mathrm{e}}(\mathbf{r}, \mathbf{r}', 0)]_{\mathbf{r} = \boldsymbol{\tau}^0_{\kappa p}, \mathbf{r}' = \boldsymbol{\tau}^0_{\kappa'' p''}}$

Giustino, Rev. Mod. Phys. (2017)

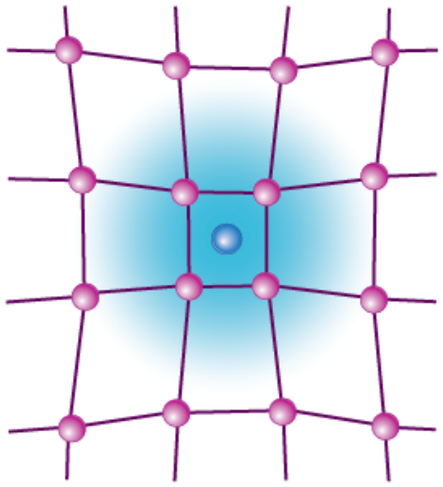
1. The concept of quasi-particles



lattice



external particle



quasi-particle

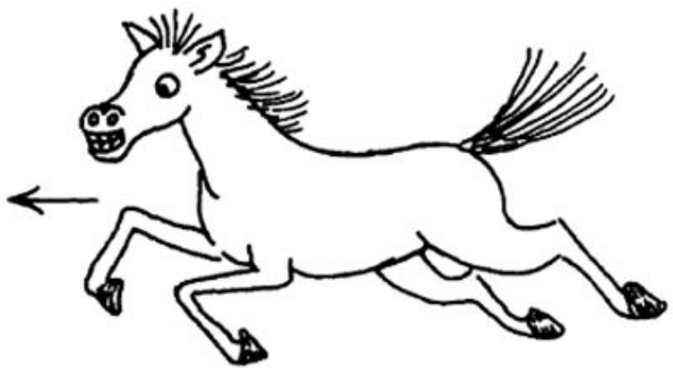
example:

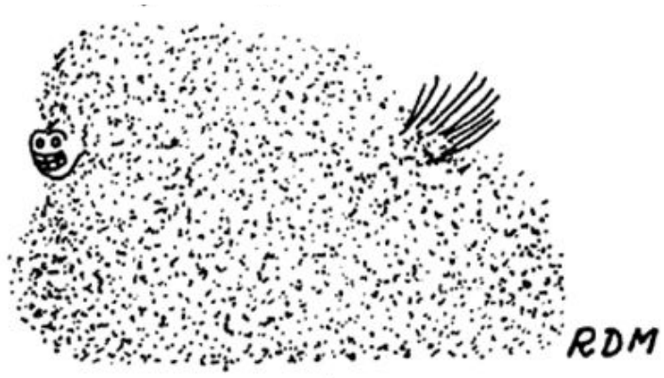
electron (hole) +
lattice distortion
= **polarons**

Quasi-particle energy: $\varepsilon_n^{\mathrm{QP}} = \varepsilon_n^{\mathrm{NI}} + \Sigma$

Energy of the non-interacting particle

Self-energy

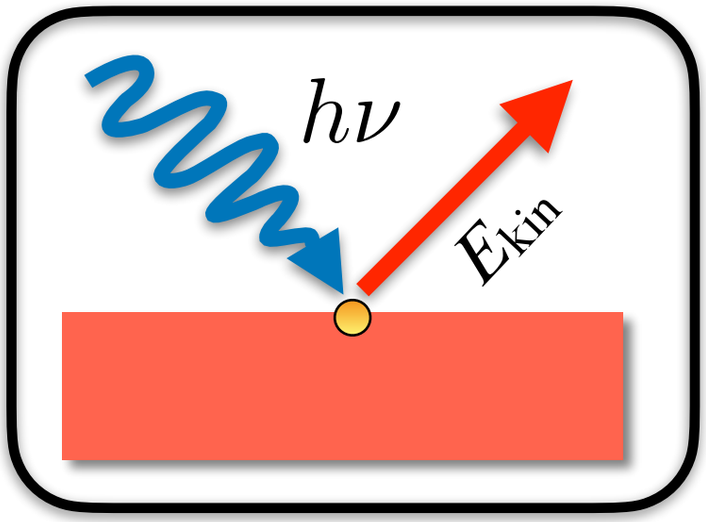




R. Mattuck

2. Relation to photoemission spectroscopy

photoemission spectroscopy



Photoelectron current (Fermi's golden rule):

$$J_{\mathbf{k}}(\omega) = \sum_{s} |\langle \Psi_{\mathbf{k},s} | \Delta | \Psi_i \rangle|^2 \delta(\omega - \varepsilon_{\mathbf{k}} + \varepsilon_s)$$

- $\Psi_{\mathbf{k},s}$: final state
- Δ: perturbation
- Ψ_i: initial state
- $\delta(\omega - \varepsilon_{\mathbf{k}} + \varepsilon_s)$: energy conservation

$$= \sum_{ij} \Delta_{\mathbf{k}j} A_{ji}(\varepsilon_k - \omega) \Delta_{i\mathbf{k}}$$

[1] sudden approximation

Spectral function

$$A(\mathbf{k},\omega) = -\frac{1}{\pi} \sum_n \mathrm{Im}\, G_{n\mathbf{k}}(\omega)$$

Migdal (diagonal) approximation:

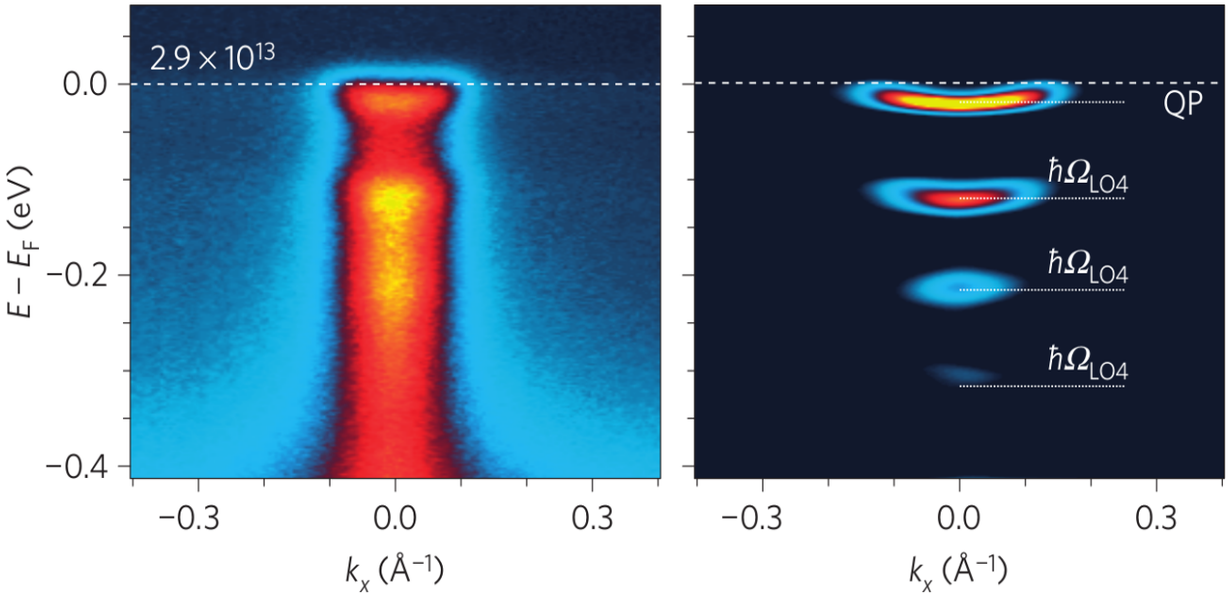
$$= \frac{1}{\pi} \sum_n \left| \mathrm{Im}[\omega - \varepsilon_{n\mathbf{k}} - \Sigma_{n\mathbf{k}}(\omega)]^{-1} \right|$$

[1] L. Hedin, et al., Phys. Rev. B **58**, 15565 (1998)

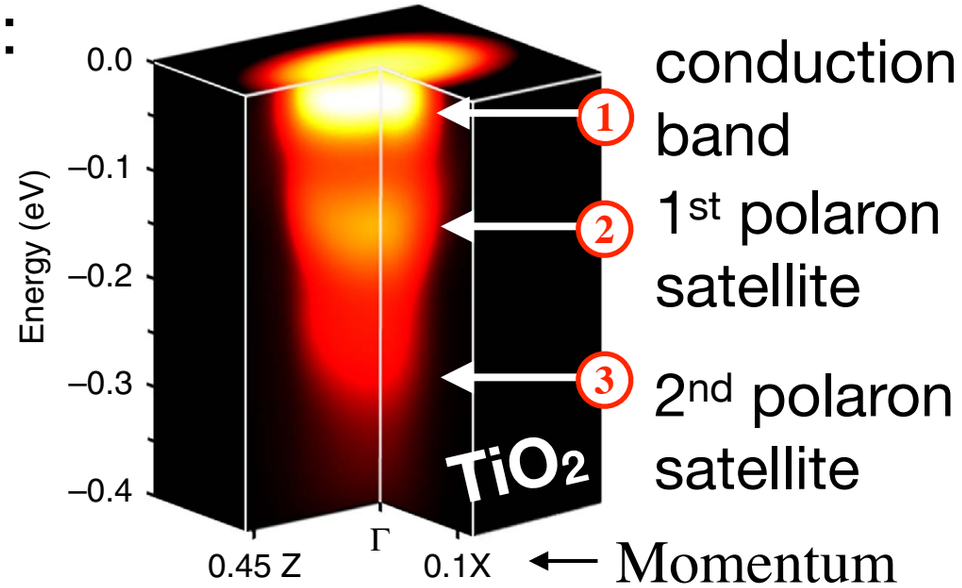
Polarons in angle-resolved photoemission spectroscopy

Polaron = electron (hole) dressed by a phonon cloud

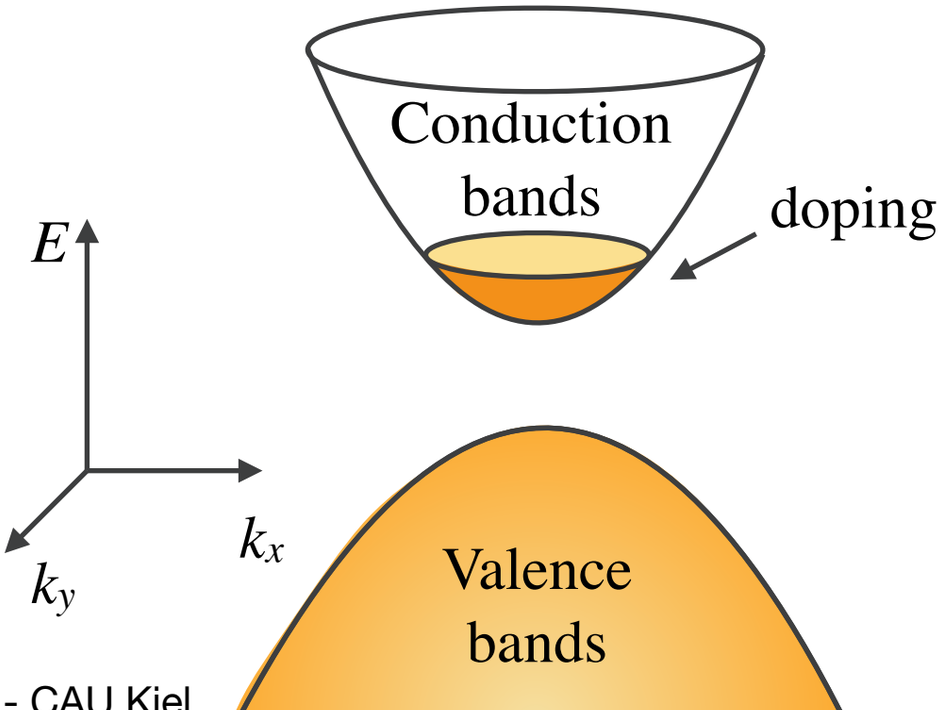
Polarons at the 2DEG at the $SrTiO_3$(001) surface:

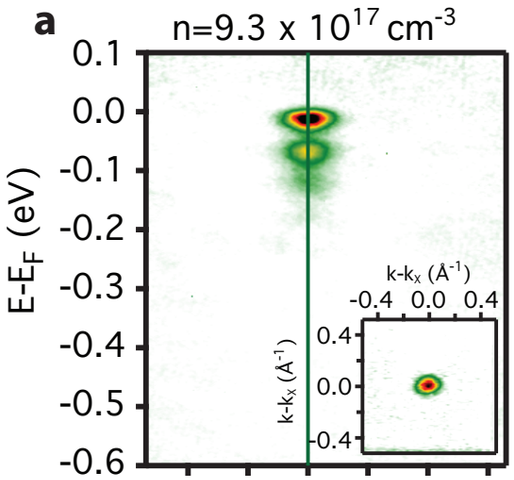


Wang et al., Nature Mater. **15**, 835 (2016)



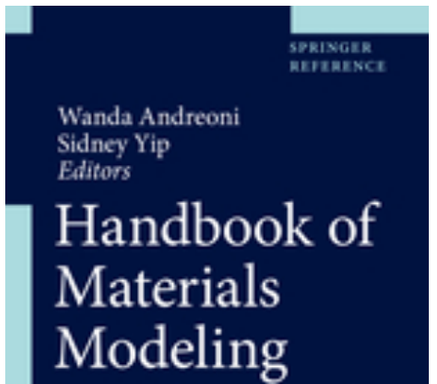
Verdi, FC, Giustino, Nature Comm. **8**, 15769 (2017)





Riley, FC, Verdi, et al. Nature Comm. **9**, 2305 (2018)

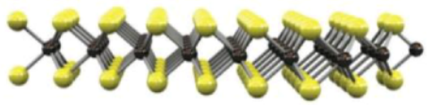
FC, Verdi, Giustino, Chapter in Handbook of Materials Modeling (2018)



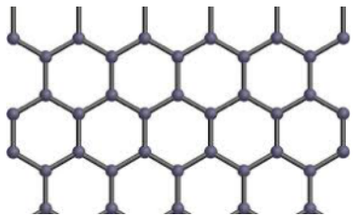
Nonequilibrium dynamics in two dimensions

Materials science in 2D

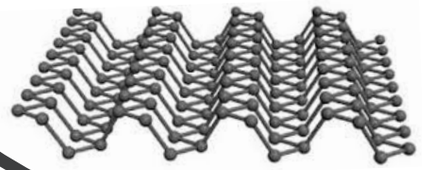
TMDCs



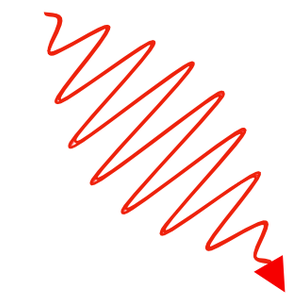
Graphene



phosphorene



Advent of femto-second lasers



$\Delta t < 10\,\mathrm{fs}$

$\Delta t < 10\,\mathrm{fs}$

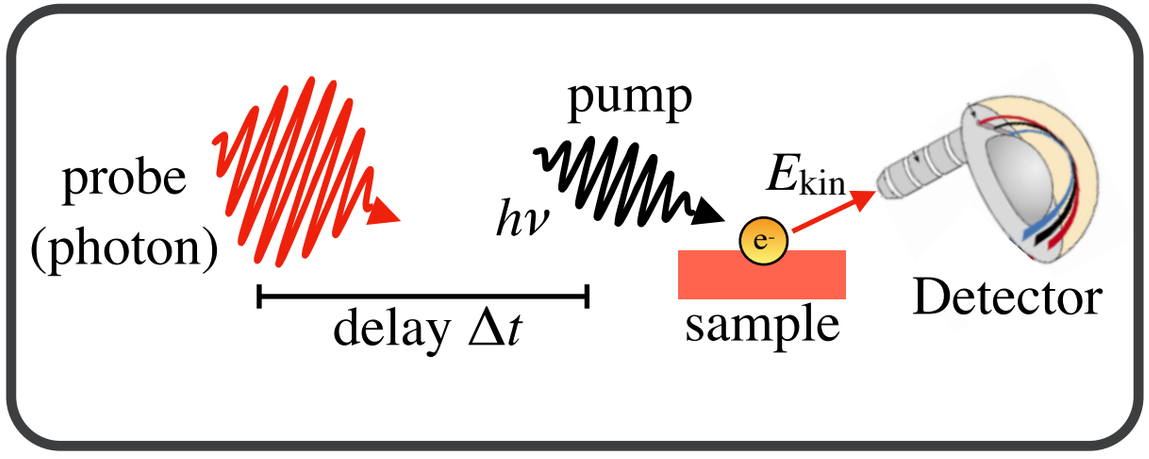
- Reduced dielectric screening
- Quantum-confinement in 2D
- Strong light-matter interactions
- Non-trivial topological properties

- Pump-probe spectroscopies
- Electron and phonon dynamics
- Light-driven phenomena

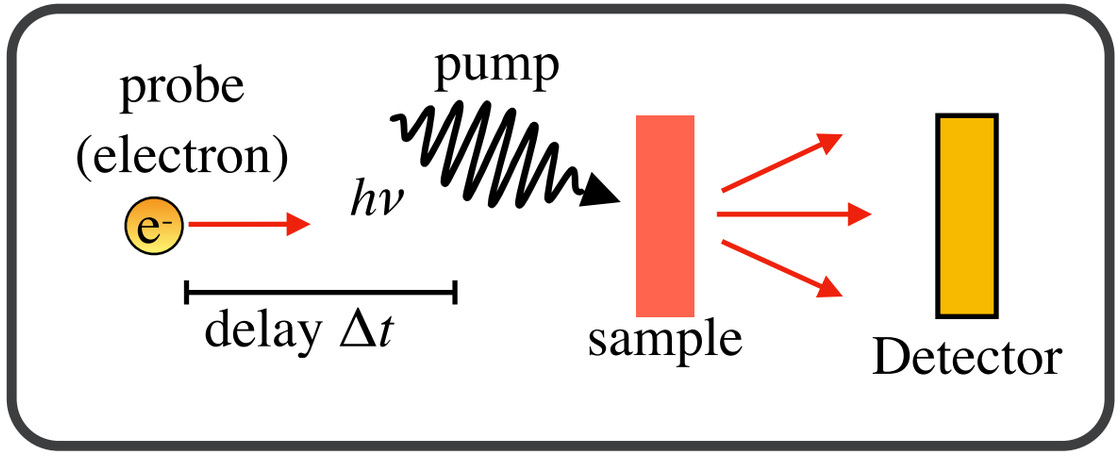
Properties on-demand using light pulses

Nonequilibrium dynamics in two dimensions

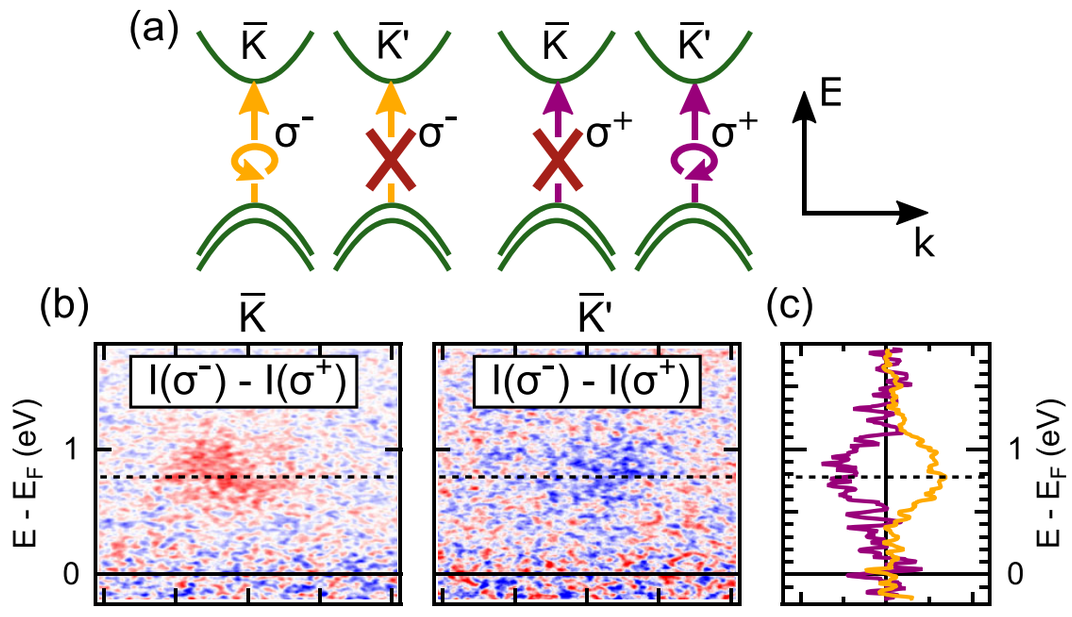
Time- and angle-resolved photoemission spectroscopy (trARPES)



Femto-second electron diffuse scattering (FEDS)

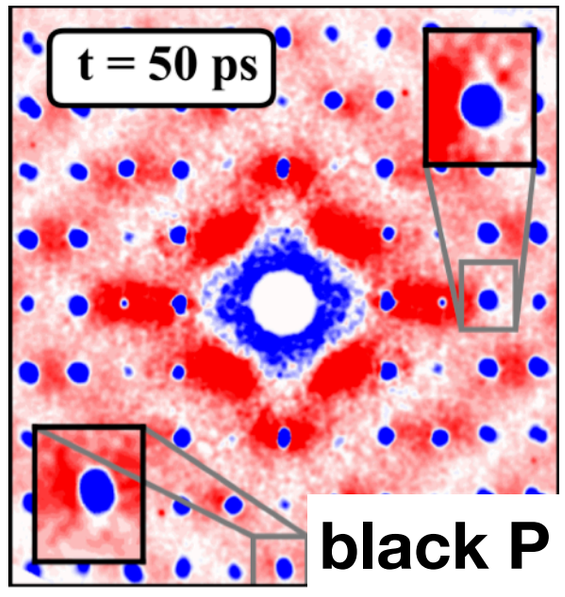


Circular Dichroism (WS_2)

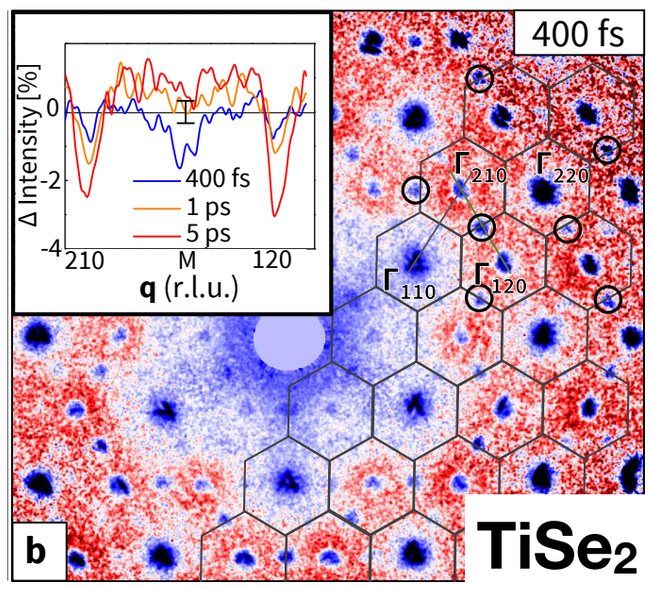


Beyer, Bauer, et al., PRL **123**, 236802 (2019)

Anisotropic lattice dynamic



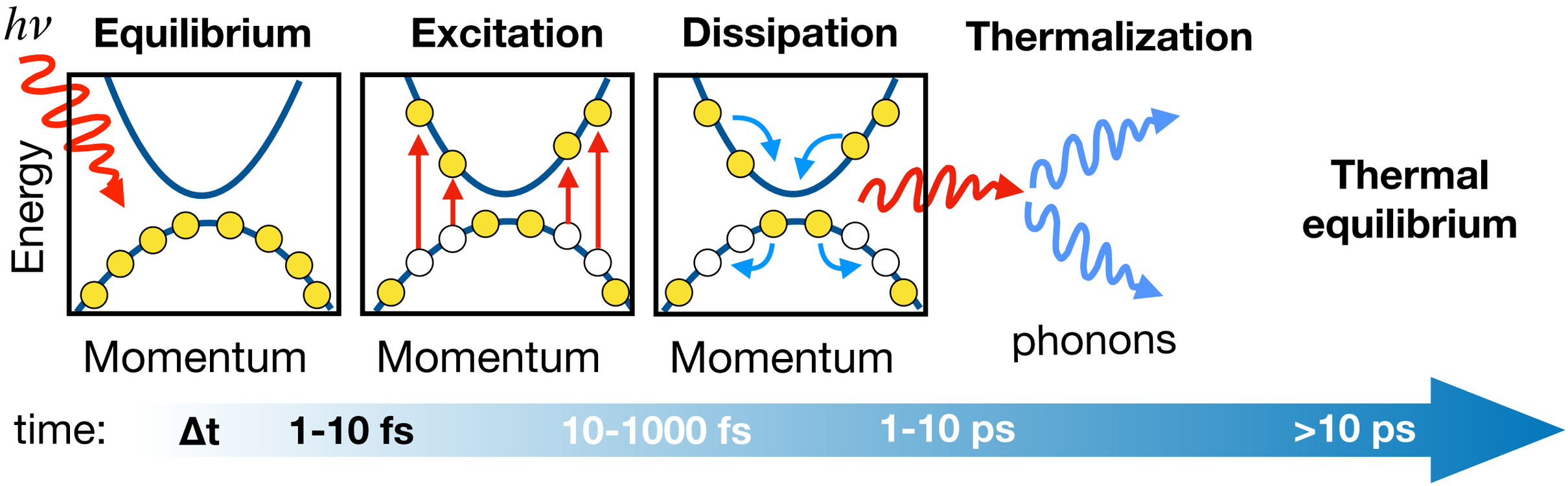
Seiler, FC, et al., arXiv:2006.12873 (2020)



Otto, Siwick, et al., arXiv:1912.03559 (2020)

What is the influence of phonons and electron-phonon interactions on the nonequilibrium dynamics?

Ultrafast dynamics from first principles



Which level of theory?

Nonequilibrium Green's functions (& DMFT)
Time-dependent density-function theory
Time-dependent Boltzmann equation
Quantum Master equation
Molecular dynamics / Path integrals
Non-thermal lattice models

Check-list:

- ☑ Electron / phonon dynamics
- ☑ Electron-phonon coupling
- ☑ Full momentum resolution

Equilibrium and nonequilibrium regimes in a quantum system

Distribution function (occupation number):

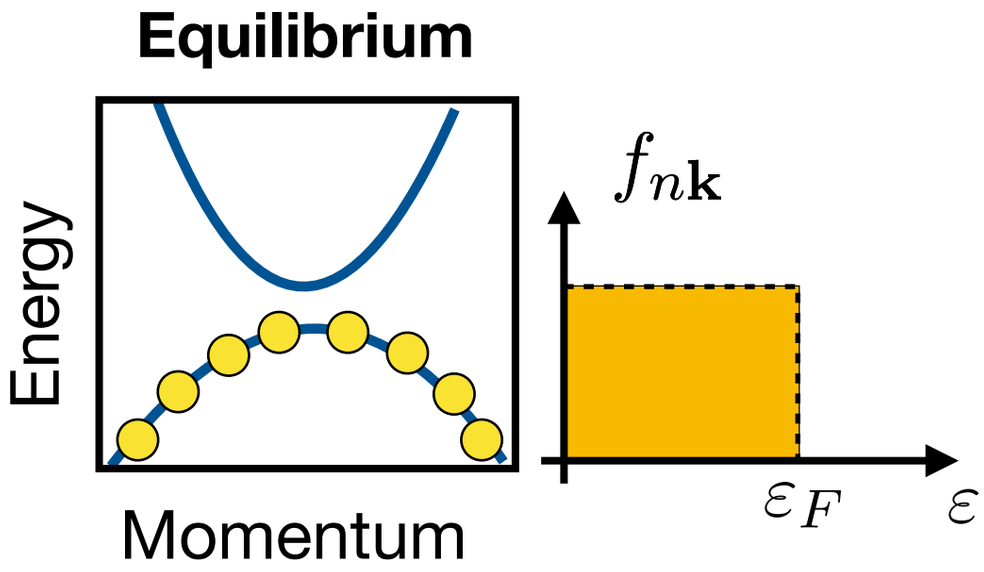
$f^{\sigma}_{n\mathbf{k}}$ Number of **electrons** in band n with momentum $\mathbf{k}$

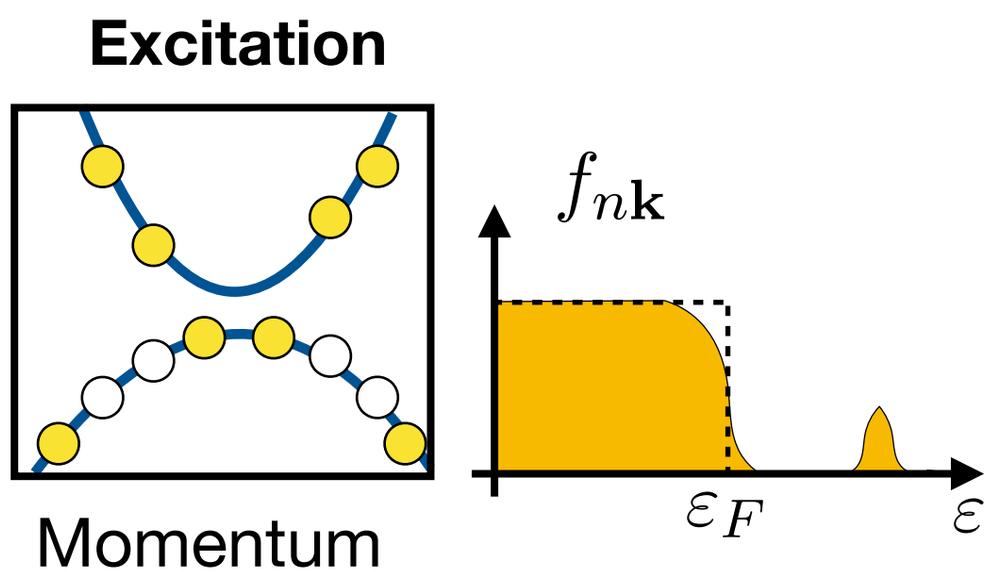
$n_{\mathbf{q}\nu}$ Number of **phonons** in branch ν with momentum $\mathbf{q}$

Equilibrium:

Fermi-Dirac statistics: $f^0_{n\mathbf{k}}(\mu,T) = [e^{(\varepsilon_{n\mathbf{k}}-\mu)/k_{\mathrm{B}}T}+1]^{-1}$

Bose-Einstein statistics: $n^0_{\mathbf{q}\nu}(T) = [e^{\hbar\omega_{\mathbf{q}\nu}/k_{\mathrm{B}}T}-1]^{-1}$





Out of equilibrium: $f_{n\mathbf{k}} \neq f^0_{n\mathbf{k}}$ and / or $n_{\mathbf{q}\nu} \neq n^0_{\mathbf{q}\nu}$

The Boltzmann equation in solid-state physics

distribution function

collision integral

$$\frac{\partial f}{\partial t} = \Gamma_{\text{collisions}}$$

+ other terms (fields, etc.)

(In a gas:)

Equilibrium is re-established via collisions

Books:

Ziman, Electrons and phonons, Oxford Universisty Press (1960)
Hang, Jauho, Quantum Kinetics in Transport and Optics of Semiconductors, Springer (1996)
Bonitz, Quantum Kinetic Theory (1998)

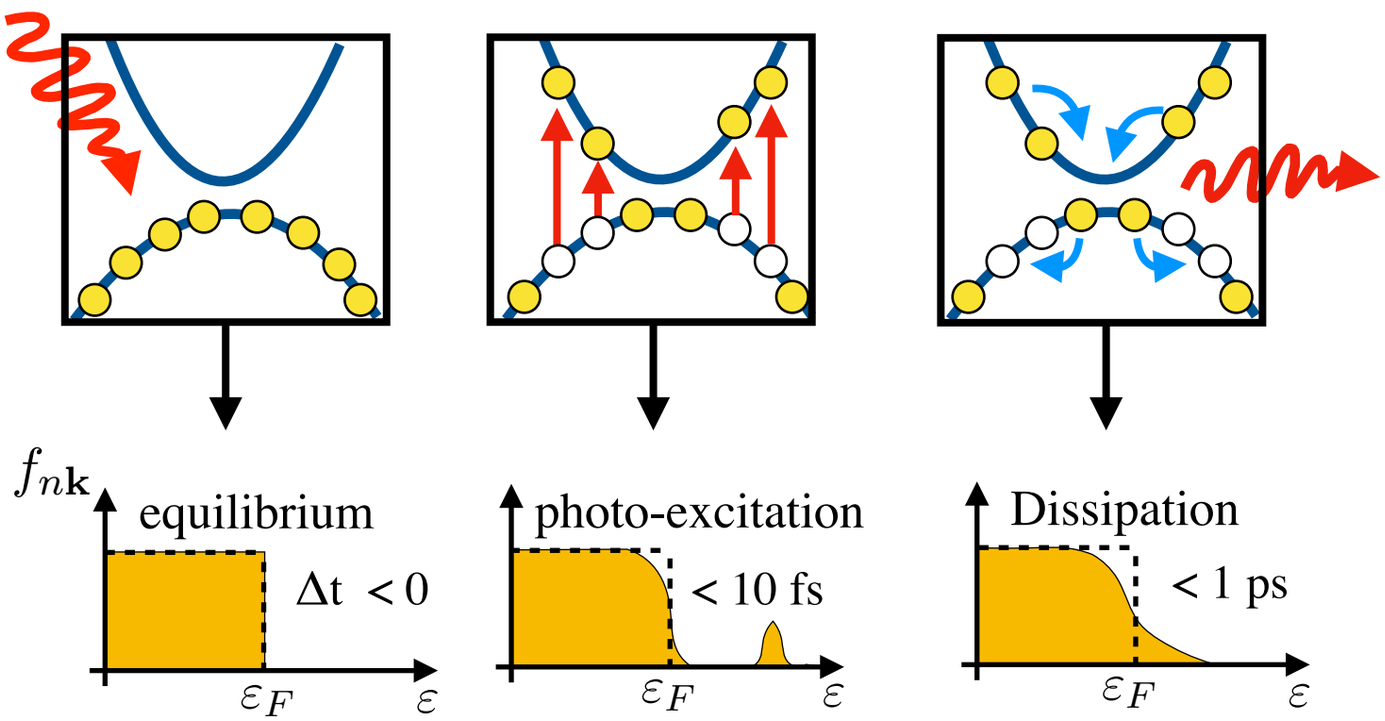
Charge and thermal Transport:

Poncé, Li, Reichard, Giustino, Rep. Prog. Phys. 83, 036501 (2019)
Li, Carrete, Katcho, Mingo, Comp. Phys. Comm.185, 1747 (2014)
Mizokami, Togo, Tanaka Phys. Rev. B 97, 224306, (2018)
Chaput, Phys. Rev. Lett 110, 265506 (2013)
Togo, Chaput, Tanaka, Phys. Rev. B 91, 094306 (2015)

Ultrafast dynamics:

Sadasivam, Chan, Darancet, Phys. Rev. Lett. 119, 136602 (2017)
Bernardi, Eur. Phys. J. B 89, 239 (2016)
Jhalani, Zhou, Bernardi, Nano Letters 17, 5012 (2017)
FC, J. Phys. Chem. Lett. 12, 1274 (2021)

Time-dependent Boltzmann equation



Nonequilibrium states are described by

$f^{\sigma}_{n\mathbf{k}}$ $n_{\mathbf{q}\nu}$

time-dependence of $f^{\sigma}_{n\mathbf{k}}$ $n_{\mathbf{q}\nu}$

time-dep. Boltzmann eq.:

electron-phonon

electron-photon (dipole int.)

electron-electron

$$\frac{\partial f_{n\mathbf{k}}}{\partial t} = I^{\mathrm{e-ph}}_{n\mathbf{k}}[f,n] + I^{\mathrm{light}}_{n\mathbf{k}}[f] + I^{\mathrm{e-e}}_{n\mathbf{k}}[f]$$

$$\frac{\partial n_{\mathbf{q}\nu}}{\partial t} = I^{\mathrm{e-ph}}_{\mathbf{q}\nu}[f,n] + I^{\mathrm{ph-ph}}_{\mathbf{q}\nu}[n]$$

phonon-electron

phonon-phonon

First-principles approach to electron and lattice dynamics

$$\frac{\partial f_{n\mathbf{k}}}{\partial t} = I^{\mathrm{e-ph}}_{n\mathbf{k}}[f,n]$$

$$\frac{\partial n_{\mathbf{q}\nu}}{\partial t} = I^{\mathrm{e-ph}}_{\mathbf{q}\nu}[f,n] + I^{\mathrm{ph-ph}}_{\mathbf{q}\nu}[n]$$

Boltzmann equation for the electron and phonon distribution function

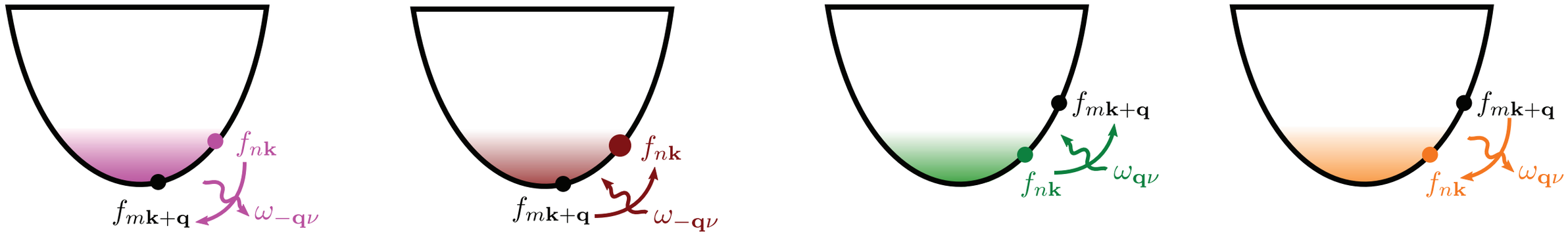
Poncé, et. al, Rep. Prog. Phys. (2019)

Example: electron-phonon scattering rate

$$I^{\mathrm{e-ph}}_{n\mathbf{k}}[f,n] = \frac{2\pi}{\hbar}\sum_{m\nu}\int\frac{d\mathbf{q}}{\Omega_{\mathrm{BZ}}}|g_{mn\nu}(\mathbf{k},\mathbf{q})|^2$$

electron-phonon coupling matrix element

$$\times\Big\{(1-f_{n\mathbf{k}})f_{m\mathbf{k+q}}\delta(\varepsilon_{n\mathbf{k}}-\varepsilon_{m\mathbf{k+q}}+\hbar\omega_{\mathbf{q}\nu})(1+n_{\mathbf{q}\nu})$$
$$+(1-f_{n\mathbf{k}})f_{m\mathbf{k+q}}\delta(\varepsilon_{n\mathbf{k}}-\varepsilon_{m\mathbf{k+q}}-\hbar\omega_{\mathbf{q}\nu})n_{\mathbf{q}\nu}$$
$$-f_{n\mathbf{k}}(1-f_{m\mathbf{k+q}})\delta(\varepsilon_{n\mathbf{k}}-\varepsilon_{m\mathbf{k+q}}-\hbar\omega_{\mathbf{q}\nu})(1+n_{\mathbf{q}\nu})$$
$$-f_{n\mathbf{k}}(1-f_{m\mathbf{k+q}})\delta(\varepsilon_{n\mathbf{k}}-\varepsilon_{m\mathbf{k+q}}+\hbar\omega_{\mathbf{q}\nu})n_{\mathbf{q}\nu}\Big\}$$



Ultrafast dynamics from first principles

WANNIER90

Wannier-function interpolation

G. Pizzi et al.,
J. Phys. Cond. Matt. 32, 165902 (2020)

EPW

Electron-phonon coupling

S. Poncé et al.,
Comp. Phys. Comm. (2017)

Coupled-dynamics of electrons and phonons

$$\frac{\partial f_{n\mathbf{k}}}{\partial t} = I_{n\mathbf{k}}^{\mathrm{e-ph}}[f,n]$$

$$\frac{\partial n_{\mathbf{q}\nu}}{\partial t} = I_{\mathbf{q}\nu}^{\mathrm{e-ph}}[f,n] + I_{\mathbf{q}\nu}^{\mathrm{ph-ph}}[n]$$

QUANTUMESPRESSO

Density functional theory

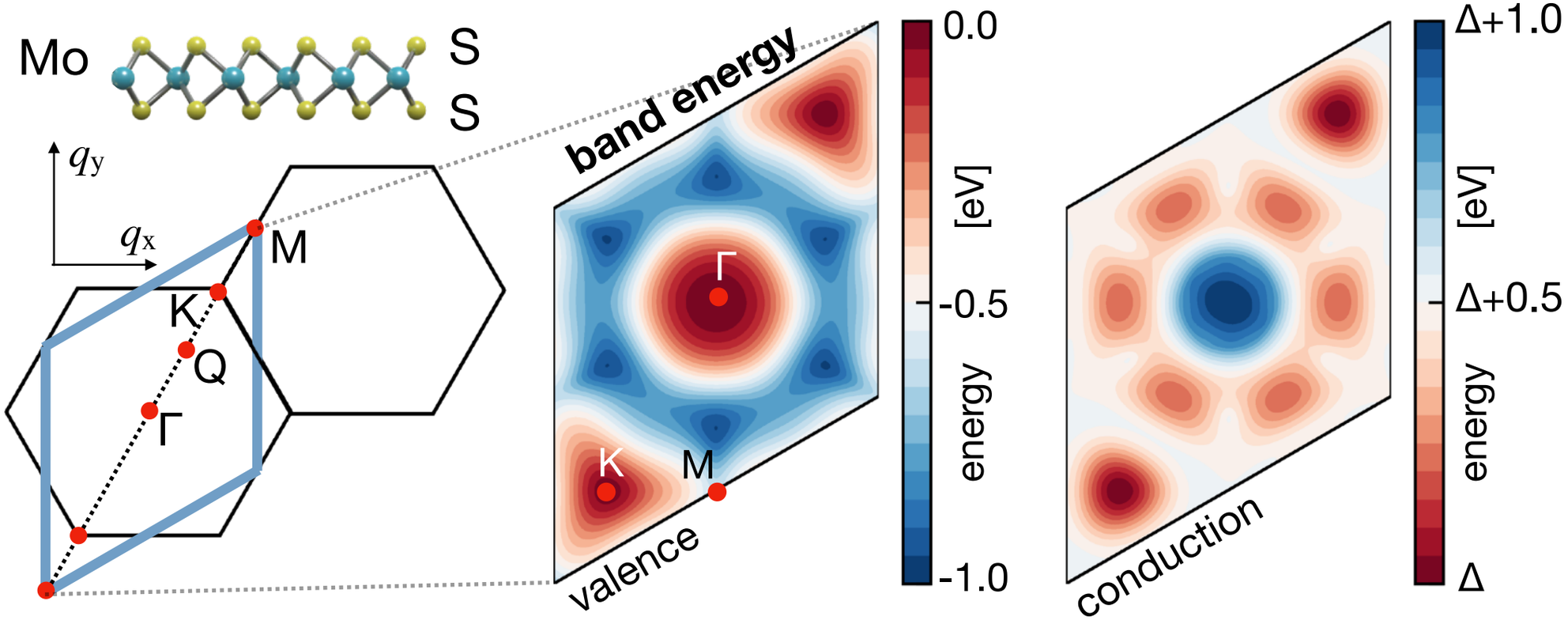
P. Giannozzi et al.,
J. Phys.: Condens. Matter 29, 465901 (2017)

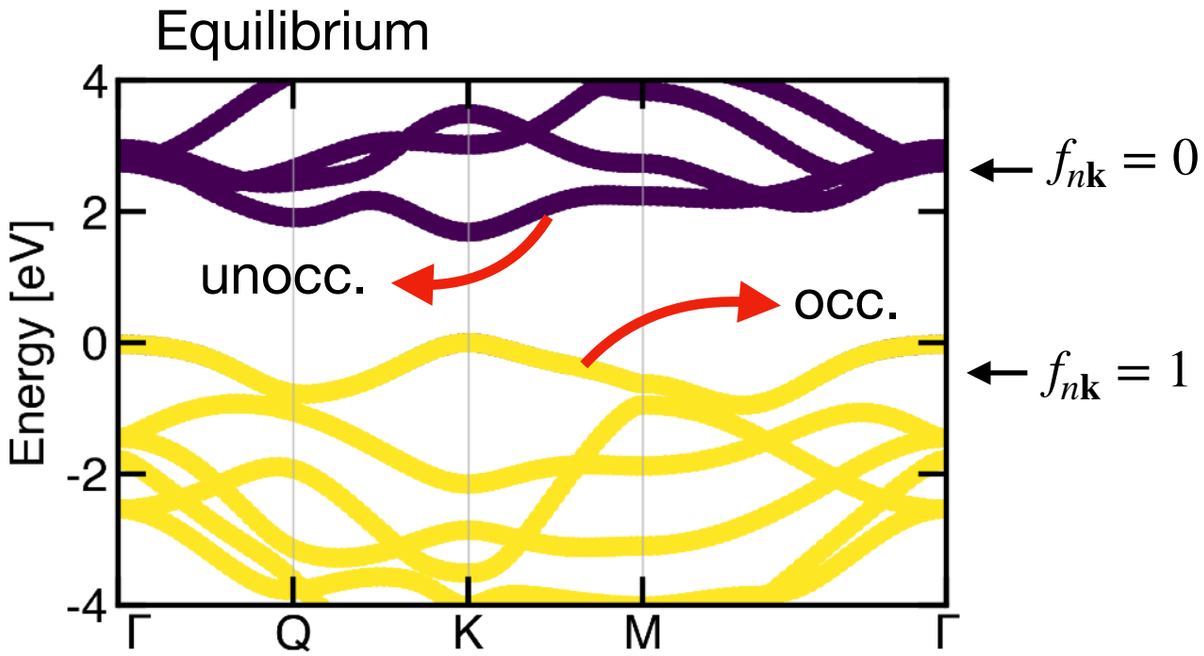
声 BTE

Third-order force constant

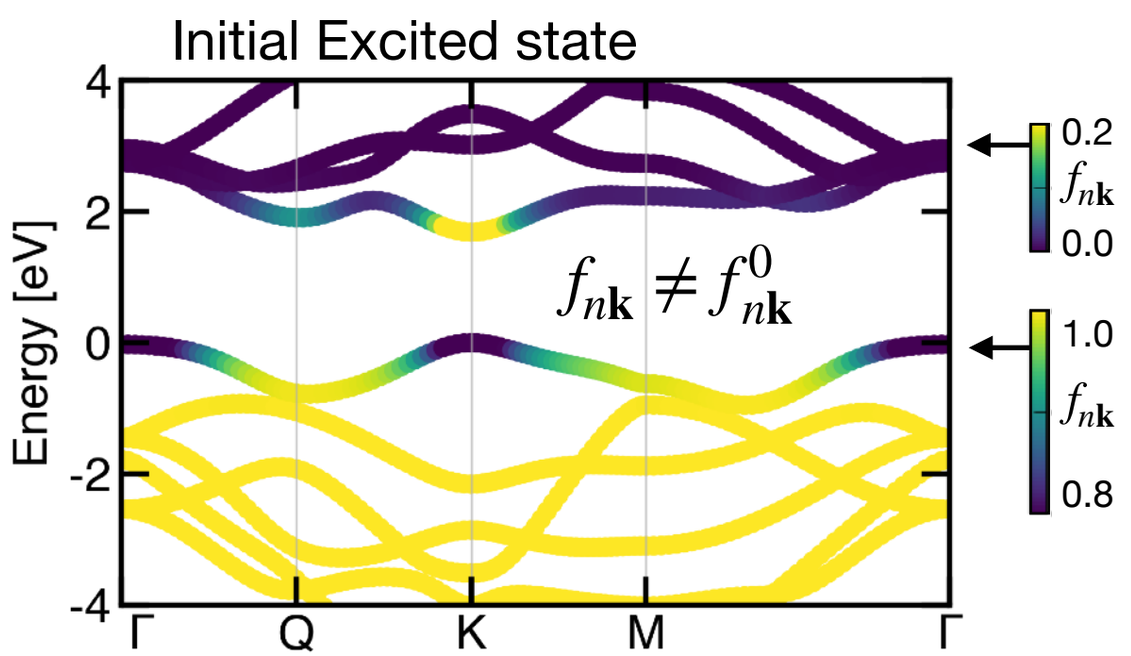
W. Li et al., Comp. Phys. Comm. 185, 1747 (2014)

Nonequilibrium electron dynamics in monolayer MoS_2





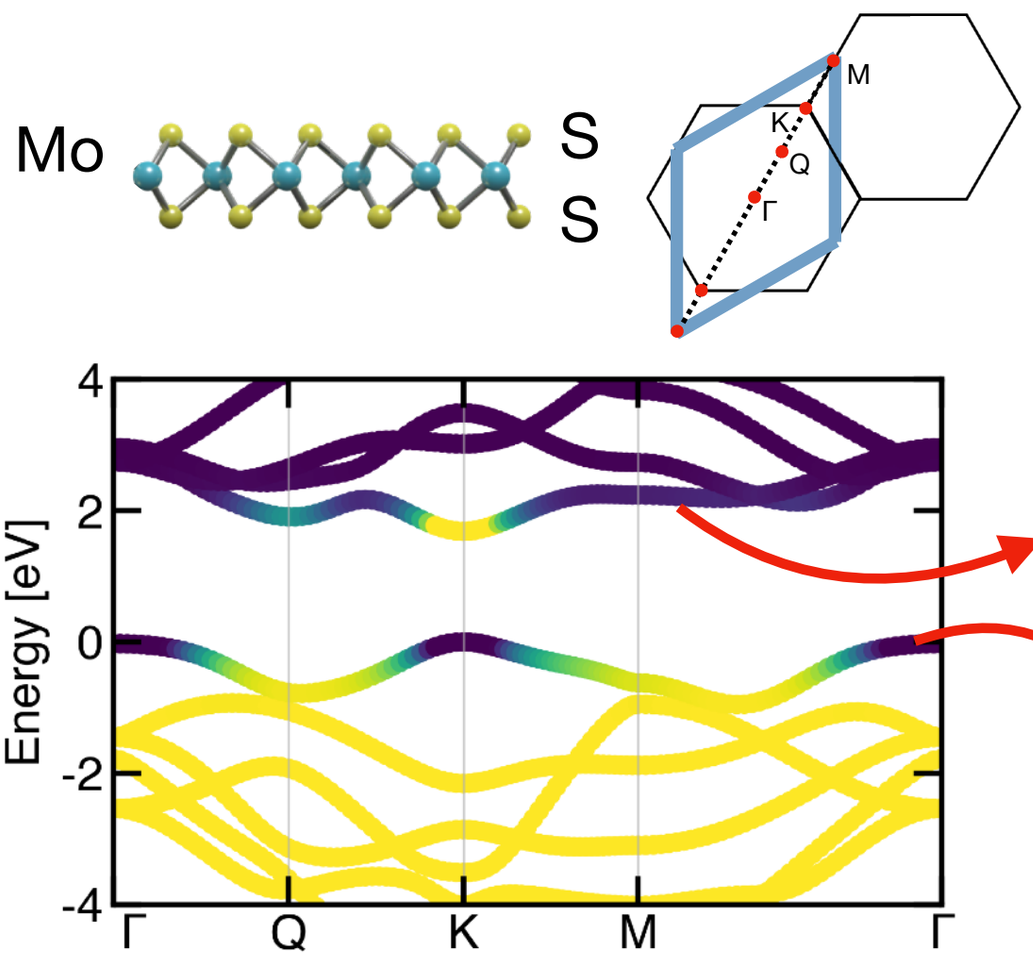
$$f^0_{n\mathbf{k}}(\mu, T) = [e^{(\varepsilon_{n\mathbf{k}} - \mu)/k_B T} + 1]^{-1}$$

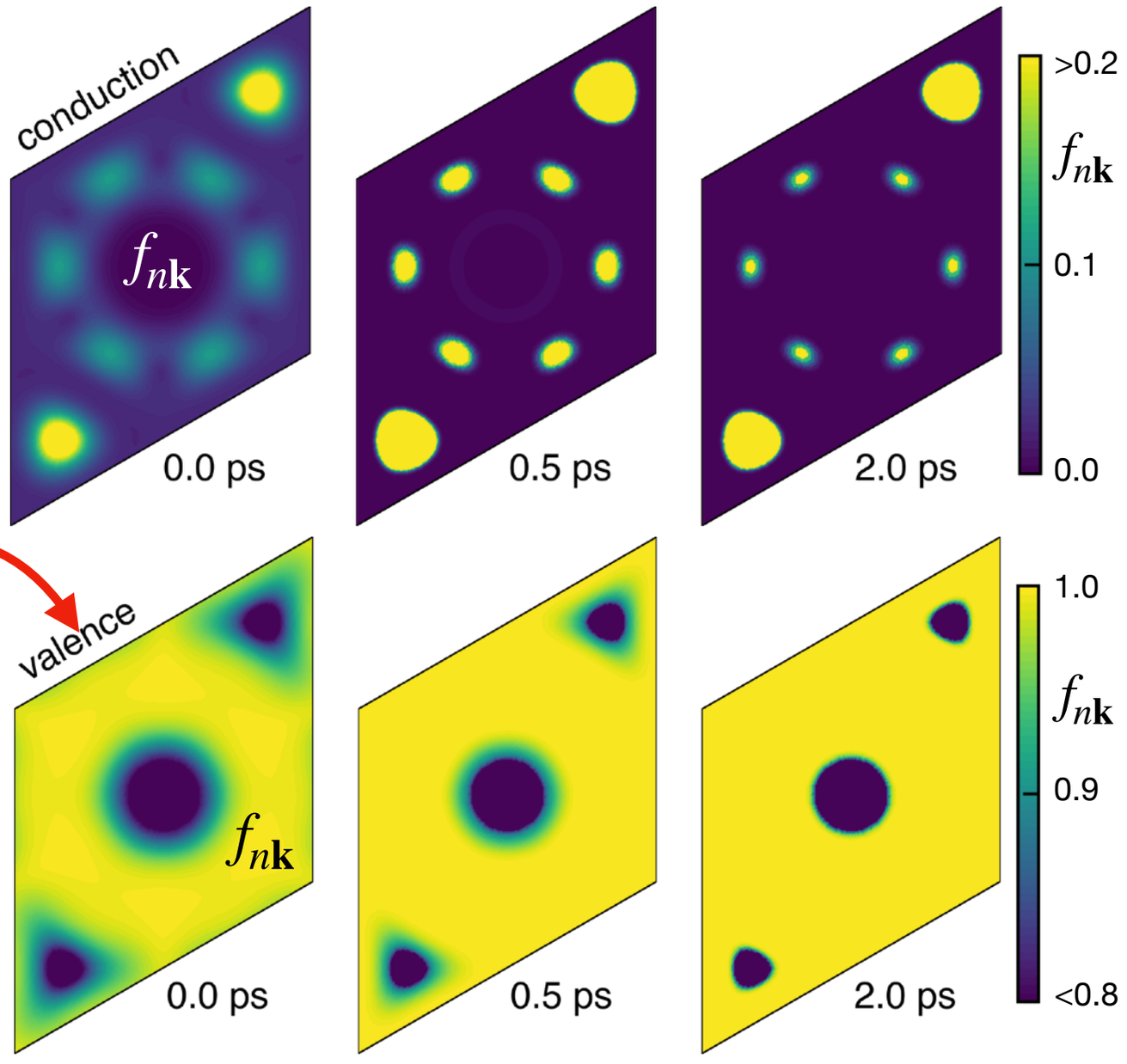


Excited carrier density: n=10^{14} cm^{-2}

FC, J. Phys. Chem. Lett. 12, 1274 (2021)

Nonequilibrium electron dynamics





Estimated timescales for electron relaxation: $< 2\,\mathrm{ps}$

Experiments from (*) (fs-electron diffraction): $1.7 \pm 0.3\,\mathrm{ps}$

(*) Mannebach et al., Nano Lett. 15, 6889 (2015)

$$\frac{\partial f_{n\mathbf{k}}}{\partial t} = I_{n\mathbf{k}}^{\mathrm{e-ph}}[f, n]$$

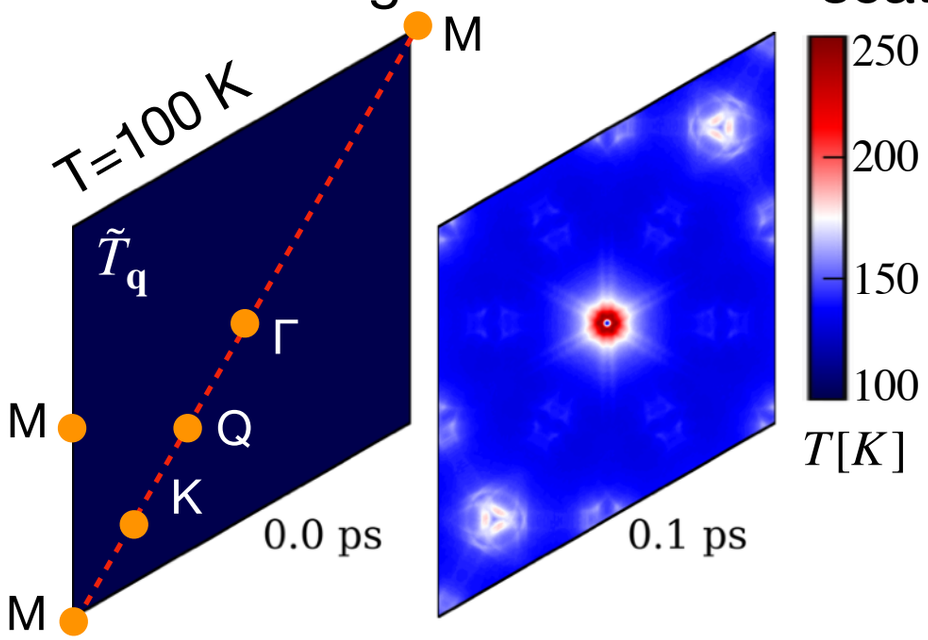
FC, J. Phys. Chem. Lett. 12, 1274 (2021)

Nonequilibrium phonon dynamics

$$\frac{\partial n_{\mathbf{q}\nu}}{\partial t} = I^{\mathrm{e-ph}}_{\mathbf{q}\nu}[f,n] + I^{\mathrm{ph-ph}}_{\mathbf{q}\nu}[n]$$

electron-phonon scattering

phonon-phonon scattering

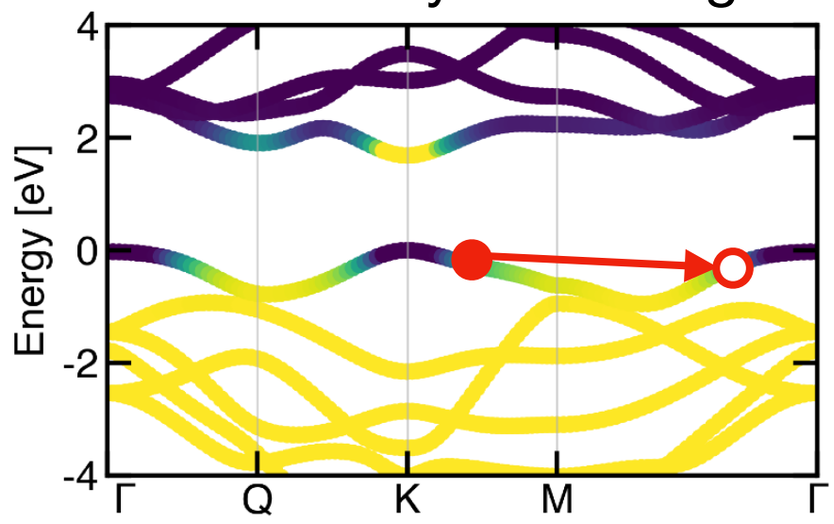


vibrational temperature

effective vibrational temperature

$$T_{\mathbf{q}\nu} = \hbar\omega_{\mathbf{q}\nu}[k_{\mathrm{B}}\ln(1+n_{\mathbf{q}\nu})]^{-1}$$

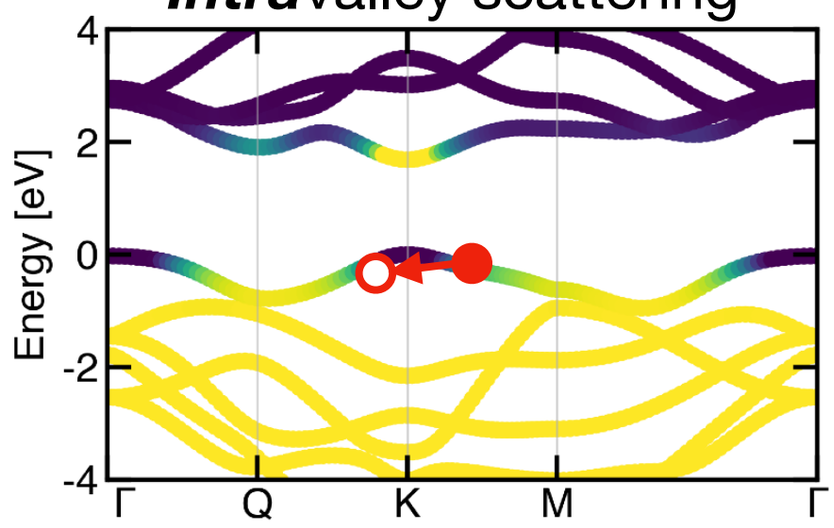
Intervalley scattering



allowed phonon momenta:

$\mathbf{q} \simeq \mathrm{K}$

Intravalley scattering



allowed phonon momenta:

$\mathbf{q} \simeq \Gamma$

Energy cons.: $\hbar\omega_{\mathbf{q}\nu} = \varepsilon_{n\mathbf{k}} - \varepsilon_{n\mathbf{k}'}$

Momentum cons.: $\mathbf{q} = \mathbf{k}' - \mathbf{k}$

FC, J. Phys. Chem. Lett. 12, 1274 (2021)

Nonequilibrium phonon dynamics

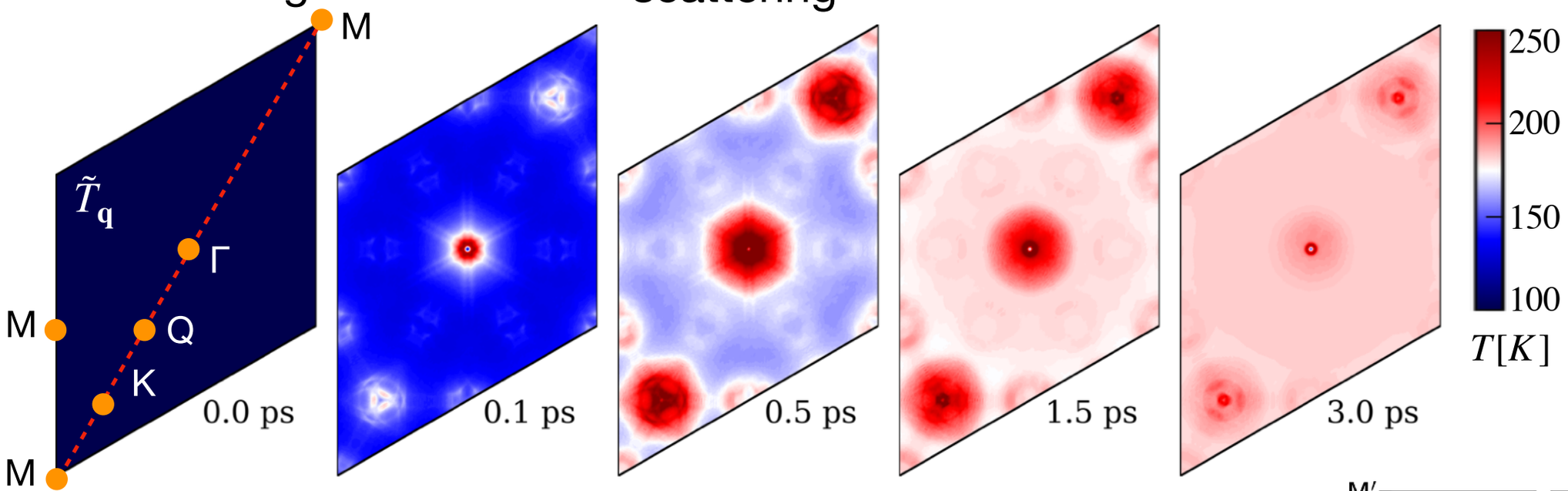
$$\frac{\partial n_{\mathbf{q}\nu}}{\partial t} = I^{\mathrm{e-ph}}_{\mathbf{q}\nu}[f,n] + I^{\mathrm{ph-ph}}_{\mathbf{q}\nu}[n]$$

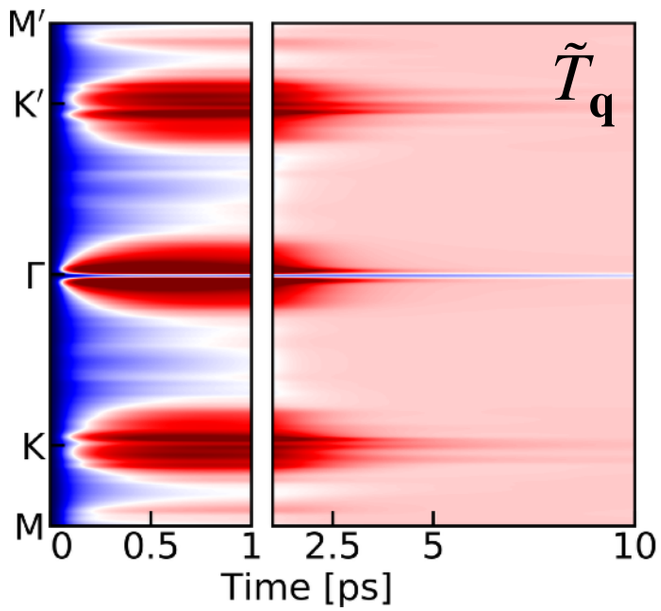
electron-phonon scattering

phonon-phonon scattering

effective vibrational temperature

$$T_{\mathbf{q}\nu} = \hbar\omega_{\mathbf{q}\nu}[k_{\mathrm{B}}\ln(1+n_{\mathbf{q}\nu})]^{-1}$$



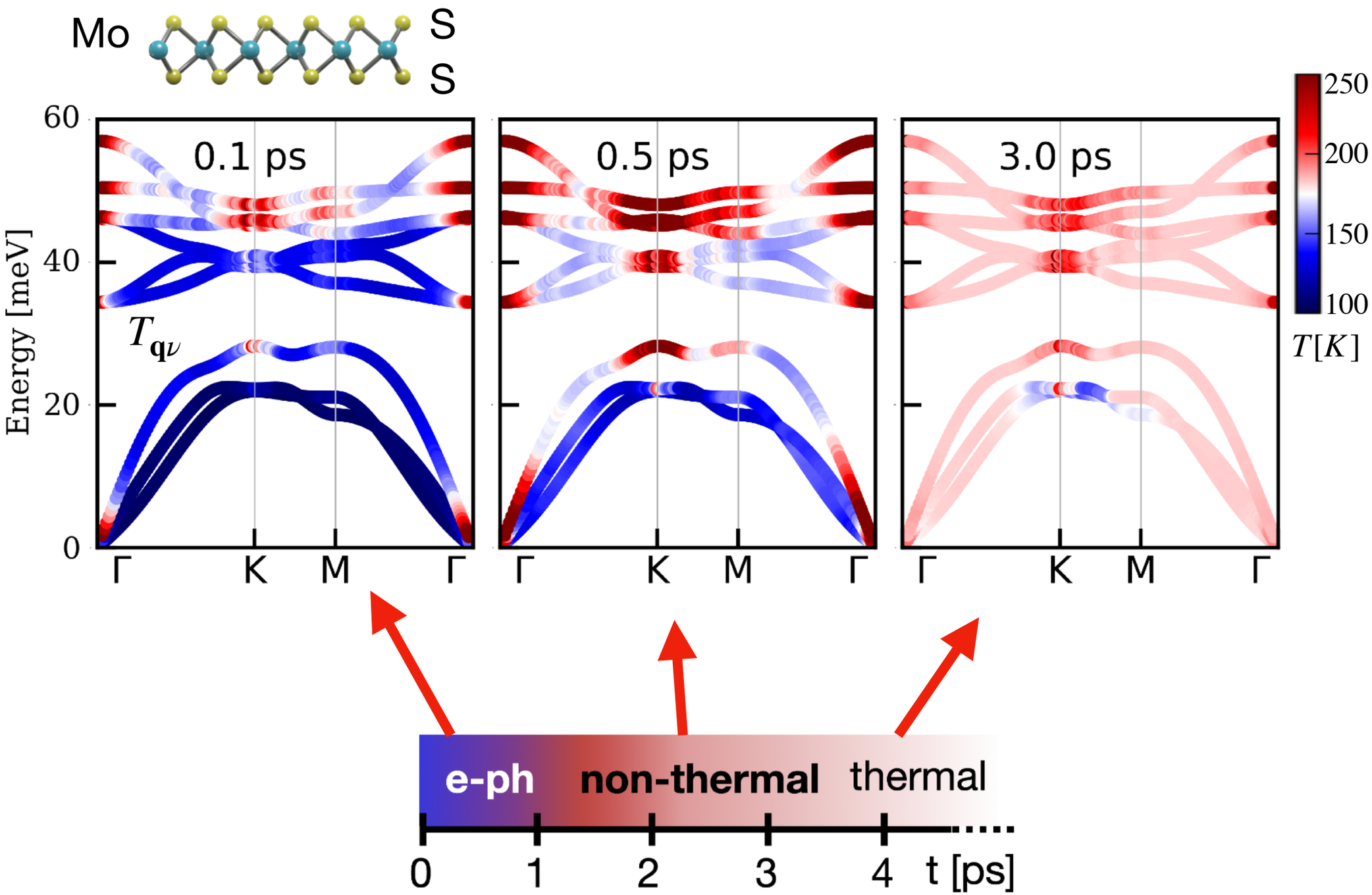


Energy cons.: $\hbar\omega_{\mathbf{q}\nu} = \varepsilon_{n\mathbf{k}} - \varepsilon_{n\mathbf{k}'}$

Momentum cons.: $\mathbf{q} = \mathbf{k}' - \mathbf{k}$

FC, J. Phys. Chem. Lett. 12, 1274 (2021)

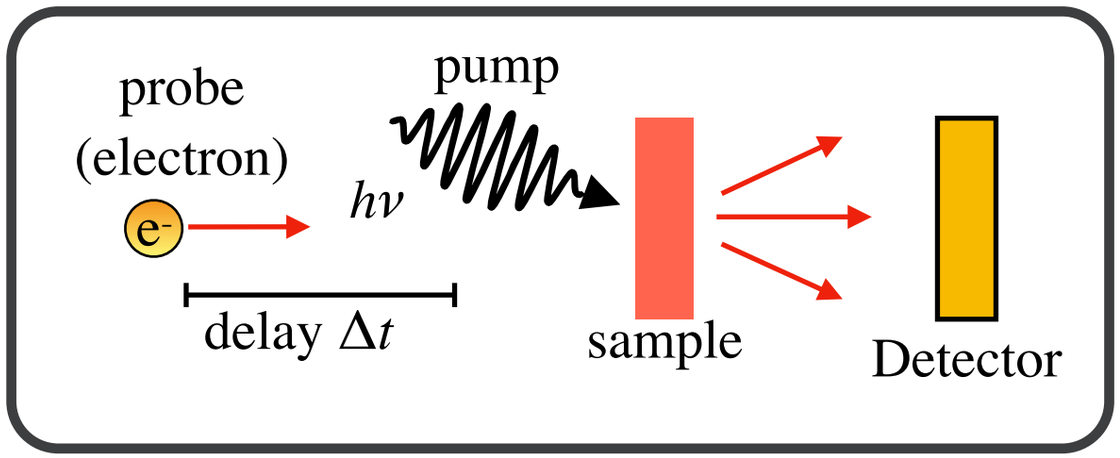
Regimes of nonequilibrium phonon dynamics



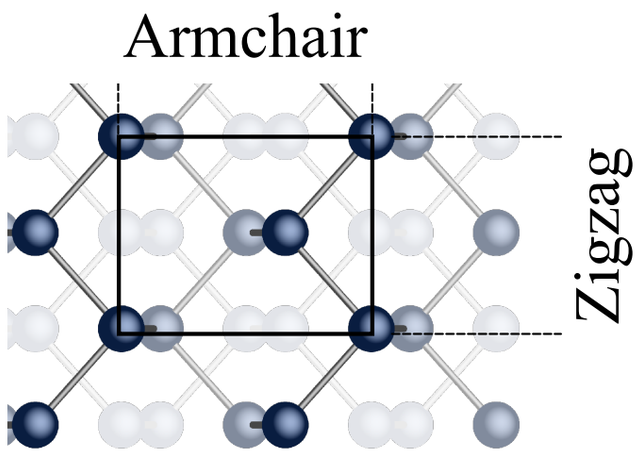
FC, J. Phys. Chem. Lett. 12, 1274 (2021)

Electron diffuse scattering: black Phosphorus

Femto-second electron diffuse scattering (FEDS)



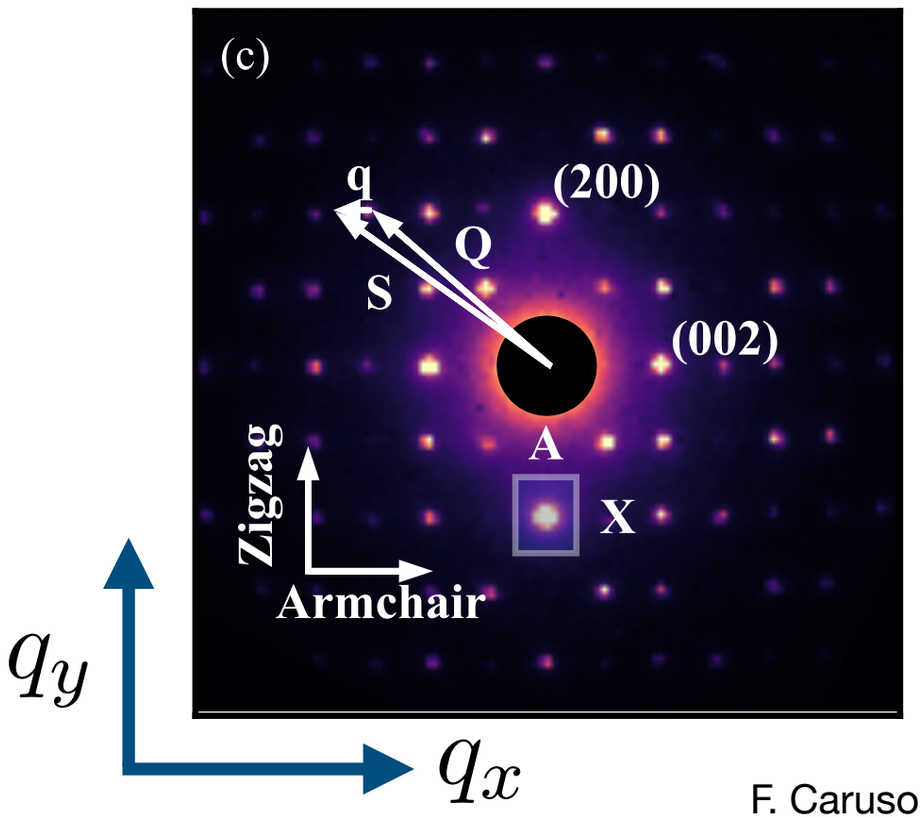
Black phosphorus



Experiments: FHI Berlin

Helene Seiler

Ralph Ernstorfer

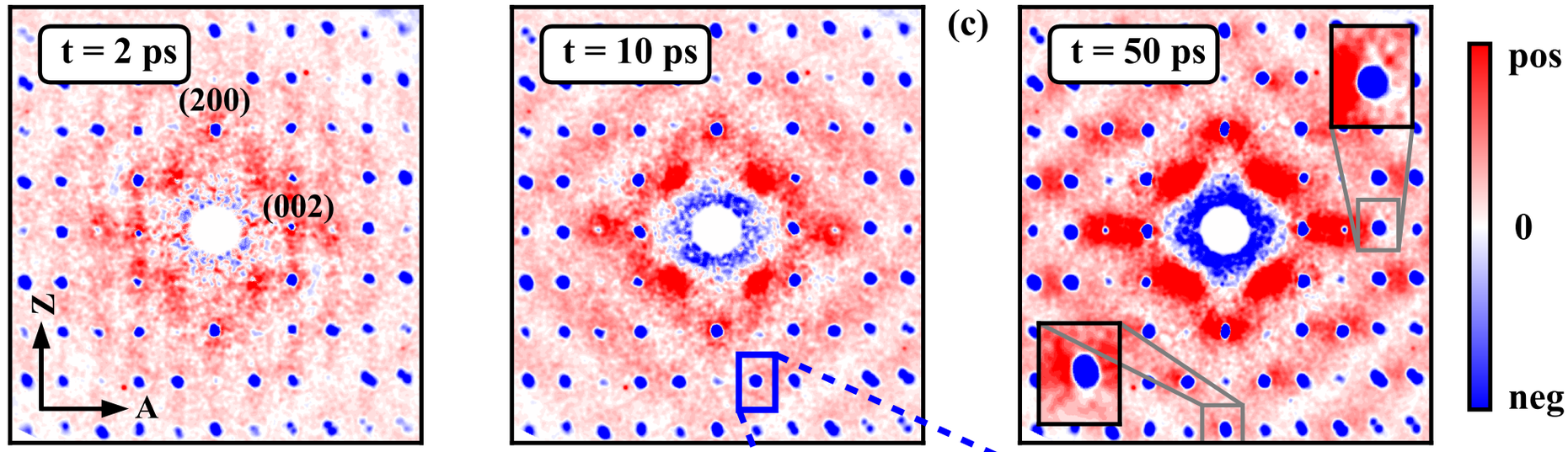


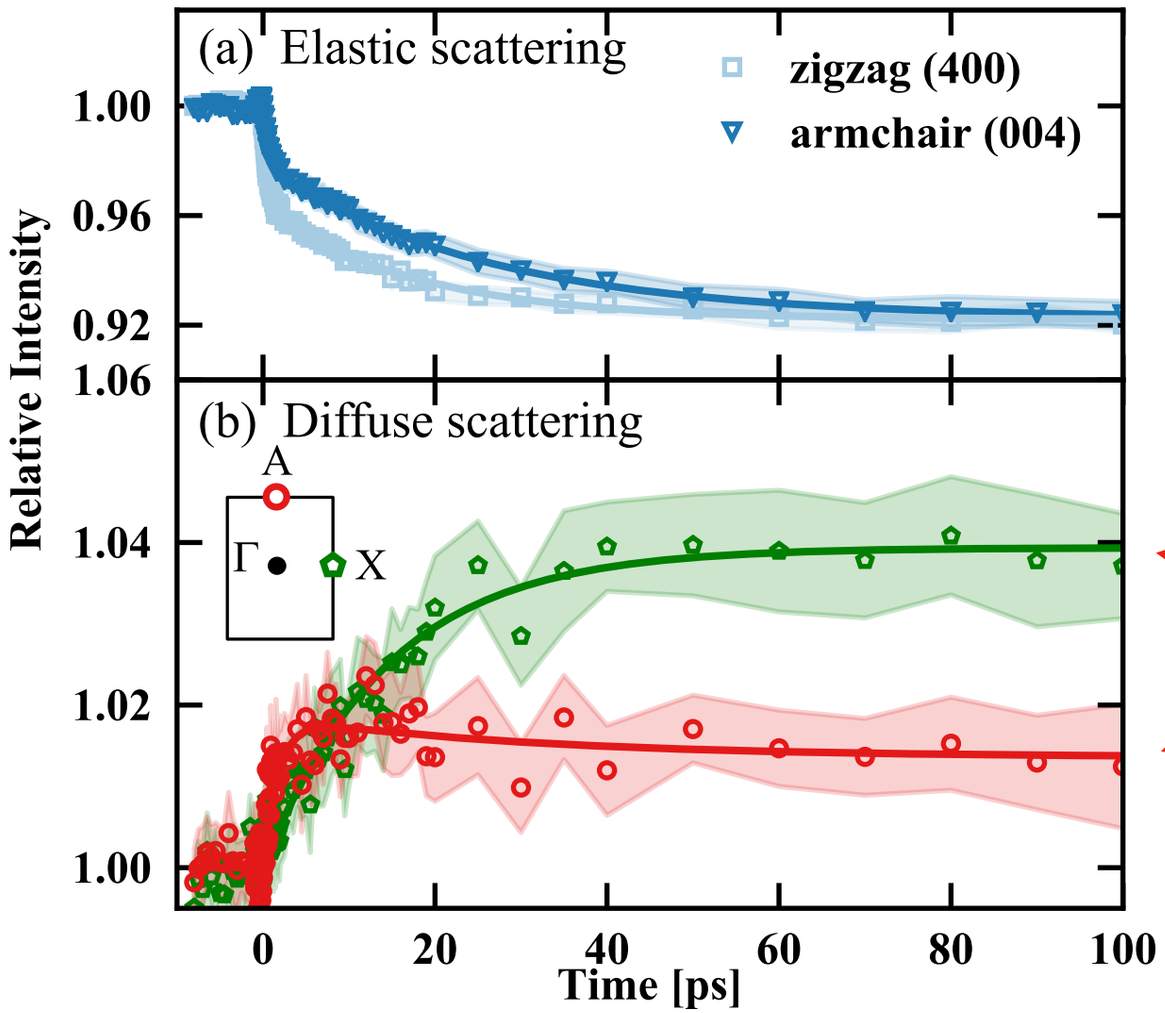
Anisotropic non-equilibrium lattice dynamics in bP

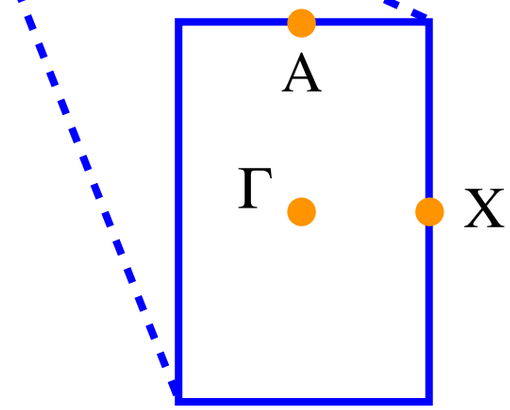
Experiment:

Intensity difference:

$$I(\mathbf{S},t) - I(\mathbf{S},t<t_0)$$

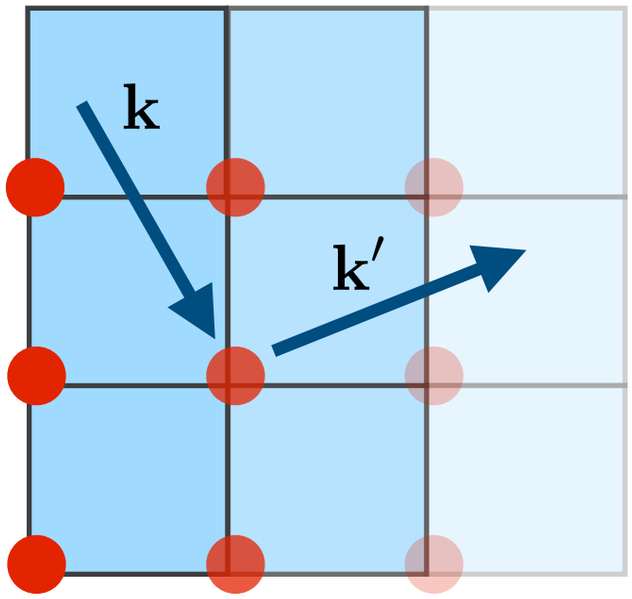






Highly-anisotropic lattice dynamics

Diffraction from a lattice



Bragg's law for interference condition:

$$A(\mathbf{q}) = \sum_{p}^{N_p} f_0 \, \exp\left[i\mathbf{q}\cdot\mathbf{R}_p\right]$$

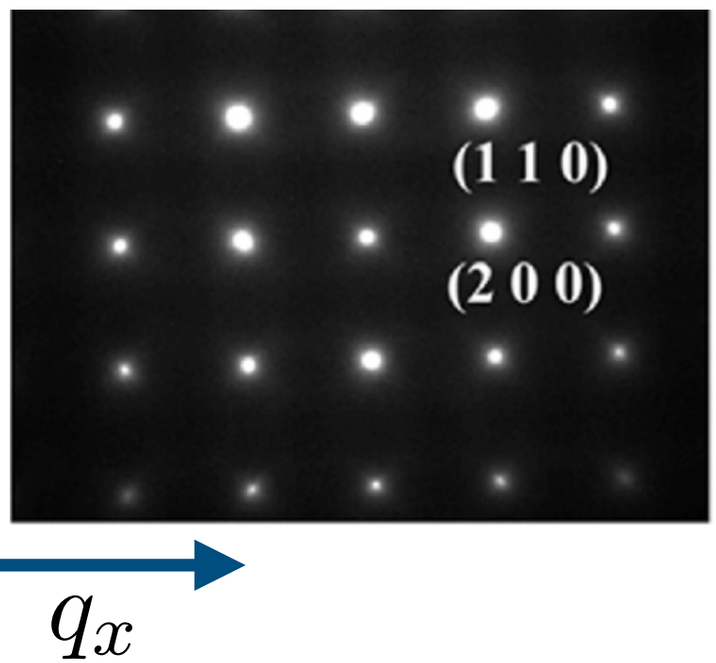
amplitude

Scattering cross section

$\mathbf{q} = \mathbf{k}' - \mathbf{k}$ Transferred momentum

Lattice vector

example: Cubic lattice

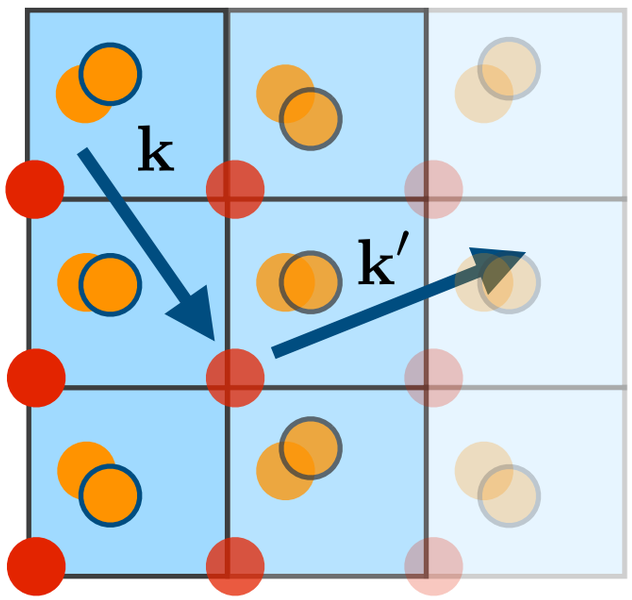


Scattering intensity for a perfect lattice:

$$I(\mathbf{q}) = |A(\mathbf{q})|^2 \simeq \delta_{\mathbf{q},\mathbf{G}}$$

Reciprocal lattice vector

Diffraction from a vibrating lattice



Zero-phonon term:

$$\langle I_0(\mathbf{S})\rangle_T \quad = \quad N_p^2 |f_0|^2 \exp(-2W_T)\delta_{\mathbf{S},\mathbf{G}}$$

Debye-Waller factor:

$$2W_T = \frac{1}{M_\kappa N_p}\sum_{\mathbf{q}\nu}\left[\sum_{\alpha\alpha'} S_\alpha S_{\alpha'} e^{\nu}_{\kappa\alpha}(\mathbf{q}) e^{\nu *}_{\kappa\alpha'}(\mathbf{q})\right] E_{\mathbf{q}\nu,T}$$

One-phonon (dynamical) structure factor

$$\begin{aligned}\langle I_1(\mathbf{S},E)\rangle_T \quad &= \quad |f_0|^2 \exp(-2W_T)\frac{\hbar^2 N_p}{2M_\kappa}\sum_{\mathbf{q}\nu}\left[\sum_{\alpha\alpha'} S_\alpha S_{\alpha'} e^{\nu}_{\kappa\alpha}(\mathbf{q}) e^{\nu *}_{\kappa\alpha'}(\mathbf{q})\right]\frac{1}{\hbar\omega_{\mathbf{q}\nu}} \quad (16)\\ &\times \quad \left[\delta(\mathbf{S}+\mathbf{q}) n_{\mathbf{q}\nu,T}\delta(E+\hbar\omega_{\mathbf{q}\nu}) + \delta(\mathbf{S}-\mathbf{q})(n_{\mathbf{q}\nu,T}+1)\delta(E-\hbar\omega_{\mathbf{q}\nu})\right].\end{aligned}$$

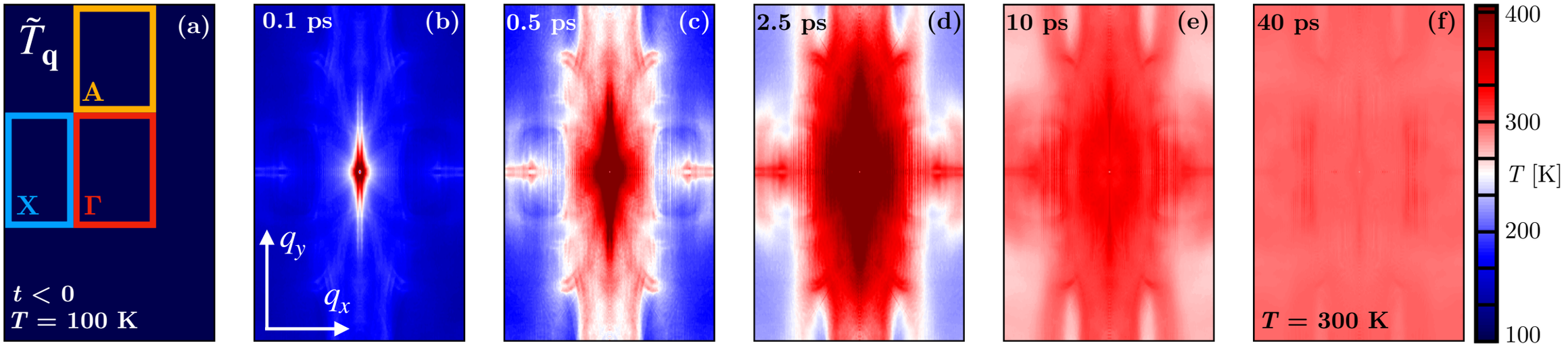
from the time-dependent Boltzmann equation

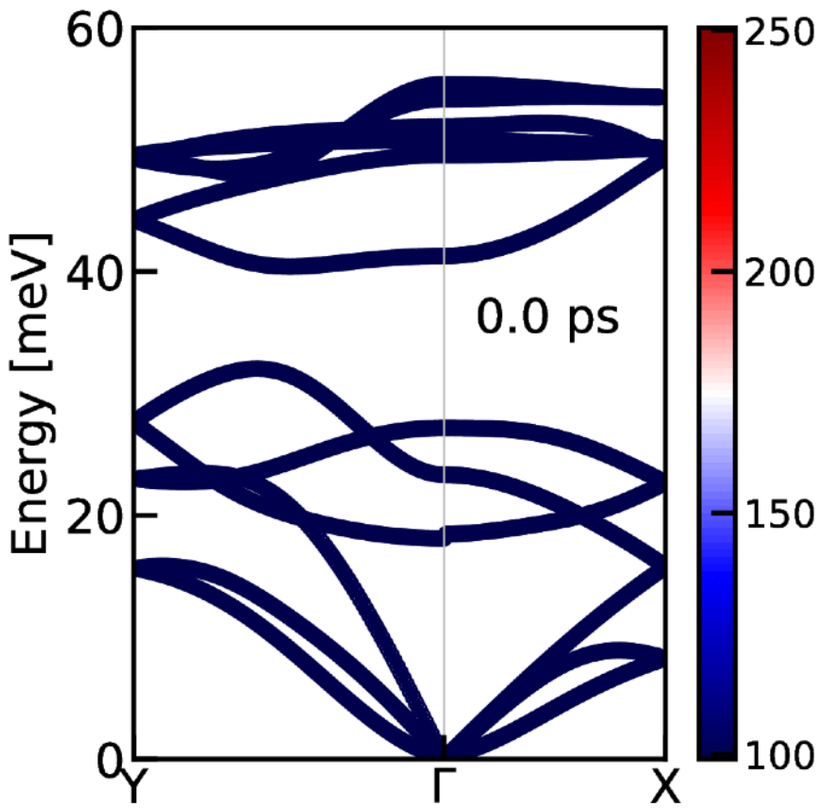
First-principles approach to Femto-second electron diffuse scattering (FEDS)

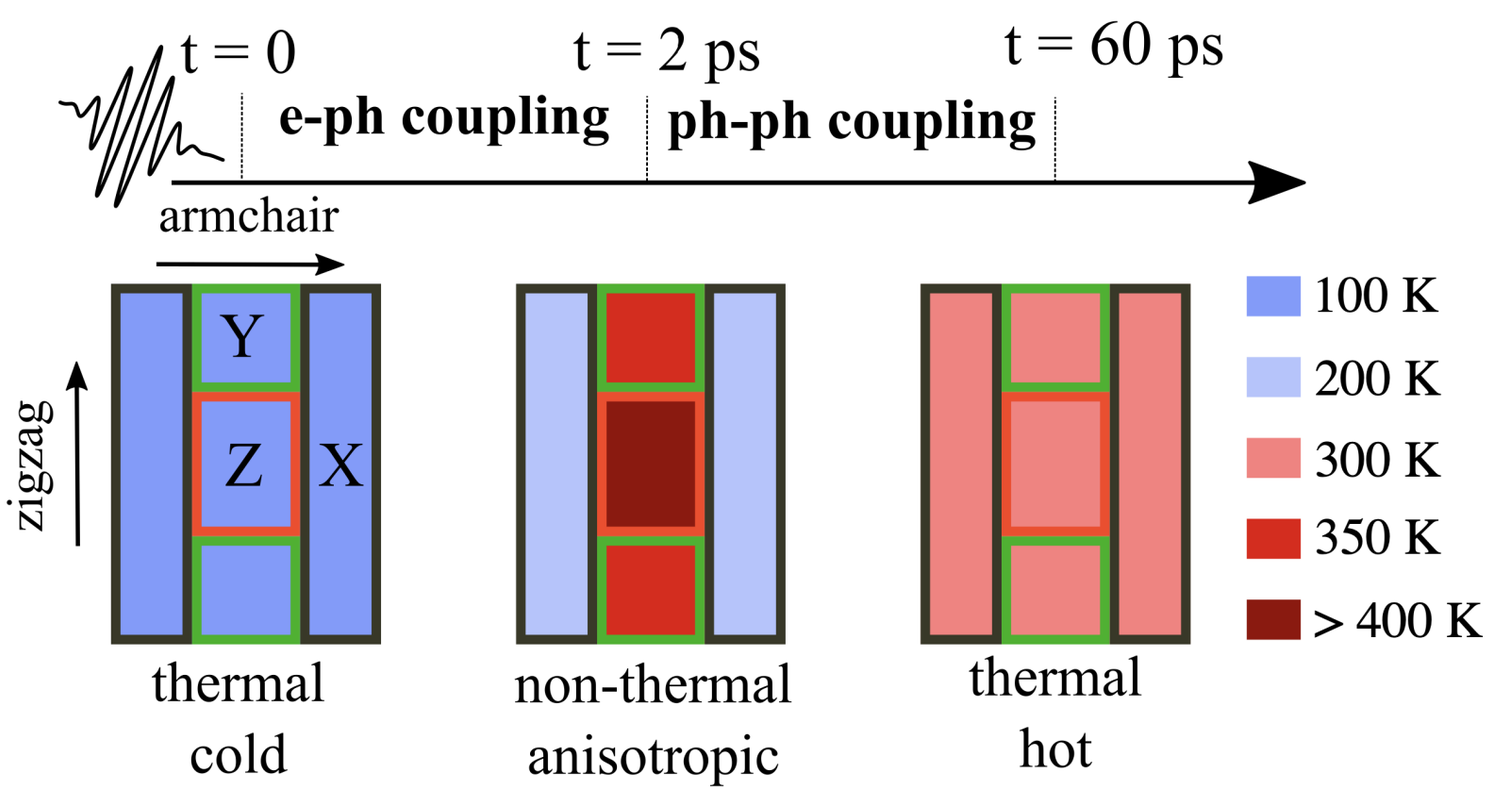
Non-equilibrium lattice dynamics in bP from first-principles

$$\frac{\partial n_{\mathbf{q}\nu}}{\partial t} = I^{\mathrm{e-ph}}_{\mathbf{q}\nu}[f,n] + I^{\mathrm{ph-ph}}_{\mathbf{q}\nu}[n]$$

Effective vibrational temperature

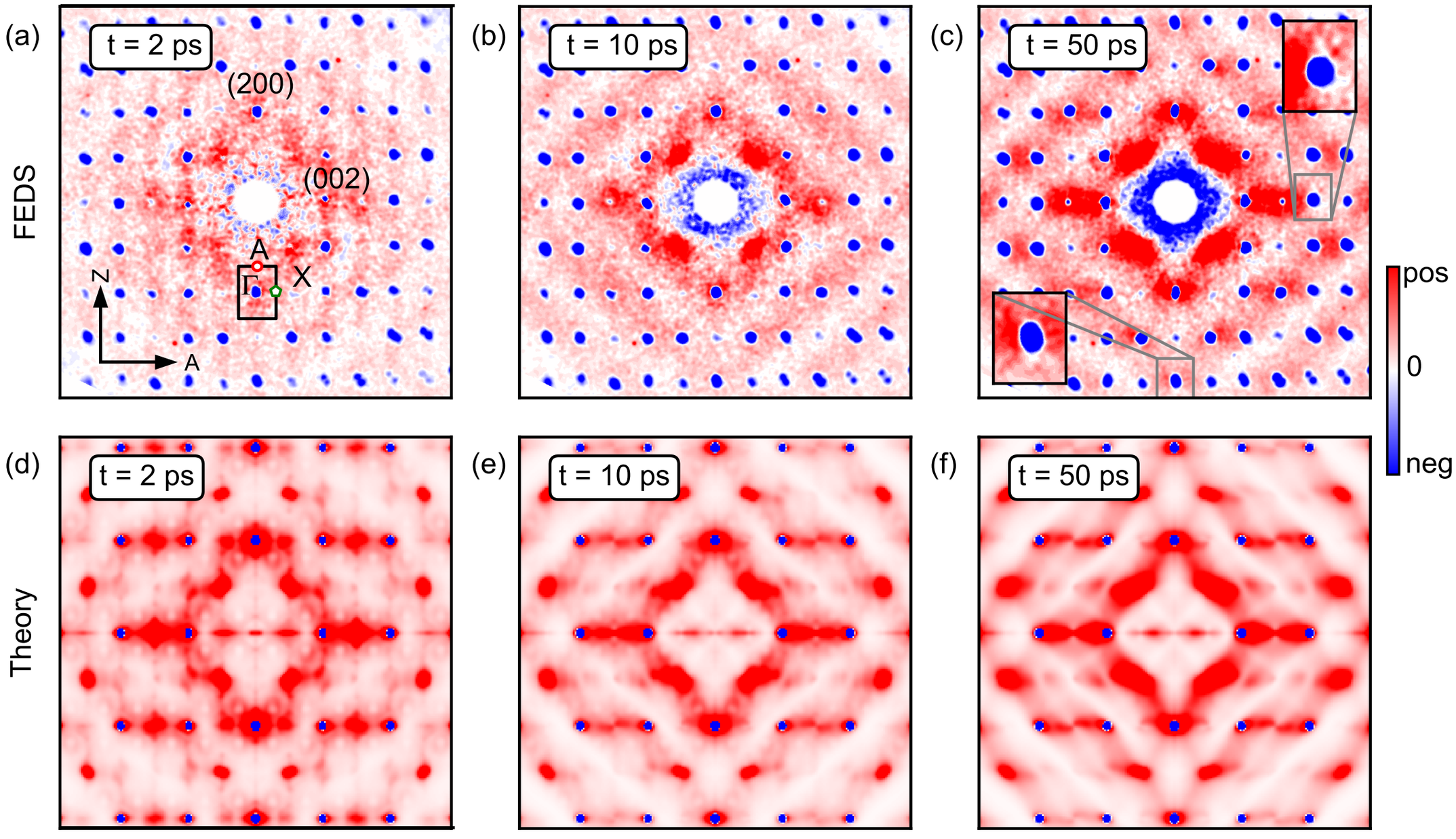




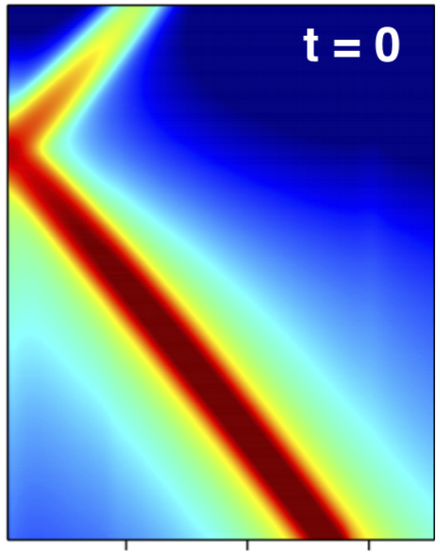


Non-equilibrium lattice dynamics in bP from first-principles

Intensity difference: $I(\mathbf{S},t) - I(\mathbf{S},t<t_0)$

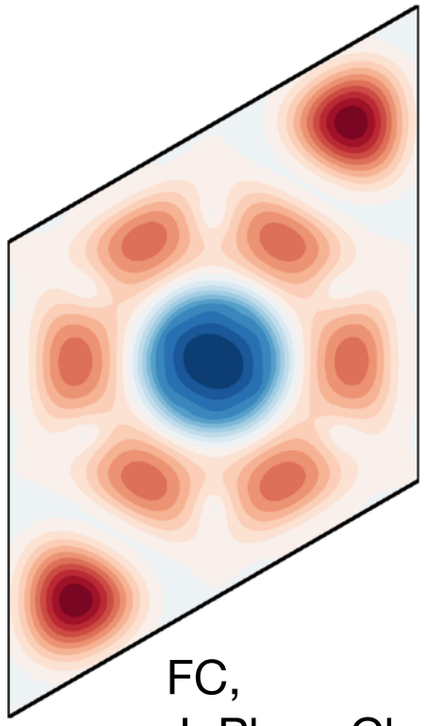


Ultrafast dynamics of electrons and phonons



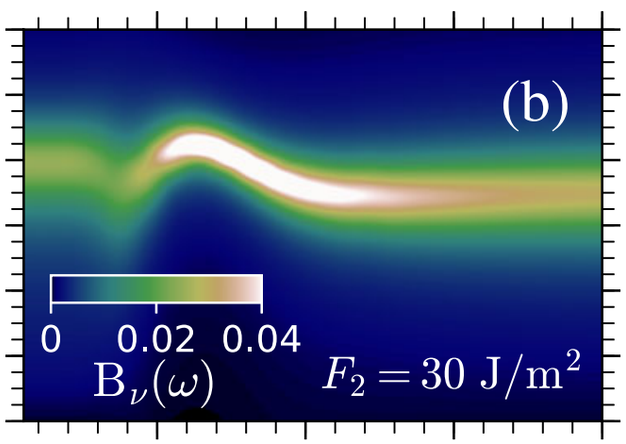
Nonequilibrium carrier dynamics in graphene

FC, Novko, Draxl,
Phys. Rev. B **101** 035128 (2020)



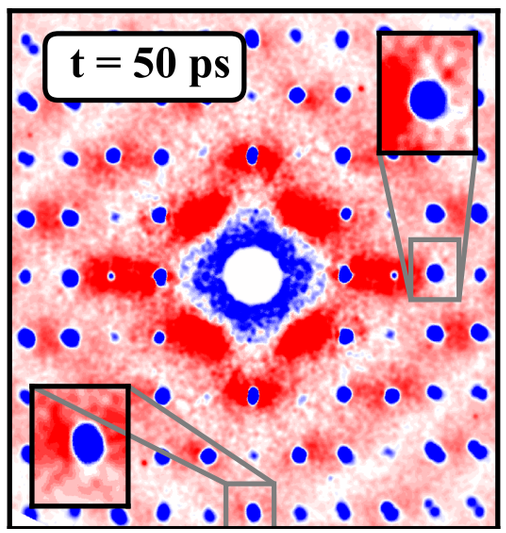
Nonequilibrium lattice dynamics in MoS_2

FC,
J. Phys. Chem. Lett. 12, 1274 (2021)



Transient phonon renormalization in MgB_2

Novko, FC, Draxl, Cappelluti, Phys.
Rev. Lett. **124** 077001 (2020)



Signatures of nonequilibrium lattice dynamics in phosphorene

Seiler, Zahn, Zacharias, et al.
arXiv (2020)

Acknowledgements

THEORY

Humboldt-Universität zu Berlin
- Claudia Draxl
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- Maximilian Schebek
- Sebastian Tillak

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- Dino Novko

University of Kiel
- Christoph Emeis

EXPERIMENTS

Fritz Haber Institute, Berlin
- Helene Seiler
- Daniela Zahn
- Ralph Ernstorfer

EPW:
S. Poncé et al.,
Comp. Phys. Comm. (2017)

Quantum Espresso:
P. Giannozzi et al., J. Phys.:
Condens. Matter 29, 465901 (2017)

Wannier90: G. Pizzi et al., J. Phys.
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