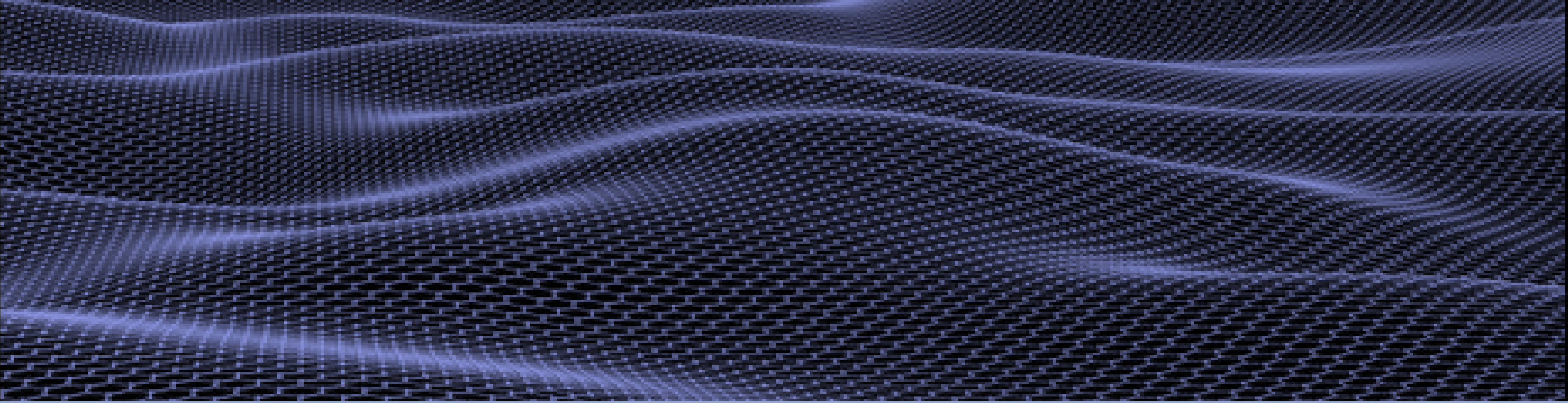

Microscopic modelling of graphene

Microscopic view on optical and electronic properties of graphene

Ermin Malić

Assistent Professor
Department of Applied Physics

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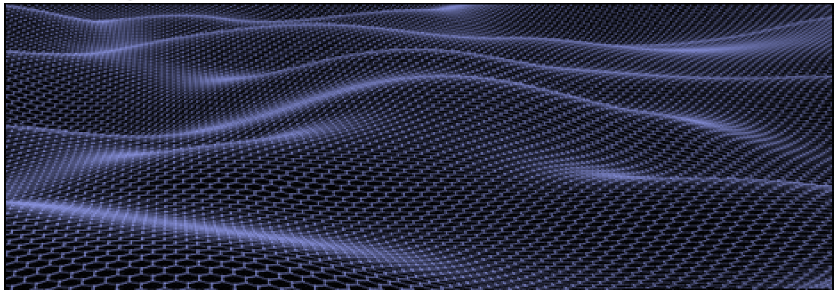
Guest lecture, FKA091 Condensed Matter Physics, December 3-4, 2015

Think-pair-share: What is graphene?

- What do you associate with the material graphene?
- What do you think makes graphene different from conventional materials, such as silicon?

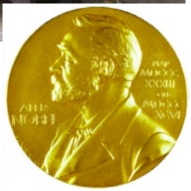
Brief history of graphene

Graphene – the perfect atomic lattice



Discovered 2004 (University of Manchester)

Nobel Prize for Physics 2010



GRAPHENE FLAGSHIP

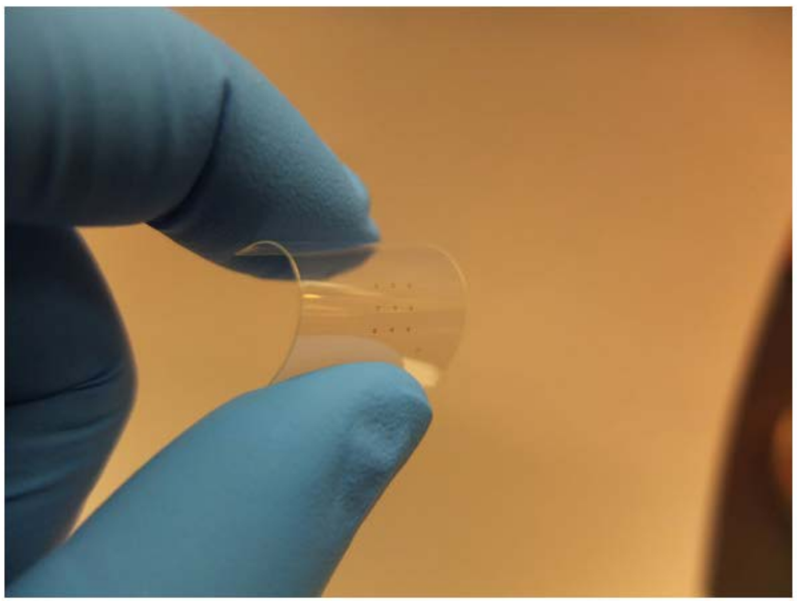
2013 EU graphene flagship launched
budget 1 billion €
(Chalmers leading university)

Graphene – the Material of Tomorrow?

The New York Times

Bend It, Charge It, Dunk It: Graphene, the Material of Tomorrow

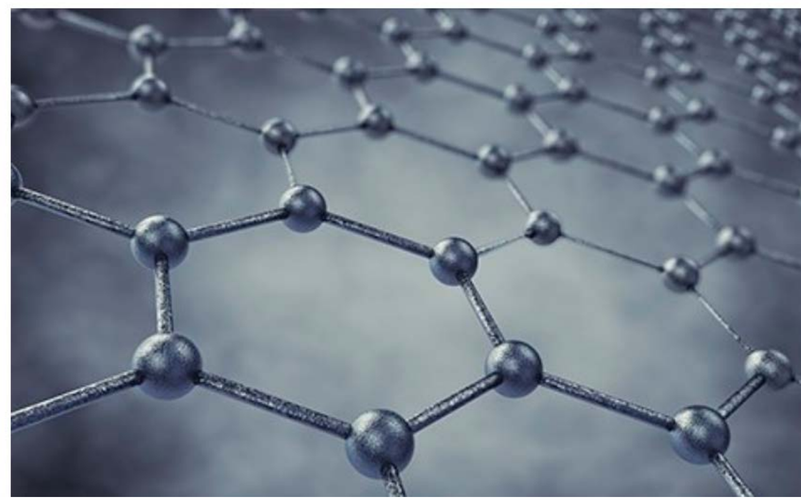
By NICK BILTON APRIL 13, 2014 11:00 AM 166 Comments



theguardian
Winner of the Pulitzer prize 2014

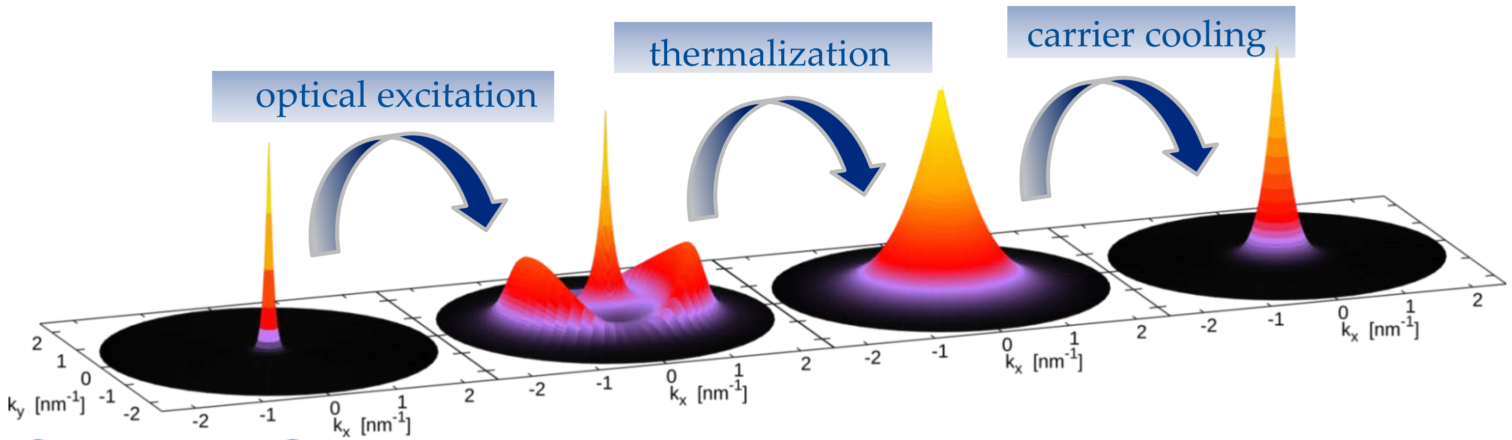
Graphene - the new wonder material

Scientific interest rolls in for a material that is more solid than steel and a better conductor than copper



Outline

- Motivation
- Microscopic modelling
- Carrier dynamics
- Many-particle phenomena



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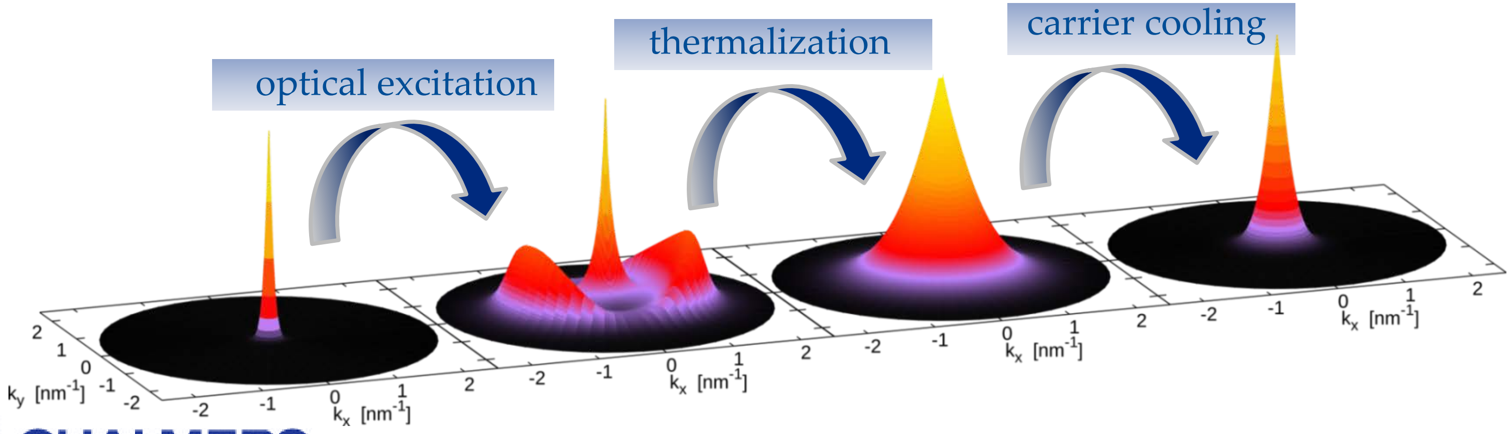
Learning Outcomes

- Recognize the **potential of graphene** for fundamental science and technological applications
- Understand how optical and electronic properties of graphene can be **microscopically modelled** (tight-binding, second quantization, Bloch equations)
- Explain how ultrafast **carrier dynamics** in graphene works
- Realize the importance of **carrier multiplication** and its relevance for highly efficient photodetectors
- Demonstrate the importance of **population inversion** for highly tunable graphene-based lasers

Outline

- Motivation
- Microscopic modelling

- Carrier dynamics
- Many-particle phenomena



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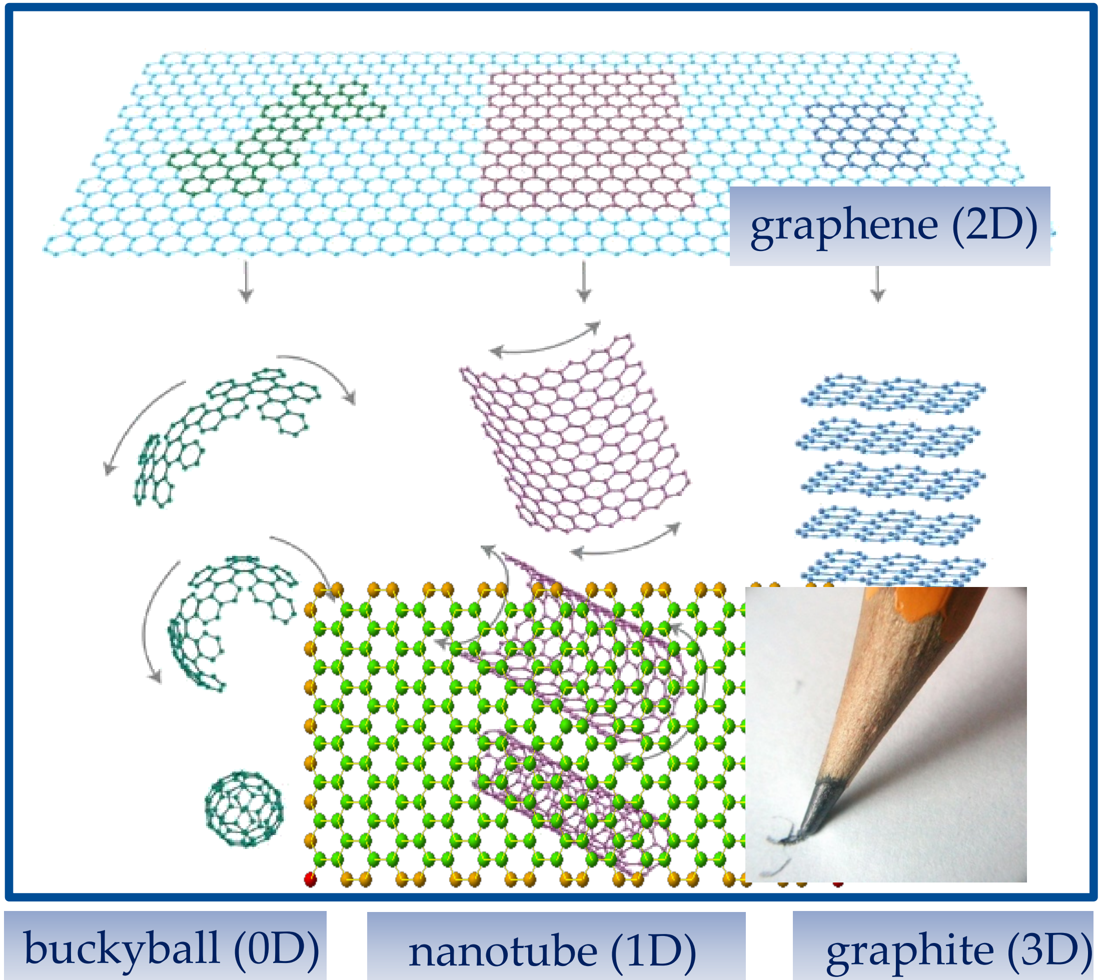
What is graphene?



Maruyama group, University of Tokyo

A. K. Geim and K. S. Novoselov, Nature Materials 6, 183 (2007)

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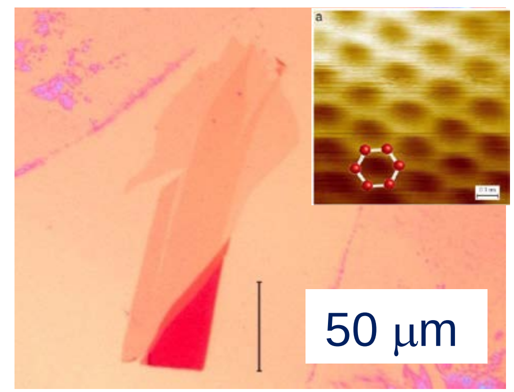
Discovery of graphene

- Discovered **2004** via mechanical exfoliation (**scotch-tape/drawing method**)

Optical microscope



- Using a **piece of graphite**, an **adhesive tape**, a substrate, and an optical microscope, graphene can be produced in **high quality**

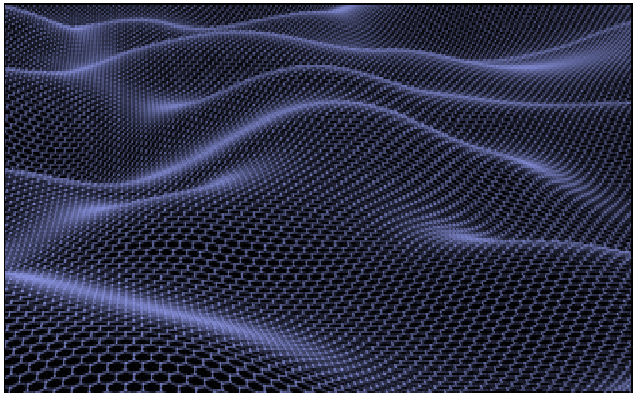
Novoselov, Rev. Mod. Phys. 83, 837 (2011)

Nobel Award in physics 2010

- **Andre Geim** and **Konstantin Novoselov** (University of Manchester) receive the Nobel Prize for „groundbreaking experiments on graphene“

→ *“New **material** with **unique properties**”*

→ *“Manifold of **practical application** areas”*



Introduction to graphene

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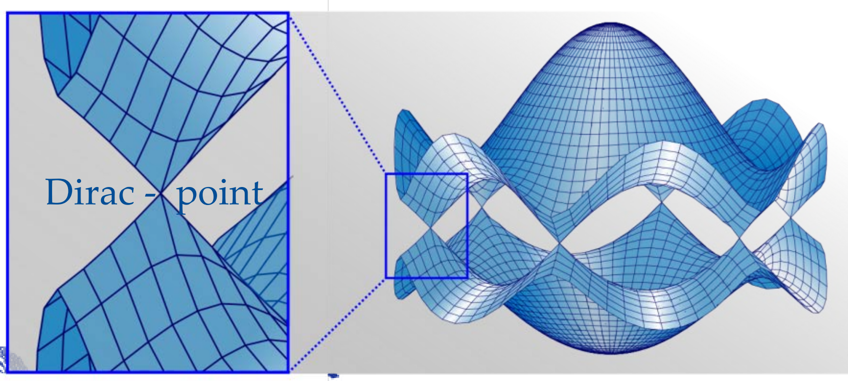
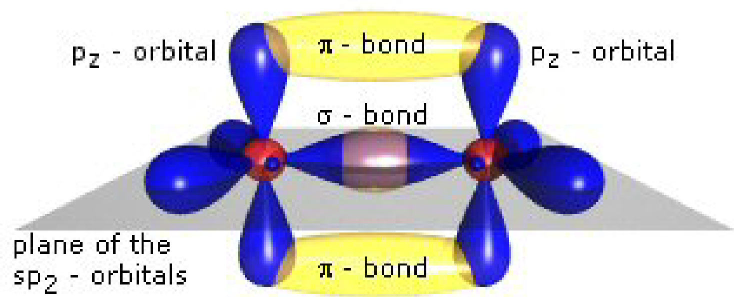
Properties of graphene

- **Andre Geim** and **Konstantin Novoselov** (University of Manchester) receive the Nobel Prize for „groundbreaking experiments on graphene“

→ *“**New material** with **unique properties**”*

  - Extraordinary conductor of current and heat (ballistic transport)
  - Very **strong** and **light** at the same time (sp^2 bonds)
  - Almost **transparent** (absorbs only 2.3 % of visible light)
  - **Linear bandstructure** close to the Dirac point



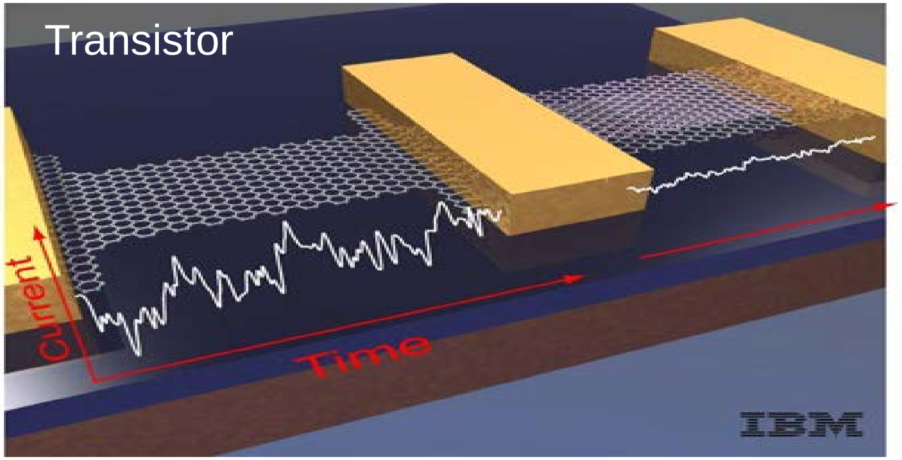


Application potential of graphene

- **Andre Geim** and **Konstantin Novoselov** (University of Manchester) receive the Nobel Prize for „groundbreaking experiments on graphene“

→ *“Manifold of **practical application** areas”*

  - Graphene-based **transistors** are much faster than silicon transistors (first IBM prototype shows a frequency of **100 GHz**)



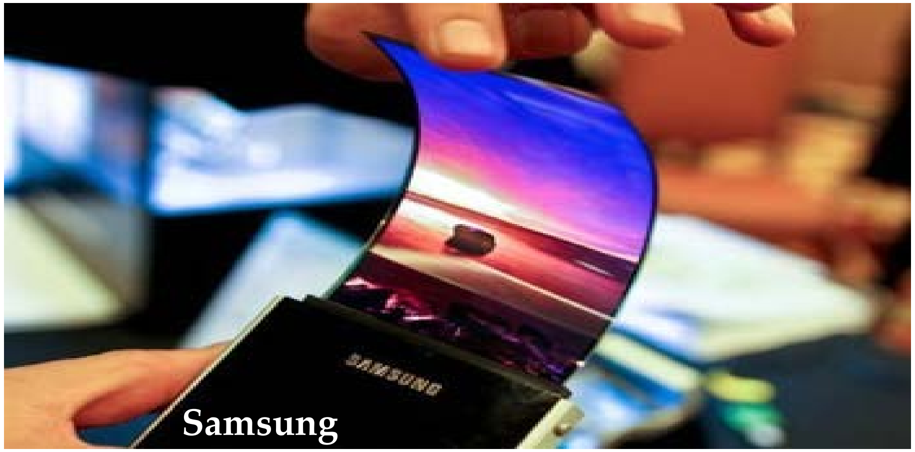
Application potential of graphene

- **Andre Geim** and **Konstantin Novoselov** (University of Manchester) receive the Nobel Prize for „groundbreaking experiments on graphene“

→ *“Manifold of **practical application** areas”*

  - Graphene-based **transistors** are much faster than silicon transistors (first IBM prototype shows a frequency of **100 GHz**)

  - Transparent and flexible **touch screens** and **solar cells**



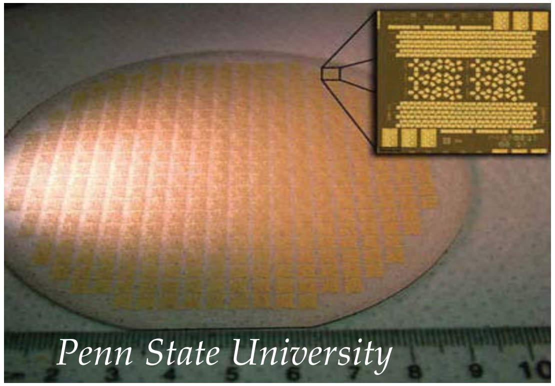
Application potential of graphene

Current challenges

- **Large-area production** of high-quality graphene

  → progress in growth techniques

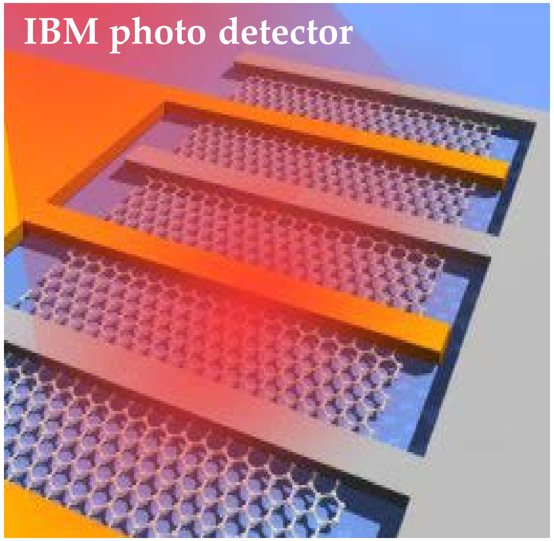
- **Lack of band gap** gives rise to **insufficient on-off ratios** in transistors



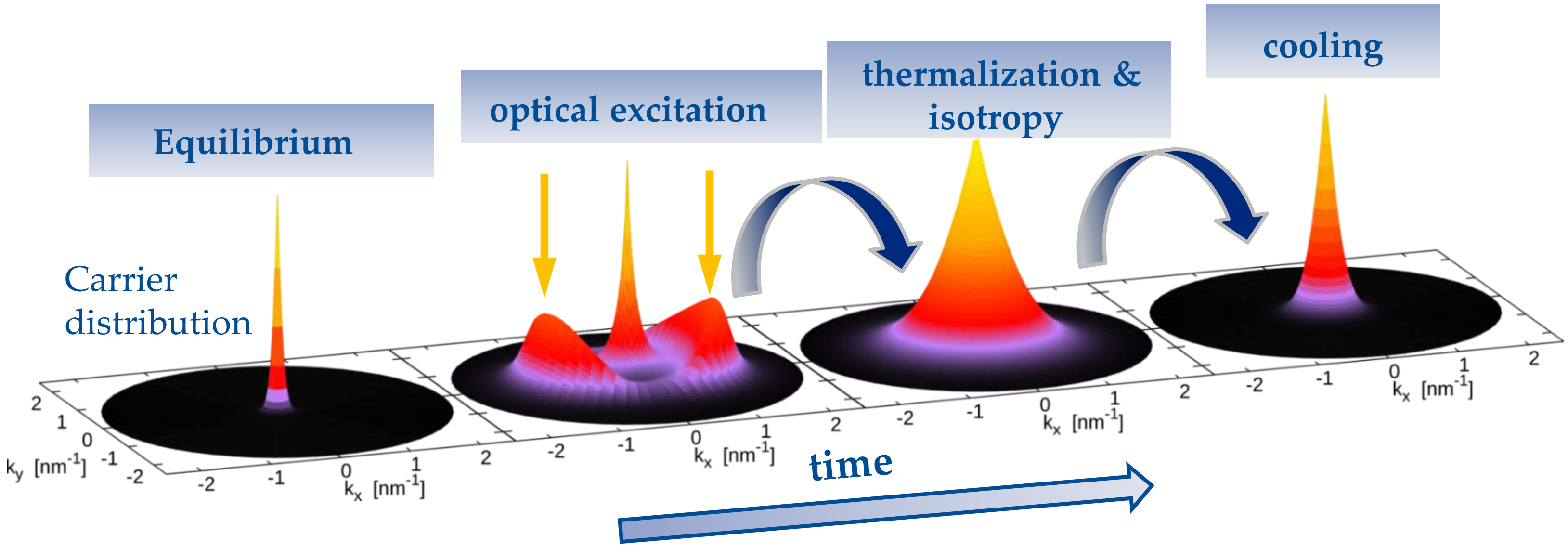
- Microscopic understanding of ultrafast carrier and phonon **relaxation dynamics**

  → Key importance for production of **opto-electronic devices** (photo detectors, lasers, solar cells, etc.)

  → Microscopic **time- and momentum-resolved** calculations of the carrier dynamics



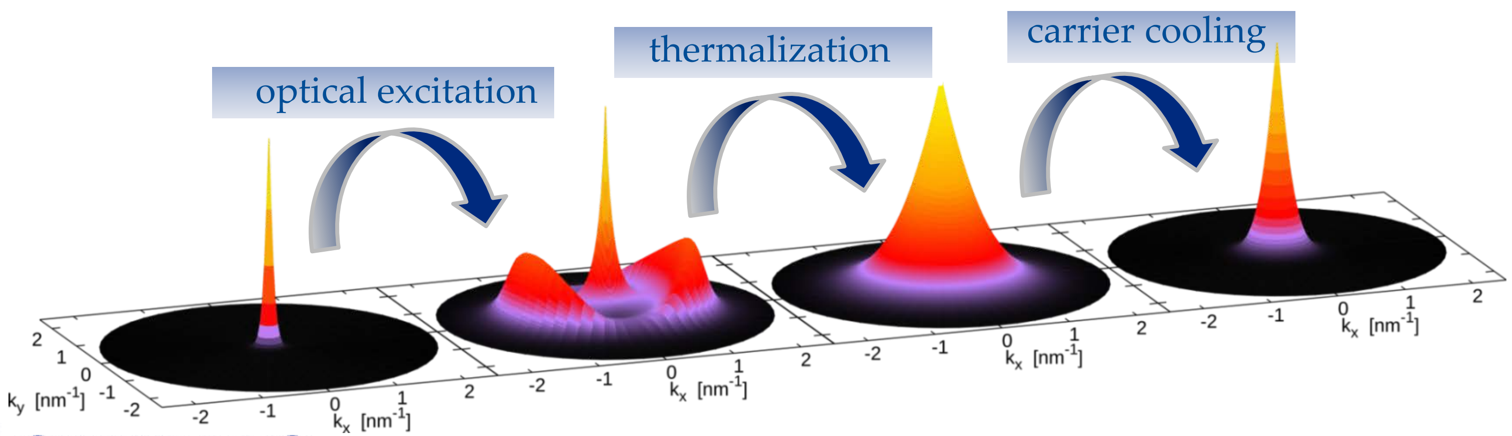
What is relaxation dynamics?



- Optically **excited carriers** relax towards **equilibrium** distribution via **carrier-carrier** and **carrier-phonon scattering**
- Important relaxation steps are **carrier thermalization** and **carrier cooling**

Outline

- Motivation
- Microscopic modelling
- Carrier dynamics
- Many-particle phenomena



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Microscopic quantities

- **Microscopic polarization**

$$p_{\boldsymbol{k}}(t) = \langle a^+_{c\boldsymbol{k}} a_{v\boldsymbol{k}} \rangle$$

- **Occupation probability**

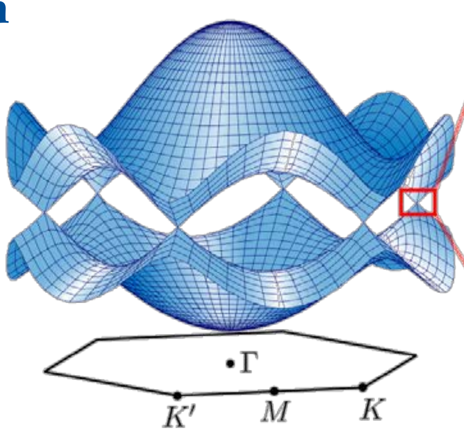
$$\rho^\lambda_{\boldsymbol{k}}(t) = \langle a^+_{\lambda\boldsymbol{k}} a_{\lambda\boldsymbol{k}} \rangle$$

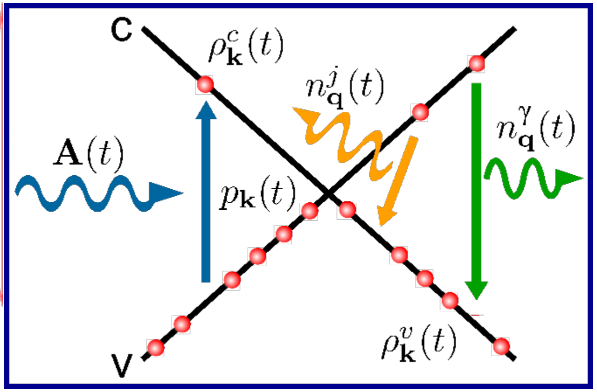
- **Phonon occupation**

$$n^j_{\boldsymbol{q}}(t) = \langle b^+_{j\boldsymbol{q}} b_{j\boldsymbol{q}} \rangle$$

- **Photon occupation**

$$n^\gamma_{\boldsymbol{q}}(t) = \langle c^+_{\gamma\boldsymbol{q}} c_{\gamma\boldsymbol{q}} \rangle$$





- **Second quantization** with creation and annihilation operators a^+, a and b^+, b

- Temporal evolution of quantity O(t) is determined by the **Heisenberg equation of motion**

$$i\hbar \frac{d}{dt} O(t) = [O(t), H]_-$$ ⟵ **Hamilton operator**

⟶ **Graphene Bloch equations** $\dot{\rho}^\lambda_{\boldsymbol{k}}, \dot{p}_{\boldsymbol{k}}, \dot{n}^j_{\boldsymbol{q}}, \dot{n}^\gamma_{\boldsymbol{q}}$

Second quantization

- **Formalism** to describe **quantum many-particle systems** avoiding complicated symmetrisation procedures of the many-particle wave function

- Introduction of **Fock states** (occupation number states)

  $|n_\alpha\rangle = |n_1, n_2, ..., n_\alpha, ...\rangle$ with n_α particles in the $|\alpha\rangle$ state

- Introduction of **creation and annihilation operators** a_α^+, a_α adding and removing a particle in the state $|\alpha\rangle$, respectively

  $$a_\alpha|n_\alpha\rangle = \sqrt{n_\alpha}\,|n_\alpha - 1\rangle, \quad a_\alpha^+|n_\alpha\rangle = \sqrt{n_\alpha + 1}\,|n_\alpha + 1\rangle, \quad a_\alpha^+ a_\alpha\,|n_\alpha\rangle = n_\alpha|n_\alpha\rangle$$

- Any Fock state can be constructed from the vacuum state $|n_\alpha\rangle = \frac{1}{\sqrt{n_\alpha!}}\,(a_\alpha^+)^{n_\alpha}\,|0\rangle$

- Creation and annihilation operators fulfil the **fundamental commutator relations** for fermions (+) and bosons (-)

  $$[a_\alpha, a_{\alpha'}^+]_\pm = \delta_{\alpha,\alpha'}, \quad [a_\alpha, a_{\alpha'}]_\pm = [a_\alpha^+, a_{\alpha'}^+]_\pm = 0$$

  with the commutator $[A, B]_\pm = AB \pm BA$

Second quantization

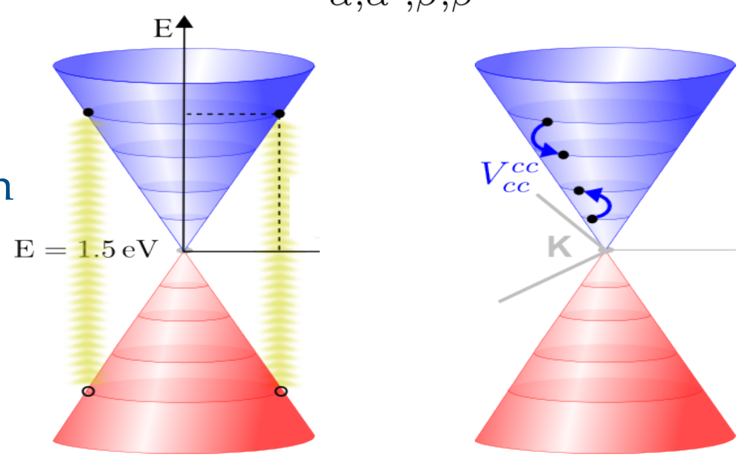
- Most physically relevant many-particle observables O_N can be expressed as a sum of **one-particle** O_1^i and **two-particle operators** $O_2^{i,j}$

$$O_N \approx \sum_i^N O_1^i + \sum_{i,j}^{i \neq j} O_2^{i,j}$$

- The **many-particle operator** reads in second quantization (in the language of creation and annihilation operators)

$$O_N \approx \sum_{\alpha,\beta} \langle \alpha | O_1 | \beta \rangle \, a_\alpha^+ a_\beta \quad + \sum_{\alpha,\alpha',\beta,\beta'} \langle \alpha \, \alpha' | O_2 | \beta \, \beta' \rangle \, a_\alpha^+ a_{\alpha'}^+ a_{\beta'} a_\beta$$

carrier-light interaction
single-particle process



carrier-carrier interaction
two-particle process

Exercise to second quantization

- Calculate the commutators for electrons

$$[\rho_\alpha, H_0]_- = [a_\alpha^+ a_\alpha, \varepsilon_\beta \, a_\beta^+ a_\beta]_- = ?$$

$$[p_{\alpha\alpha'}, H_0]_- = [a_\alpha^+ a_{\alpha'}, \varepsilon_\beta \, a_\beta^+ a_\beta]_- = ?$$

Exercise to second quantization

- Calculate the commutators for electrons

$$[\rho_\alpha, H_0]_- = [a_\alpha^+ a_\alpha, \varepsilon_\beta \, a_\beta^+ a_\beta]_- = 0$$

$$[p_{\alpha\alpha'}, H_0]_- = [a_\alpha^+ a_{\alpha'}, \varepsilon_\beta \, a_\beta^+ a_\beta]_- = (\varepsilon_{\alpha'} - \varepsilon_\alpha) \, a_\alpha^+ a_{\alpha'} = \Delta E \, p_{\alpha\alpha'}$$

Microscopic quantities

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- **Occupation probability**

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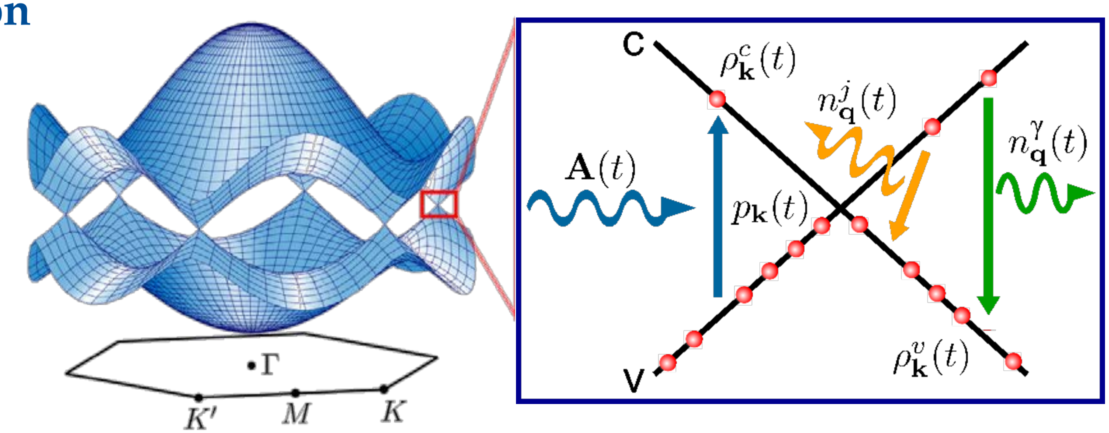

- **Second quantization** with creation and annihilation operators a^+, a and b^+, b

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→ **Graphene Bloch equations** $\dot{\rho}^{\lambda}_{\boldsymbol{k}}, \dot{p}_{\boldsymbol{k}}, \dot{n}^{j}_{\boldsymbol{q}}, \dot{n}^{\gamma}_{\boldsymbol{q}}$

Many-particle Hamilton operator

- Many-particle Hamiltonian in the language of second quantization

$$H = H_0 + H_{c-l} + H_{c-c} + H_{c-ph}$$

free-particle **carrier-light interaction** **carrier-carrier interaction**

$$= \sum_{l} \varepsilon_{l} a_{l}^{+} a_{l} + \frac{ie_0\hbar}{m_0} \sum_{l,l'} M_{l,l'} \cdot \boldsymbol{A}(t) a_{l}^{+} a_{l'} + \frac{1}{2} \sum_{l_1,l_2,l_3,l_4} V_{l_3,l_4}^{l_1,l_2} a_{l_1}^{+} a_{l_2}^{+} a_{l_4} a_{l_3}$$

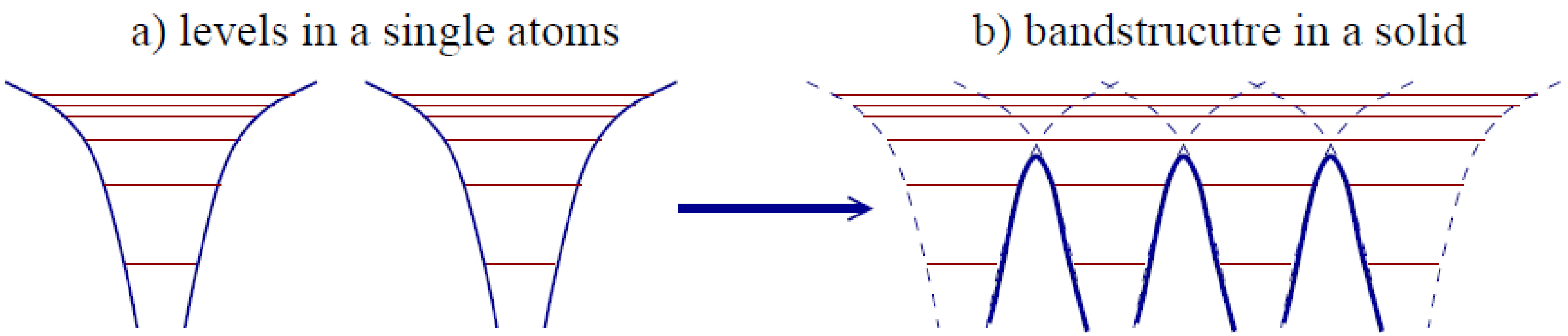
$$+ \sum_{i} \hbar\omega_{i} b_{i}^{+} b_{i} + \sum_{l,l'} \sum_{i} \left(g_{l,l'}^{i} a_{l}^{+} b_{i}\, a_{l'} + h.c. \right)$$

carrier-phonon interaction

- To calculate the material-specific bandstructure and matrix elements, we need the **many-particle wave function**

Tight-binding aproach

- The tight-binding (TB) method is based on the assumption that **electrons are tightly bound** to their nuclei
- Start from isolated atoms, their **wave functions overlap** and lead to chemical bonds and to the formation of crystals, when the atoms get close enough
- Due to the appearing interactions, the **electronic energies broaden and build continuous bands**



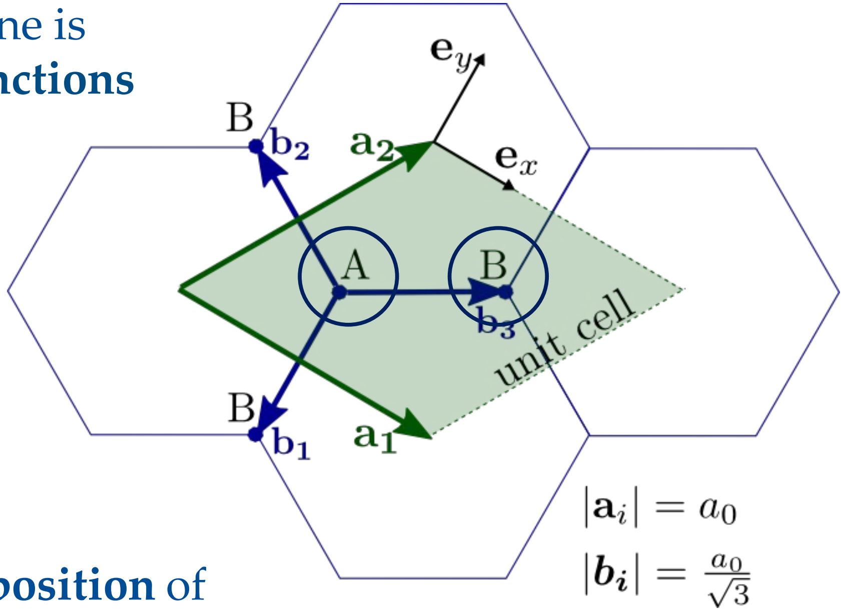
Tight-binding wave functions

- The required band structure for graphene is calculated with **tight-binding wave functions**

$$\psi^{\lambda}_{\boldsymbol{k}}(\boldsymbol{r}) = C^{\lambda}_{\boldsymbol{k},A}\Phi_{\boldsymbol{k},A}(\boldsymbol{r}) + C^{\lambda}_{\boldsymbol{k},B}\Phi_{\boldsymbol{k},B}(\boldsymbol{r})$$

$$\Phi_{\boldsymbol{k},i}(\boldsymbol{r}) = \frac{1}{\sqrt{N}}\sum_{\boldsymbol{R}_j} e^{i\boldsymbol{k}\cdot\boldsymbol{R}_j}\phi_j(\boldsymbol{r}-\boldsymbol{R}_j)$$

with **$2p_z$-orbital** functions $\phi_j(\boldsymbol{r}-\boldsymbol{R}_j)$



- TB wave functions are based on **superposition** of wave functions for isolated atoms located at each atomic site

- We take $2p_z$ orbitals **from hydrogen atom with an effective atomic number**

- We apply the **nearest-neighbor TB approximation** considering only overlaps of the next lying three neighboring atoms

$$\frac{1}{N}\sum_{\boldsymbol{R}_A,\boldsymbol{R}_B} e^{i\boldsymbol{k}\cdot(\boldsymbol{R}_B-\boldsymbol{R}_A)} = \sum_{i=1}^{3} e^{i\boldsymbol{k}\cdot\boldsymbol{b}_i} = e(\boldsymbol{k})$$

Electronic bandstructure

- Solve the **eigenvalue problem** $H\,\psi_{\boldsymbol{k}}^{\lambda}(\boldsymbol{r}) = E_{\boldsymbol{k}}^{\lambda}\,\psi_{\boldsymbol{k}}^{\lambda}(\boldsymbol{r})$

- Multiply with $\Phi_{\boldsymbol{k},A}^{*}(\boldsymbol{r})$ and $\Phi_{\boldsymbol{k},B}(\boldsymbol{r})$, separately and integrate over r leading to a set of coupled equations

$$\begin{pmatrix} H_{AA} - \varepsilon_{\mathrm{k}} S_{AA} & H_{AB} - \varepsilon_{\mathrm{k}} S_{AB} \\ H_{BA} - \varepsilon_{\mathrm{k}} S_{BA} & H_{BB} - \varepsilon_{\mathrm{k}} S_{BB} \end{pmatrix} \begin{pmatrix} c_A(\mathrm{k}) \\ c_B(\mathrm{k}) \end{pmatrix} = 0$$

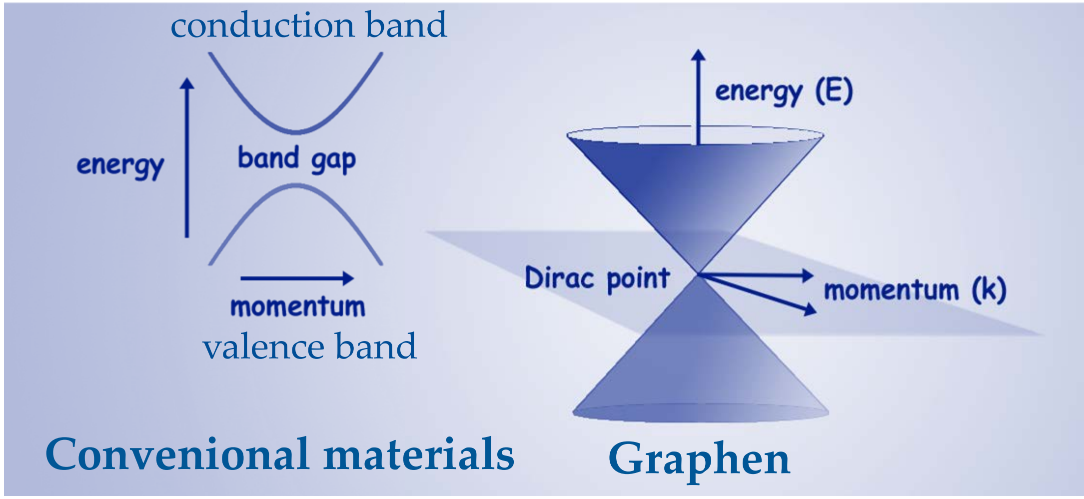
that can be solved by evaluating the **secular equation**

$\det[\hat{\boldsymbol{H}} - E_{\boldsymbol{k}}^{\lambda}\,\hat{\boldsymbol{S}}] = 0$ with $H_{i,i'} = \langle \Phi_i | H | \Phi_{i'} \rangle$ and $S_{i,i'} = \langle \Phi_i | \Phi_{i'} \rangle$

- Exploit the equivalence of the A and B atoms with $H_{AA} = H_{BB},\, H_{AB} = H_{BA}^{*}$, and assume the **nearest-neighbour approximation** with

$$H_{AB} = \frac{1}{N} \sum_{\boldsymbol{R}_A, \boldsymbol{R}_B} e^{i\boldsymbol{k}\cdot(\boldsymbol{R}_B - \boldsymbol{R}_A)} \langle \phi(\boldsymbol{r} - \boldsymbol{R}_A) | H | \phi(\boldsymbol{r} - \boldsymbol{R}_B) \rangle = \gamma_0\, e(\boldsymbol{k})$$

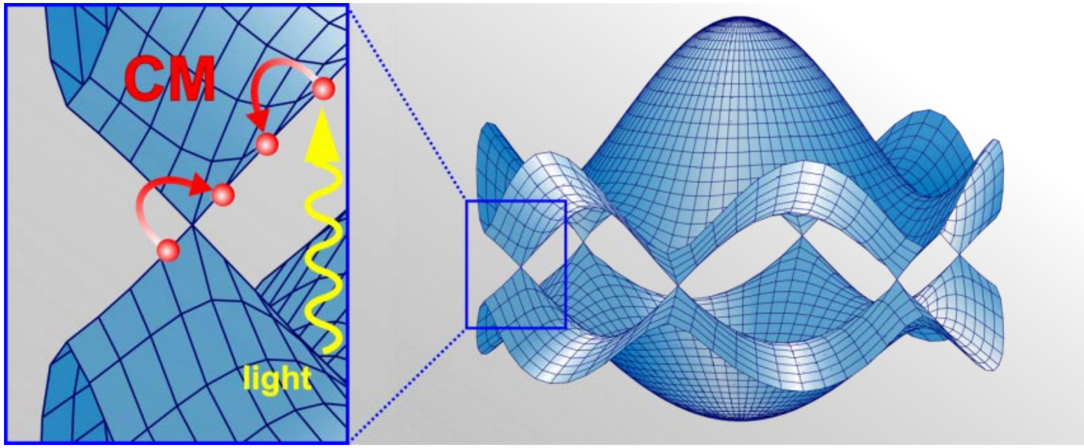
Electronic bandstructure



- **Linear energy-impulse dependence** close to the Dirac point

$$E = \alpha |k|$$

- Graphene has **no band gap** (semi-metal or zero-gap semiconductor)



- **Linear and gapless bandstructure** gives rise to **new carrier relaxation channels**

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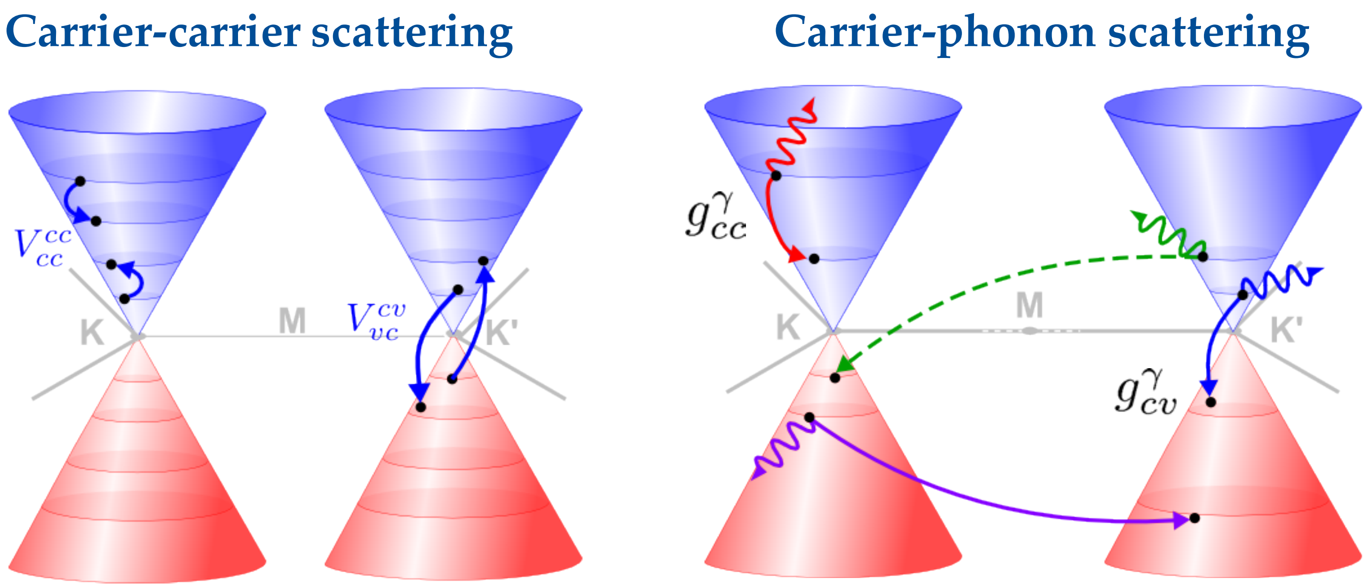
Many-particle scattering channels



- Excited carriers relax towards lower energies via **intra-** and **inter-band scattering**
- **Carrier-phonon** scattering gives rise to **carrier cooling**
- Phonon-induced **intervalley** processes can be very efficient

Think-pair-share: Linear bandstructure

- What do you think why the linear and gapless bandstructure of graphene can be important in terms of technological application?

Many-particle Hamilton operator

- Many-particle Hamiltonian in the language of second quantization

$$H = H_0 + H_{c-l} + H_{c-c} + H_{c-ph}$$

free-particle **carrier-light interaction** **carrier-carrier interaction**

$$= \sum_{\boldsymbol{l}} \varepsilon_{\boldsymbol{l}} a_{\boldsymbol{l}}^{+} a_{\boldsymbol{l}} + \frac{ie_0\hbar}{m_0} \sum_{\boldsymbol{l},\boldsymbol{l}'} M_{\boldsymbol{l},\boldsymbol{l}'} \cdot \boldsymbol{A}(t) a_{\boldsymbol{l}}^{+} a_{\boldsymbol{l}'} + \frac{1}{2} \sum_{\boldsymbol{l}_1,\boldsymbol{l}_2,\boldsymbol{l}_3,\boldsymbol{l}_4} V_{\boldsymbol{l}_3,\boldsymbol{l}_4}^{\boldsymbol{l}_1,\boldsymbol{l}_2} a_{\boldsymbol{l}_1}^{+} a_{\boldsymbol{l}_2}^{+} a_{\boldsymbol{l}_4} a_{\boldsymbol{l}_3}$$

$$+ \sum_{\boldsymbol{i}} \hbar\omega_{\boldsymbol{i}} b_{\boldsymbol{i}}^{+} b_{\boldsymbol{i}} + \sum_{\boldsymbol{l},\boldsymbol{l}'} \sum_{\boldsymbol{i}} \left(g_{\boldsymbol{l},\boldsymbol{l}'}^{\boldsymbol{i}} a_{\boldsymbol{l}}^{+} b_{\boldsymbol{i}}\, a_{\boldsymbol{l}'} + h.c. \right)$$

carrier-phonon interaction

- **To calculate the material-specific bandstructure and matrix elements,** we need the many-particle wave function

Optical matrix element

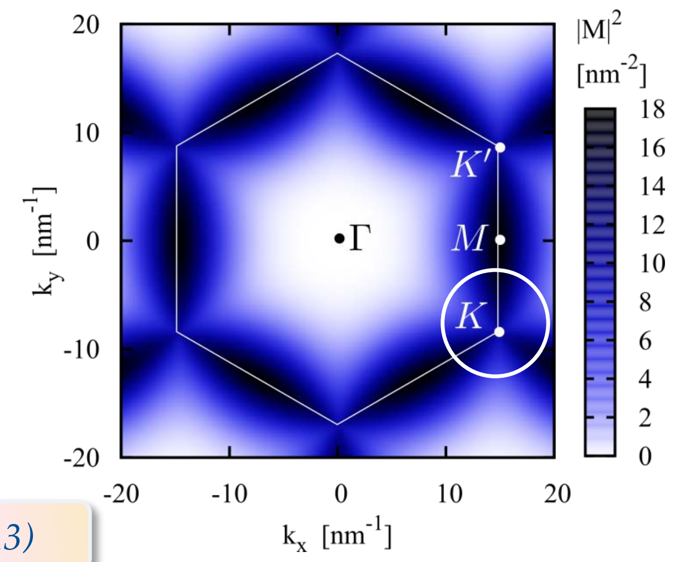
- Optical matrix element $M_{l,l'} = \langle \Psi_l(\boldsymbol{r}) \,|\, \boldsymbol{p} \,|\, \Psi_{l'}(\boldsymbol{r}) \rangle$ determines the **strength** of the **carrier-light coupling** and includes **optical selection rules**

- Analytic expression can be obtained within the **nearest- neighbor tight-binding** approximation yielding

$$M_{\boldsymbol{k}}^{\lambda\lambda'} = m \sum_{i=1}^{3} \frac{\boldsymbol{b}_i}{|\boldsymbol{b}_i|} \left(C_{A*}^{\lambda}(\boldsymbol{k}) C_{B}^{\lambda'}(\boldsymbol{k}) e^{i\boldsymbol{k}\cdot\boldsymbol{b}_i} - C_{B*}^{\lambda}(\boldsymbol{k}) C_{A}^{\lambda'}(\boldsymbol{k}) e^{-i\boldsymbol{k}\cdot\boldsymbol{b}_i} \right)$$

- **Carrier-light coupling** is strongly **anisotropic** around the Dirac points (K points)

- It shows **maxima** at **M points** and **vanishes** at the **Γ point** of the Brillouin zone (selection rule)



PRB 84, 205406 (2011)

E. Malic and A. Knorr, Wiley (2013)

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Coulomb matrix element

- The Coulomb matrix element reads (with compound indices $l_i = k_i, \lambda_i$)

$$V^{l_1,l_2}_{l_3,l_4} = \int d\boldsymbol{r} \int d\boldsymbol{r}' \Psi^*_{l_1}(\boldsymbol{r})\Psi^*_{l_2}(\boldsymbol{r}')\, V(\boldsymbol{r}-\boldsymbol{r}')\, \Psi_{l_4}(\boldsymbol{r}')\Psi_{l_3}(\boldsymbol{r})$$

- Within the **nearest-neighbor tight-binding** approximation, we obtain

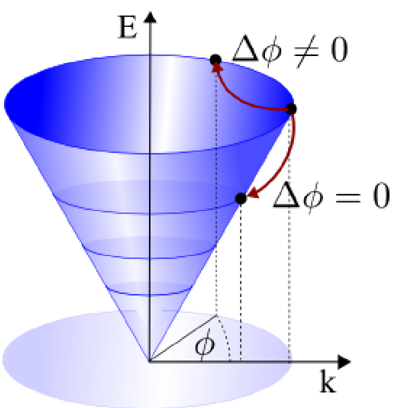
$$V^{l_1,l_2}_{l_3,l_4} = \frac{e_o^2}{2\varepsilon_0 q}\left[\left(\frac{q\, a_B}{Z_{eff}}\right)^2 + 1\right]^{-6} \alpha^{l_1,l_2}_{l_3,l_4}\, \boxed{\delta_{\boldsymbol{q},\boldsymbol{k}_3-\boldsymbol{k}_1}\,\delta_{\boldsymbol{q},\boldsymbol{k}_4-\boldsymbol{k}_2}}$$

← **momentum conservation**

with TB-coefficients $\alpha^{l_1,l_2}_{l_3,l_4} = \frac{1}{4}\left(1 + c_{\lambda_1\lambda_3}\frac{e^*(\boldsymbol{k}_1)e(\boldsymbol{k}_3)}{|e(\boldsymbol{k}_1)e(\boldsymbol{k}_3)|}\right)\left(1 + c_{\lambda_2\lambda_4}\frac{e^*(\boldsymbol{k}_2)e(\boldsymbol{k}_4)}{|e(\boldsymbol{k}_2)e(\boldsymbol{k}_4)|}\right)$

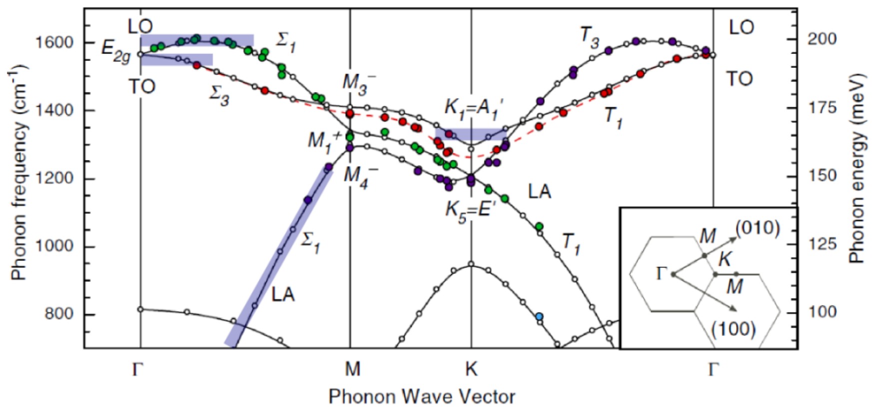
- Coulomb processes with **large momentum transfer** are **strongly suppressed** (decay scales with ***$1/q^{13}$***)

- Coulomb interaction $V \propto 1 + e^{i\Delta\phi}$ **prefers parallel intraband scattering** along the Dirac cone



Carrier-phonon matrix element

- Focus on **strongly coupling optical phonons** (*ΓLO, ΓTO, K*)



J. Maultsch et al., PRL 92, 75501 (2004)

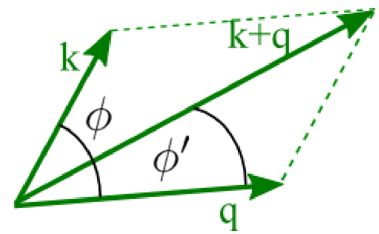
- Carrier-phonon **matrix elements**

$$g_{\boldsymbol{q},j}^{\boldsymbol{k}\boldsymbol{k}',\lambda\lambda'} = \langle \Psi_{\boldsymbol{k},\lambda}(\boldsymbol{r}) \,|\, \Delta V_{\boldsymbol{q},\gamma} \,|\, \Psi_{\boldsymbol{k}',\lambda'}(\boldsymbol{r}) \rangle$$

can be expressed as (Mauri et al.):

$$|g_{\boldsymbol{q}\Gamma j}^{\boldsymbol{k}\boldsymbol{k}',\lambda\lambda'}|^2 = \frac{a_0\sqrt{3}}{2A}\tilde{g}_{\Gamma}^2 \left(1 + c_j^{\lambda\lambda'} cos(\phi + \phi')\right)$$

$$|g_{\boldsymbol{q}K}^{\boldsymbol{k}\boldsymbol{k}',\lambda\lambda'}|^2 = \frac{a_0\sqrt{3}}{2A}\tilde{g}_{K}^2 \left(1 - c_K^{\lambda\lambda'} cos(\phi - \phi')\right)$$



with $\tilde{g}_{\Gamma}^2 = 0.0405eV^2, \tilde{g}_K^2 = 0.0994eV^2$

which can be extracted from experiment exploiting **Kohn anomalies**

- Phonon-induced intra- ($\lambda = \lambda$\`) and interband ($\lambda \neq \lambda$\`) scattering shows a distinct **angle-dependence** for different phonon modes

Correlation expansion

- Hamilton operator H is known → derivation of **Bloch equations** $\dot{\rho}^{\lambda}_{\boldsymbol{k}}, \dot{p}_{\boldsymbol{k}}, \dot{n}^{j}_{\boldsymbol{q}}, \dot{n}^{\gamma}_{\boldsymbol{q}}$ applying the Heisenberg equation $i\hbar\dot{\rho}^{\lambda}_{\boldsymbol{k}} = [\rho^{\lambda}_{\boldsymbol{k}}, H]_-$

- Many-particle interaction (e.g. carrier-carrier coupling) leads to a **hierarchy problem** (system of equations is not closed)

$$\frac{d}{dt}\langle a_1^+ a_2 \rangle \;\propto\; \langle a_A^+ a_B^+ a_C a_D \rangle$$

$$\frac{d}{dt}\langle a_A^+ a_B^+ a_C a_D \rangle \;\propto\; \langle a_1^+ a_2^+ a_3^+ a_4\, a_5\, a_6 \rangle \quad \ldots$$

- Solution by applying the **correlation expansion** and systematic truncation

  Example: **Hartree-Fock** factorization (single-particle quantities only)

$$\langle a_A^+ a_B^+ a_C a_D \rangle = \langle a_A^+ a_D \rangle \langle a_B^+ a_C \rangle - \langle a_A^+ a_C \rangle \langle a_B^+ a_D \rangle + \langle a_A^+ a_B^+ a_C a_D \rangle^c$$

→ **closed system of equations** (already sufficient for description of **excitons**)

Markov approximaiton

- For description of **scattering processes**, dynamics of two-particle quantities $\sigma_{ABCD} = \langle a_A^+ a_B^+ a_C a_D \rangle$ is necessary (**second-order Born**)

$$\frac{d}{dt}\sigma_{ABCD}(t) = \frac{i}{\hbar}\Delta\varepsilon\, \sigma_{ABCD}(t) + \frac{i}{\hbar}Q(t) - \gamma\, \sigma_{ABCD}(t)$$

with the scattering term *Q(t)* including only single-particle quantities

- Für 2-dim systems, such as graphene with $A = (k_x, k_y, \lambda)$, the evaluation of equations is a **numerical challenge** (memory, CPU time)

- **Markov approximation** neglects quantum-kinetic memory effects:

$$\sigma_{ABCD}(t) = \frac{i}{\hbar}\int_0^{\infty} e^{\left(\frac{i}{\hbar}\Delta\varepsilon+\gamma\right)s}\, Q(t-s)\, ds \quad \approx \quad -i\pi\, Q(t)\, \delta\left(\Delta\varepsilon\right) \qquad (\gamma \to 0)$$

→ **closed system of equations**

Graphene Bloch equations

$$\dot{\rho}^{\lambda}_{\mathbf{k}}(t) = 2\mathrm{Im}\left(\Omega^{*}_{\mathrm{k}}(t)\mathrm{p}_{\mathrm{k}}(t)\right) + \Gamma^{in}_{\boldsymbol{k},\lambda}(t)\left(1-\rho^{\lambda}_{\boldsymbol{k}}(t)\right) - \Gamma^{out}_{\boldsymbol{k},\lambda}(t)\rho^{\lambda}_{\boldsymbol{k}}(t)$$

$$\dot{p}_{\mathbf{k}}(t) = -i\omega_{\boldsymbol{k}}p_{\boldsymbol{k}}(t) - i\Omega_{\boldsymbol{k}}(t)\left(\rho^{c}_{\boldsymbol{k}}(t) - \rho^{v}_{\boldsymbol{k}}(t)\right) - \gamma_{2,\boldsymbol{k}}(t)\,p_{\boldsymbol{k}}(t) + \tilde{\gamma}_{2,\boldsymbol{k}'}(t)$$

$$\dot{n}^{j}_{\mathbf{q}}(t) = \Gamma^{out}_{j,\mathbf{q}}(t)\left(n^{j}_{\mathbf{q}}(t)+1\right) - \Gamma^{in}_{j,\mathbf{q}}(t)n^{j}_{\mathbf{q}}(t) - \gamma_j\left(n^{j}_{\boldsymbol{q}}(t) - n_0\right)$$

$$H = H_0 + H_{c-l} + H_{c-c} + H_{c-ph}$$

- **Carrier-light coupling** gives rise to a **non-equilibrium distribution** of electrons after optical excitation with a laser pulse

Graphene Bloch equations

$$\dot{\rho}^{\lambda}_{\mathbf{k}}(t) = 2\mathrm{Im}\left(\Omega^*_{\mathrm{k}}(\mathrm{t})\mathrm{p}_{\mathrm{k}}(\mathrm{t})\right) + \Gamma^{in}_{\boldsymbol{k},\lambda}(t)\left(1-\rho^{\lambda}_{\boldsymbol{k}}(t)\right) - \Gamma^{out}_{\boldsymbol{k},\lambda}(t)\rho^{\lambda}_{\boldsymbol{k}}(t)$$

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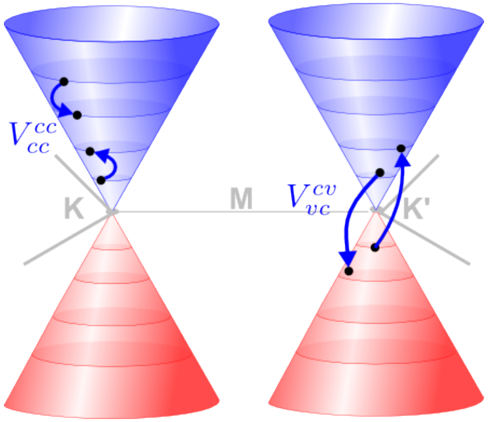
- **Time-** and **momentum-dependent** carrier-carrier and carrier-phonon **scattering rates** $\Gamma^{in}_{\boldsymbol{k},\lambda}(t) = \Gamma^{in,cc}_{\boldsymbol{k},\lambda}(t) + \Gamma^{in,cp}_{\boldsymbol{k},\lambda}(t)$

Coulomb matrix elements

$$\Gamma^{in,cc}_{\boldsymbol{k},\lambda} = \frac{2\pi}{\hbar} \sum_{\boldsymbol{l}_1,\boldsymbol{l}_2,\boldsymbol{l}_3} W^{\boldsymbol{k}\lambda\,\boldsymbol{l}_1\,*}_{\boldsymbol{l}_2\,\boldsymbol{l}_3}\left(2W^{\boldsymbol{k}\lambda\,\boldsymbol{l}_1\,*}_{\boldsymbol{l}_2\,\boldsymbol{l}_3} - W^{\boldsymbol{k}\lambda\,\boldsymbol{l}_1\,*}_{\boldsymbol{l}_3\,\boldsymbol{l}_2}\right) \times \rho_{\boldsymbol{l}_2}\rho_{\boldsymbol{l}_3}(1-\rho_{\boldsymbol{l}_1})\delta(\varepsilon_{\boldsymbol{k}\lambda} + \varepsilon_{\boldsymbol{l}_1} - \varepsilon_{\boldsymbol{l}_2} - \varepsilon_{\boldsymbol{l}_3})$$

Pauli blocking



Graphene Bloch equations

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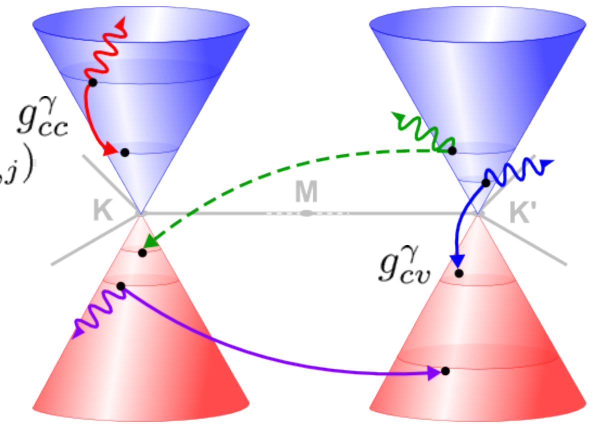
phonon emission

$$\Gamma^{in,cp}_{\boldsymbol{k},\lambda} = \sum_{\boldsymbol{q},j,\lambda'} |g^{\boldsymbol{k}\boldsymbol{k}',\lambda\lambda'}_{\boldsymbol{q},j}|^2 f^{\lambda'}_{\boldsymbol{k}+\boldsymbol{q}}\Big((n^j_{\boldsymbol{q}}+1)\delta(\varepsilon_{\boldsymbol{k}+\boldsymbol{q},\lambda'}-\varepsilon_{\boldsymbol{k},\lambda}-\hbar\omega_{\boldsymbol{q},j}) + n^j_{\boldsymbol{q}}\delta(\varepsilon_{\boldsymbol{k}+\boldsymbol{q},\lambda'}-\varepsilon_{\boldsymbol{k},\lambda}+\hbar\omega_{\boldsymbol{q},j})\Big)$$

phonon absorption



Graphene Bloch equations

$$\dot{\rho}^{\lambda}_{\mathbf{k}}(t) = 2\mathrm{Im}\left(\Omega^{*}_{\mathrm{k}}(\mathrm{t})\mathrm{p}_{\mathrm{k}}(\mathrm{t})\right) + \Gamma^{in}_{\boldsymbol{k},\lambda}(t)\left(1 - \rho^{\lambda}_{\boldsymbol{k}}(t)\right) - \Gamma^{out}_{\boldsymbol{k},\lambda}(t)\rho^{\lambda}_{\boldsymbol{k}}(t)$$

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$$H = H_0 + H_{c-l} + H_{c-c} + H_{c-ph}$$

- **Diagonal** and **non-diagonal dephasing** of microscopic polarization

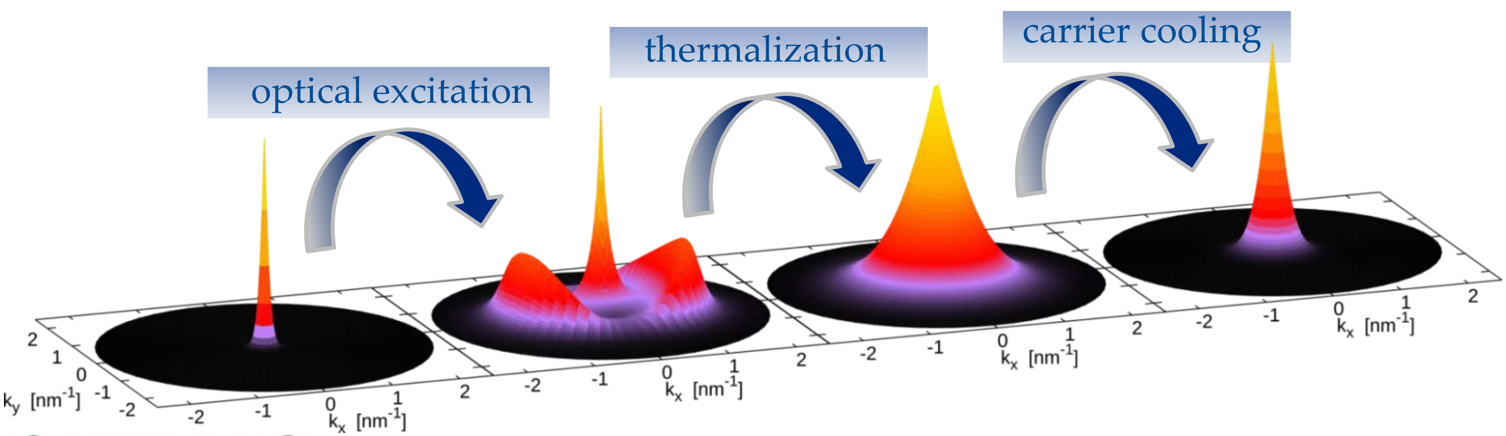
$$\gamma_{2,\boldsymbol{k}}(t) = \tfrac{1}{2}\sum_{\lambda}\left(\Gamma^{in}_{\boldsymbol{k},\lambda}(t) + \Gamma^{out}_{\boldsymbol{k},\lambda}(t)\right)$$

$$\tilde{\gamma}_{2,\boldsymbol{k}}(t) = \sum_{\boldsymbol{k}'}\left(T^{a}_{\boldsymbol{k}\boldsymbol{k}'}(t)p_{\boldsymbol{k}'}(t) + T^{b}_{\boldsymbol{k}\boldsymbol{k}'}(t)p^{*}_{\boldsymbol{k}'}(t)\right)$$

E. Malic and A. Knorr, Wiley (2013)

Outline

- Motivation
- Microscopic modelling
- **Carrier dynamics**
- Many-particle phenomena



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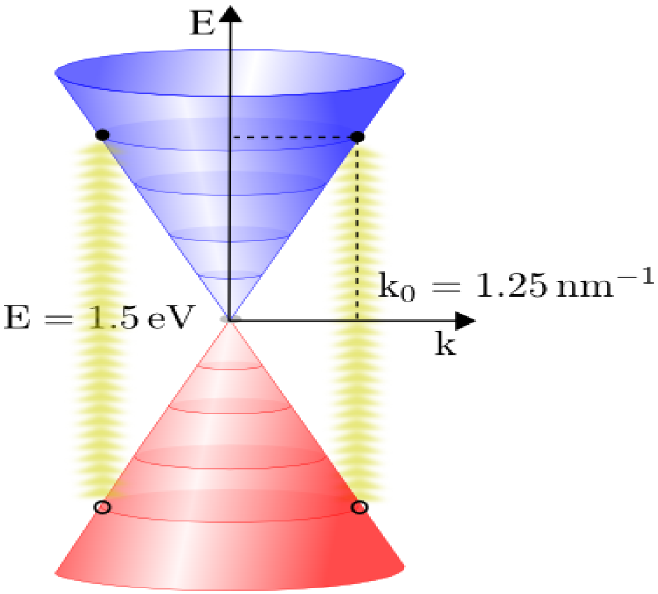
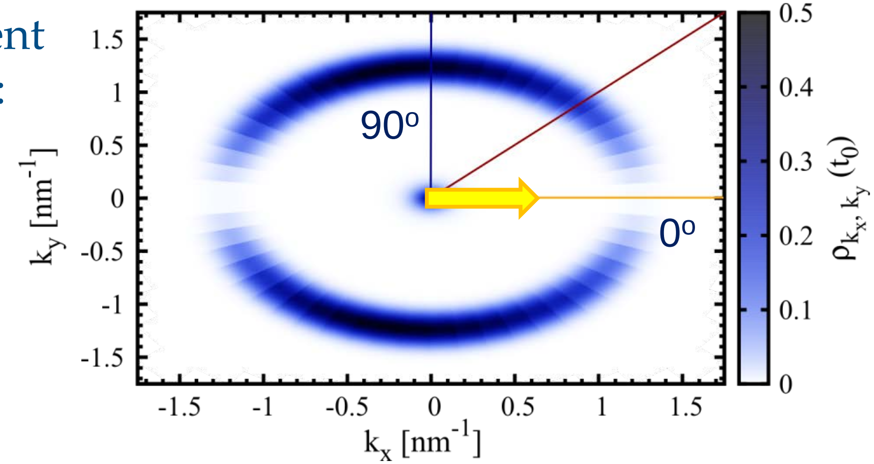
Generation of a non-equilibrium

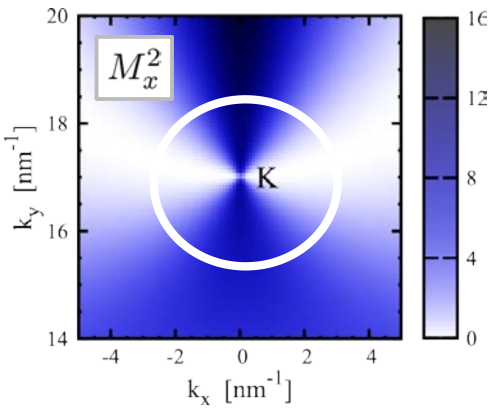
- **Optical excitation** according to a recent **experiment** (T. Elsaesser, MBI Berlin):
  - pulse width **10 fs**
  - excitation energy **1.5 eV**
  - pump fluence **1 μJcm^{-2}**



Generation of a non-equilibrium

- **Optical excitation** according to a recent **experiment** (T. Elsaesser, MBI Berlin):
  - pulse width **10 fs**
  - excitation energy **1.5 eV**
  - pump fluence **1 μJcm^{-2}**
- Generation of an **anisotropic non-equilibrium** carrier distribution
- **Maximal occupation** perpendicular to polarization of excitation pulse (**90°**)
- Origin lies in the **anisotropy** of the **carrier-light coupling** element





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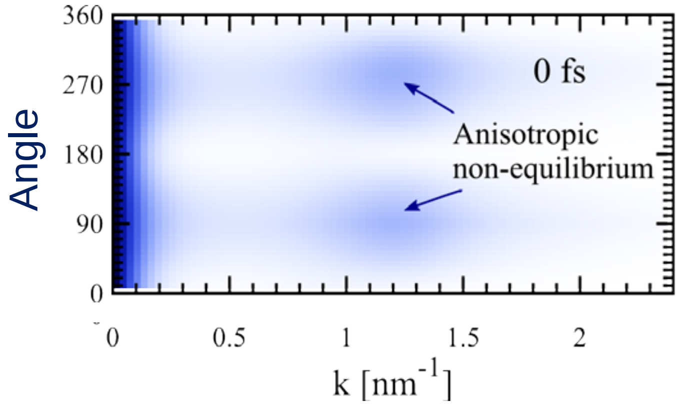
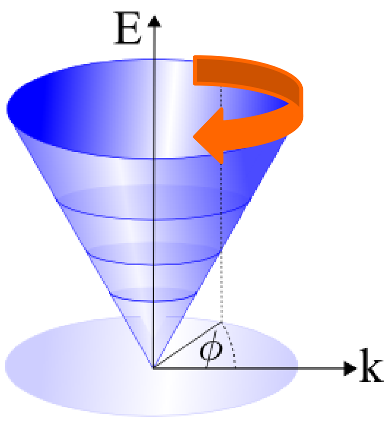
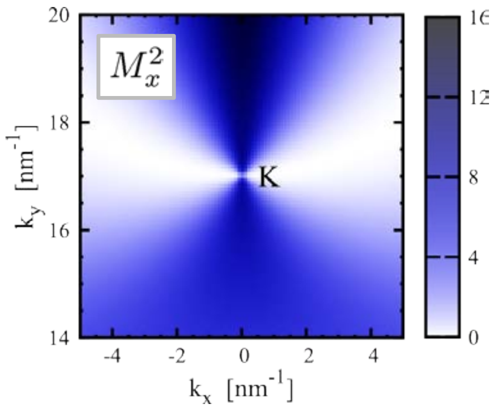
PRB 84, 205406 (2011)

Anisotropic carrier distribution

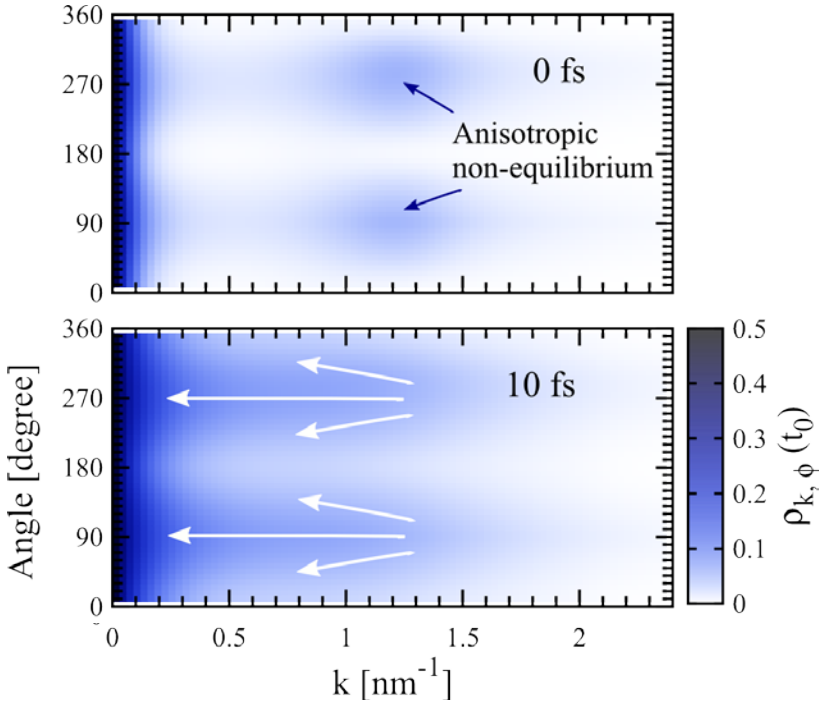




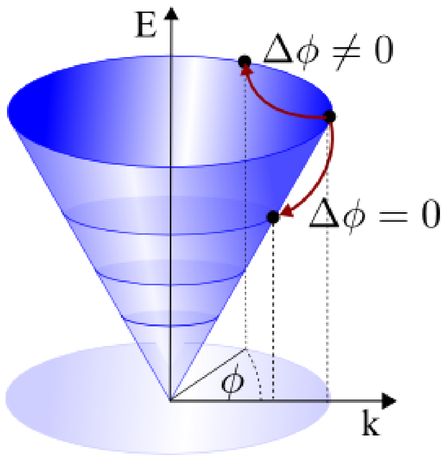
- Generation of an **anisotropic non-equilibrium** carrier distribution



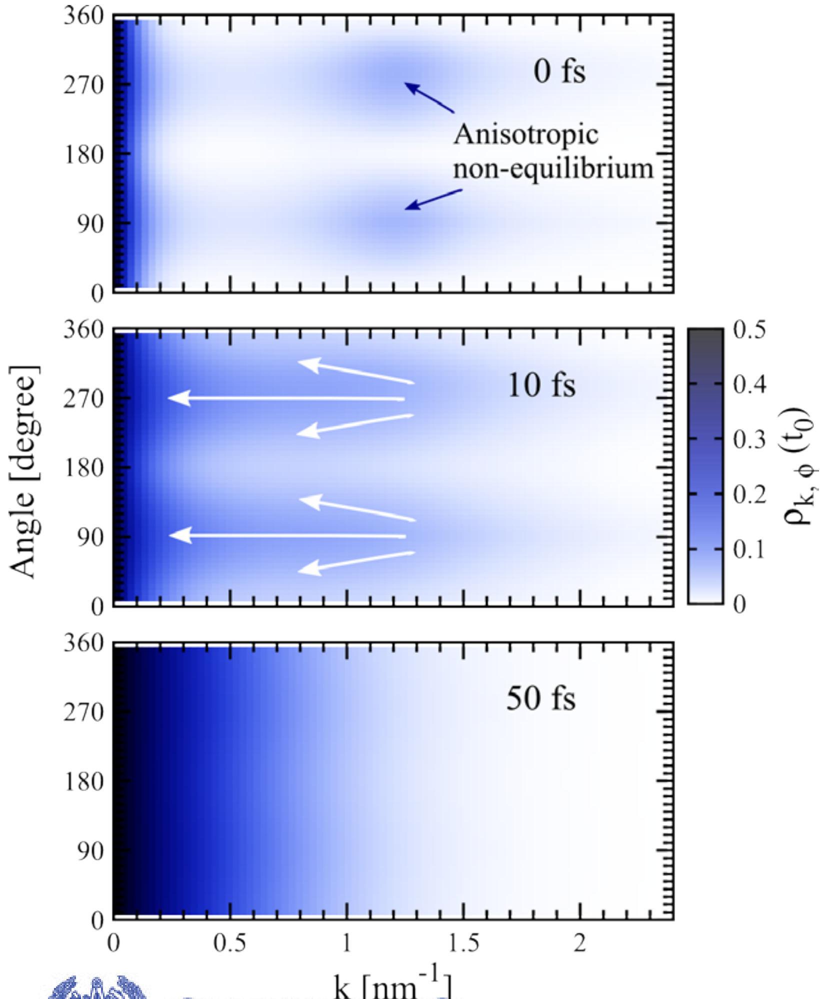
Anisotropic carrier dynamics



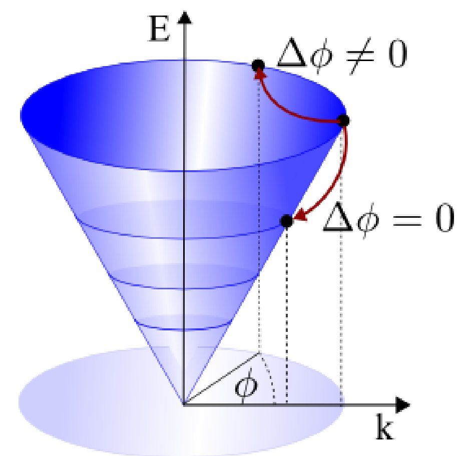
- Generation of an **anisotropic non-equilibrium** carrier distribution
- Scatering **across** the Dirac **cone reduces** the **anisotropy**



Anisotropic carrier dynamics



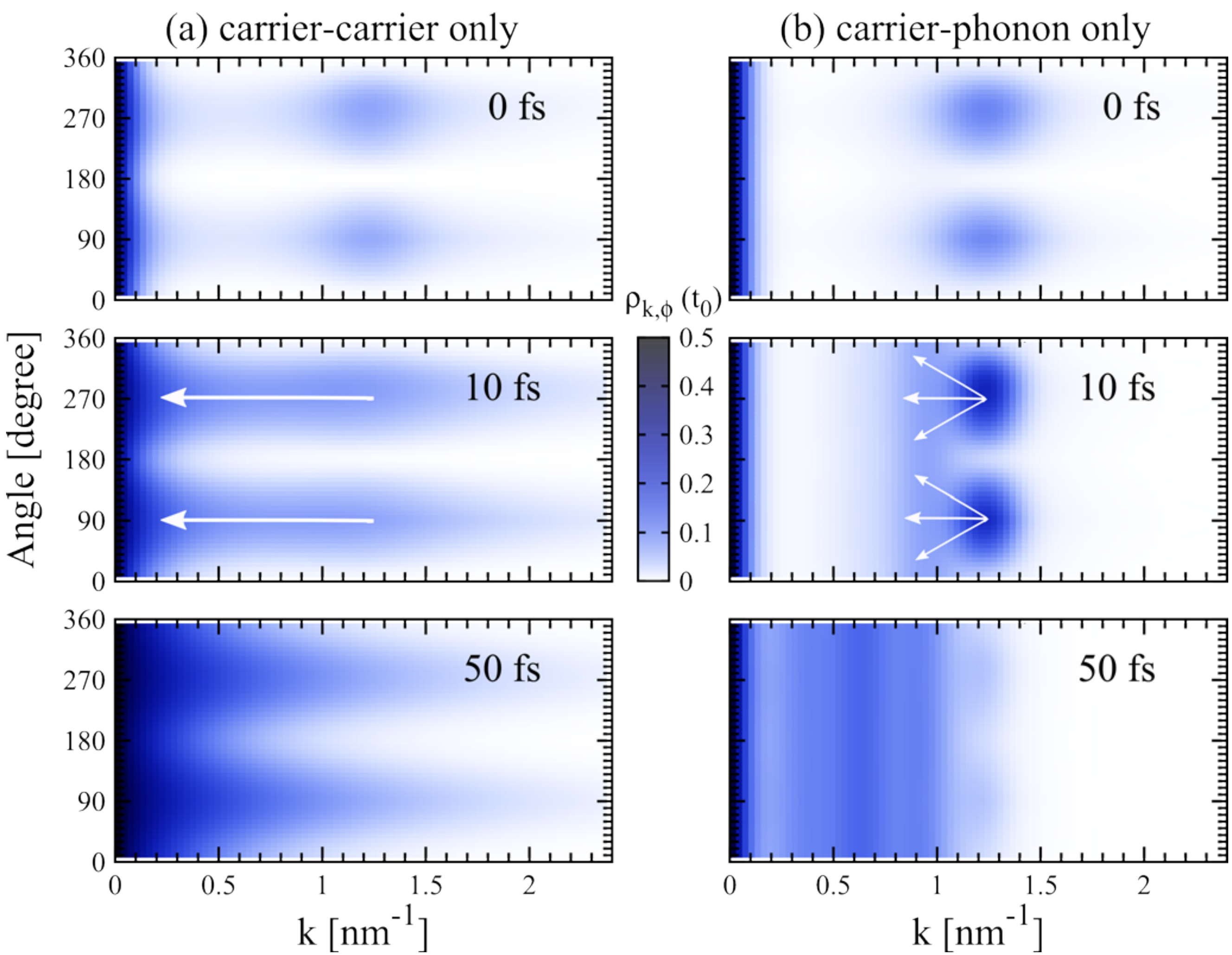
- Generation of an **anisotropic non-equilibrium** carrier distribution
- Scatering **across** the Dirac **cone reduces** the **anisotropy**



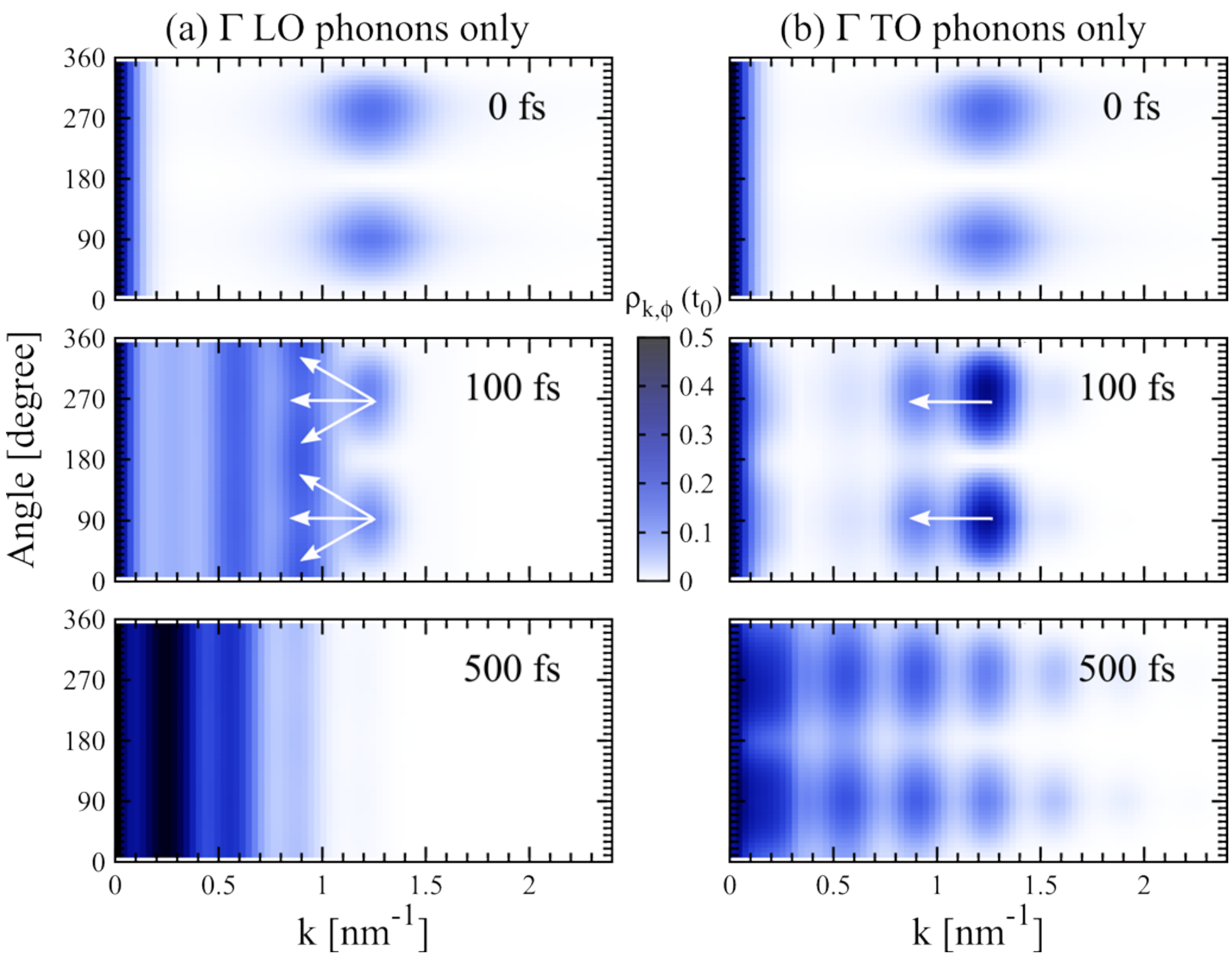
- Carrier distribution becomes entirely **isotropic** within the first **100 fs**

APL 101, 213110 (2012)

Microscopic mechanism



Different phonon modes



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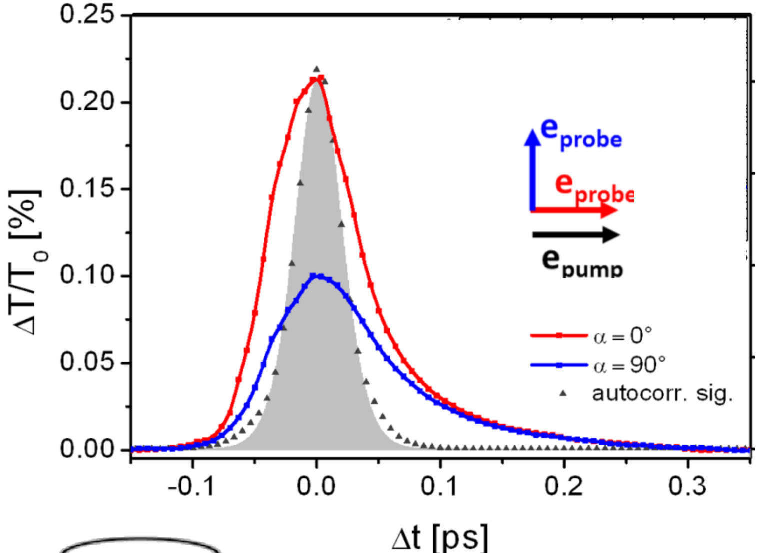
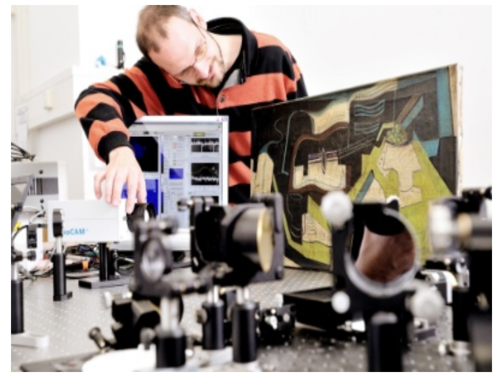
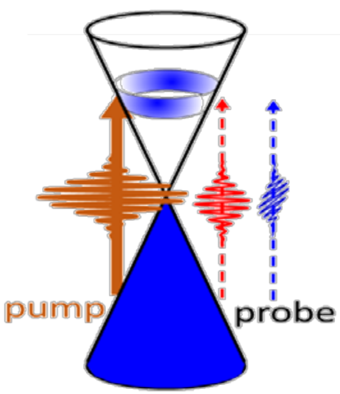
Experiment-theory comparison

Experiment



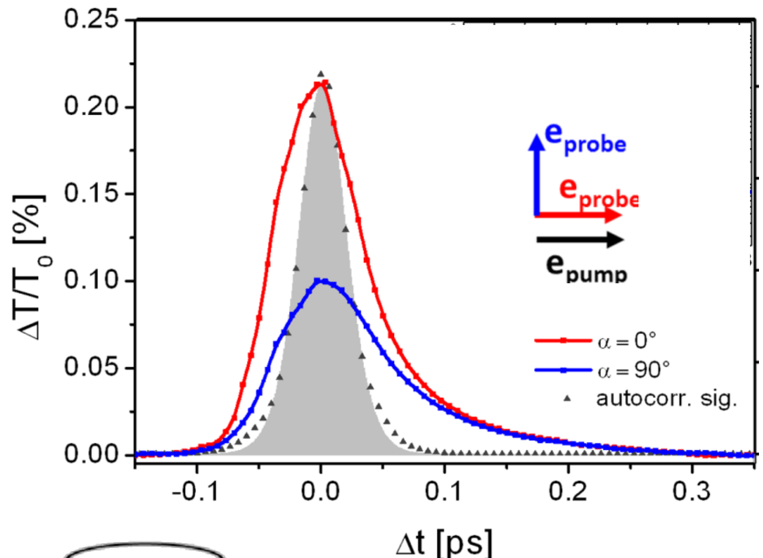




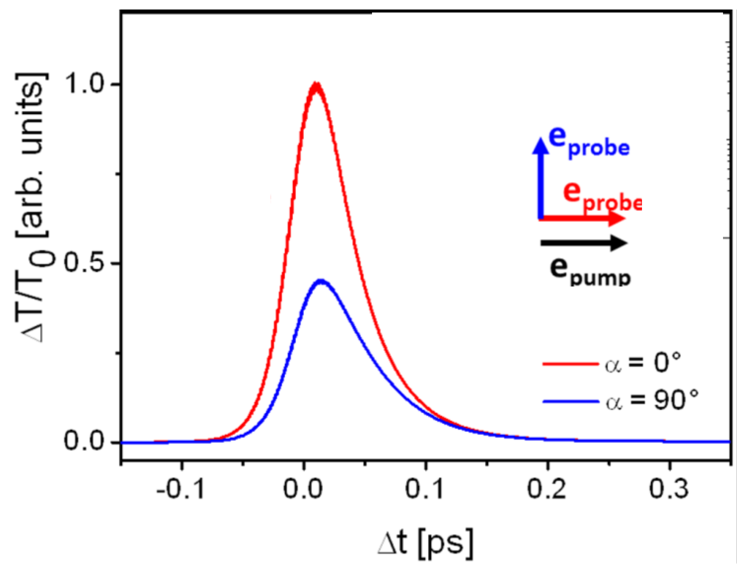
- **Polarization-dependent** high-resolution **pump-probe** experiment (**Stephan Winnerl, Manfred Helm,** Helmholtz-Zentrum Dresden)

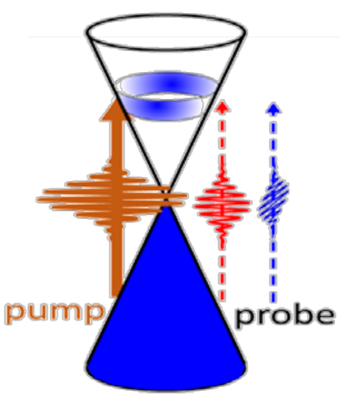
Experiment-theory comparison

Experiment



Theory





- **Polarization-dependent** high-resolution **pump-probe** experiment (**Stephan Winnerl, Manfred Helm,** Helmholtz-Zentrum Dresden)
- Theoretical prediction is in **excellent agreement** with experiment:

→ **Anisotropic differential transmission** can be observed within the **first 100 fs**

Nano Lett. 14, 1504 (2014)

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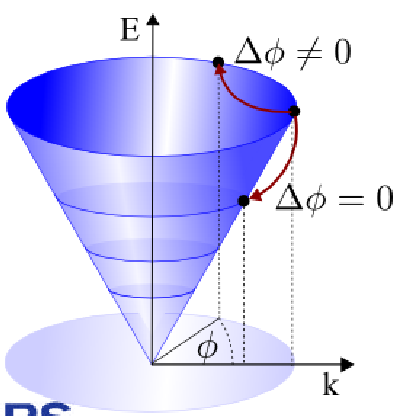
Phonons account for isotropy

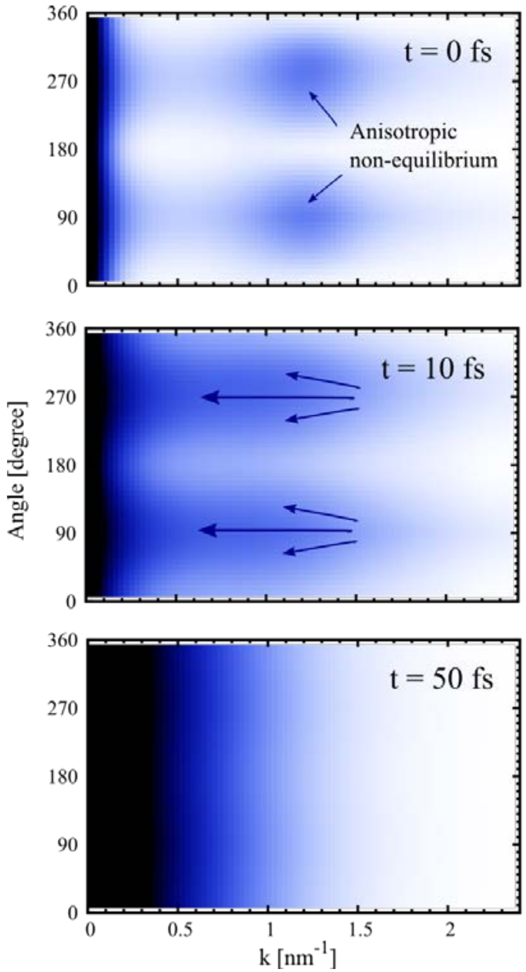
- **Carrier- phonon** coupling is **efficient** for scattering **across** the Dirac cone $\Delta\phi \neq 0$

  → **isotropic** distribution

- Carrier-carrier and carrier-phonon **channels in competition** for scattering **along** the Dirac cone with $\Delta\phi = 0$

  → **thermalization**





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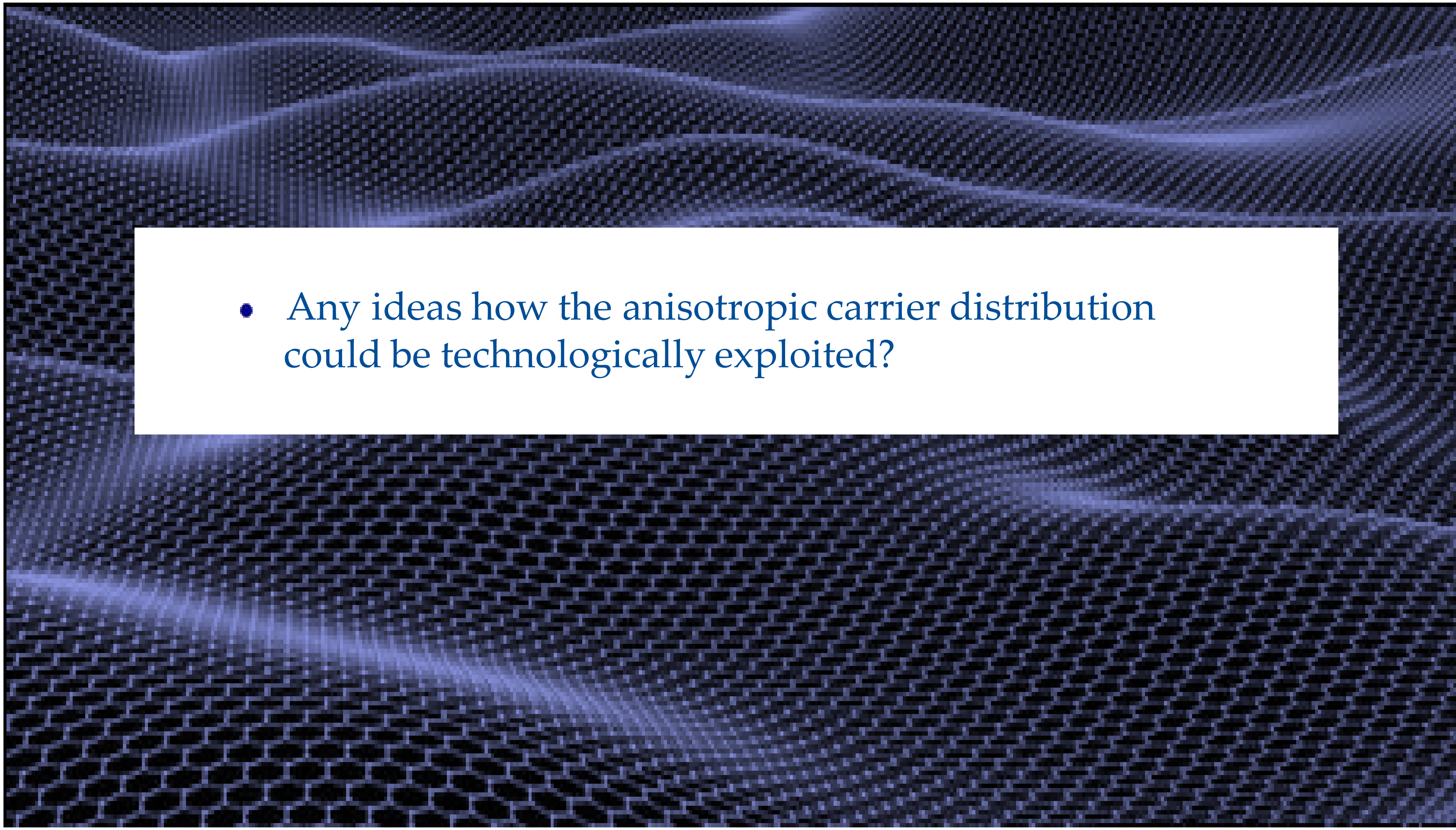
Think-pair-share: Anisotropy



- Any ideas how the anisotropic carrier distribution could be technologically exploited?

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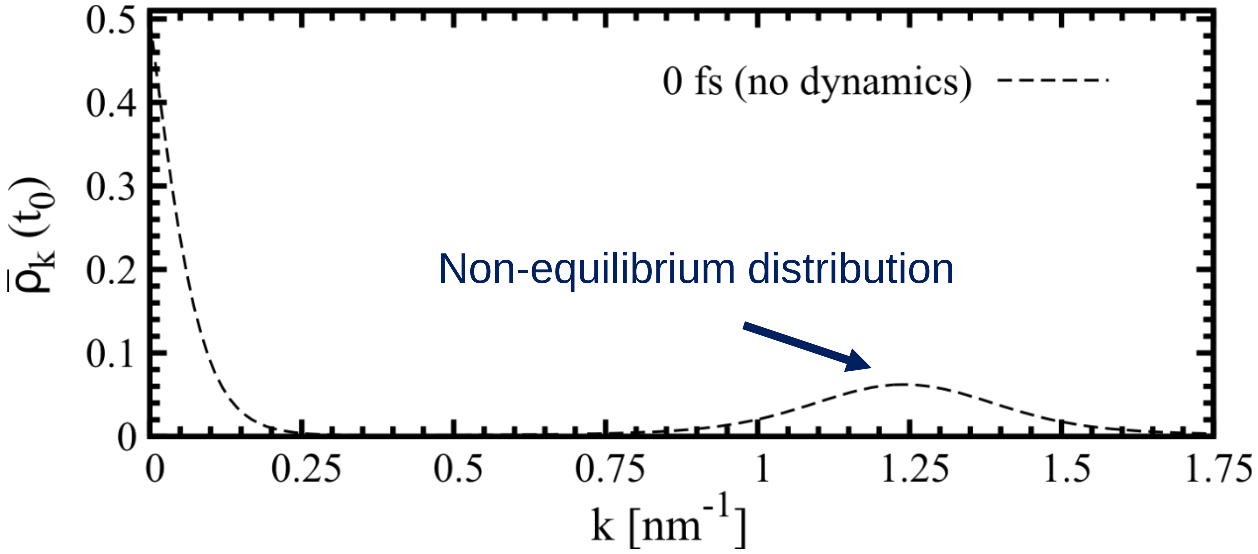
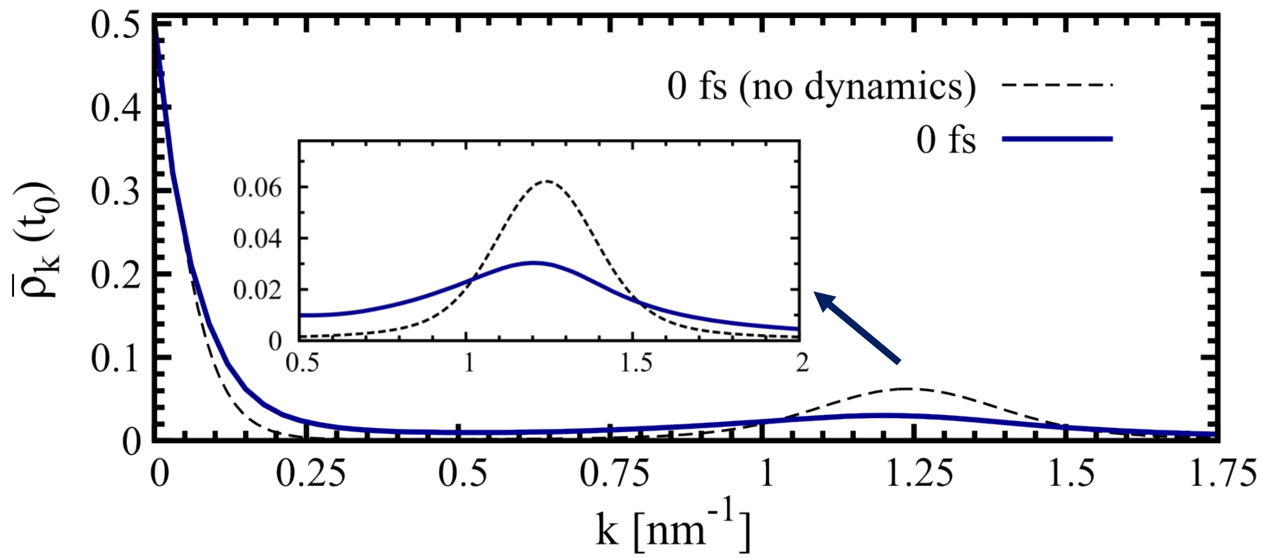
Carrier thermalization

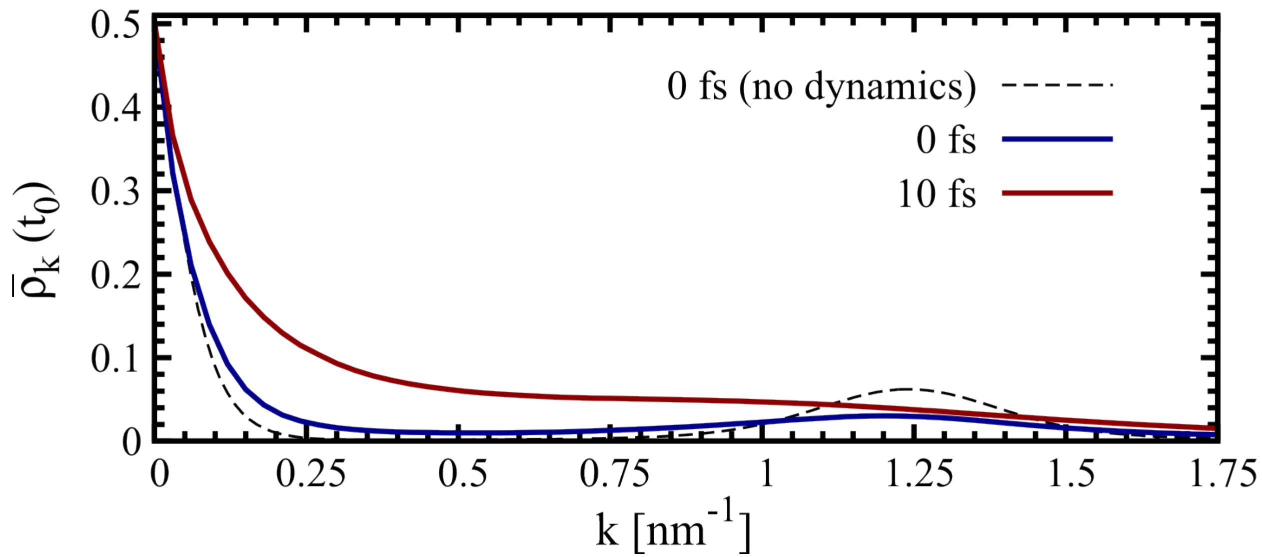


Carrier thermalization



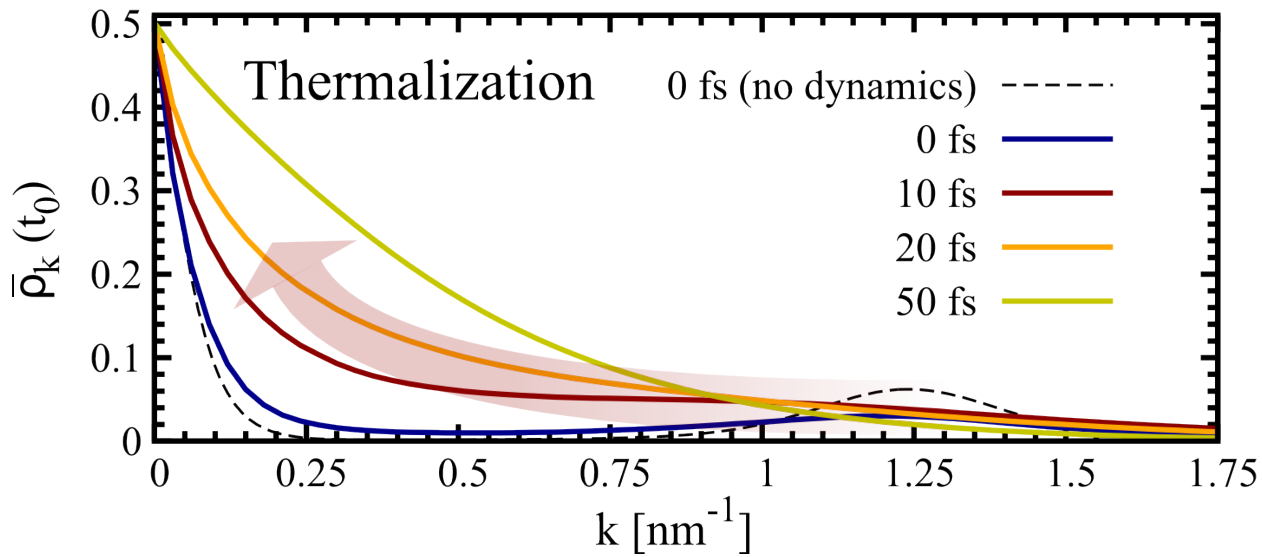
- Significant **relaxation** takes place already **during** the excitation **pulse**

Carrier thermalization



- Significant **relaxation** takes place already **during** the excitation **pulse**
- Carrier-carrier and carrier-phonon scattering are in direct **competition**

Carrier thermalization



- Significant **relaxation** takes place already **during** the excitation **pulse**
- Carrier-carrier and carrier-phonon scattering are in direct **competition**
- **Thermalized distribution** reached within the first **50-100 fs**

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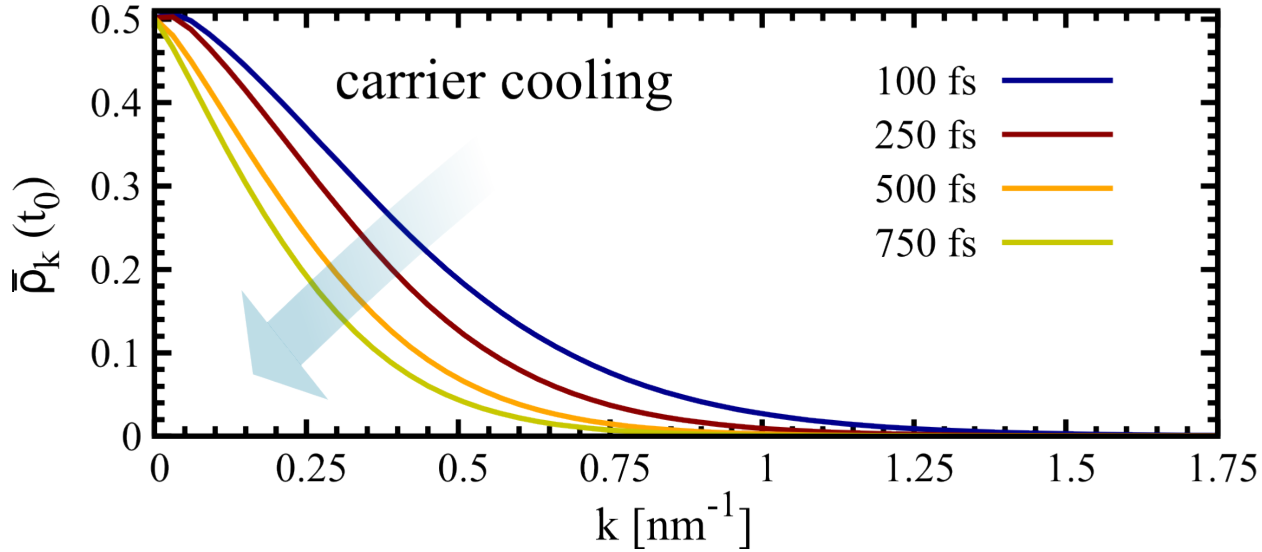
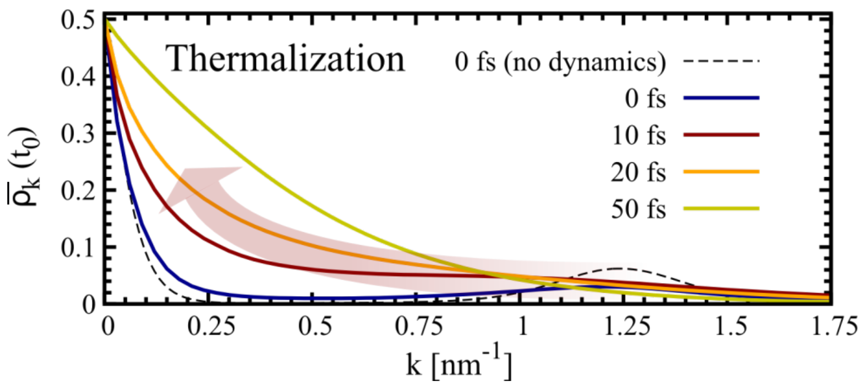
Carrier cooling

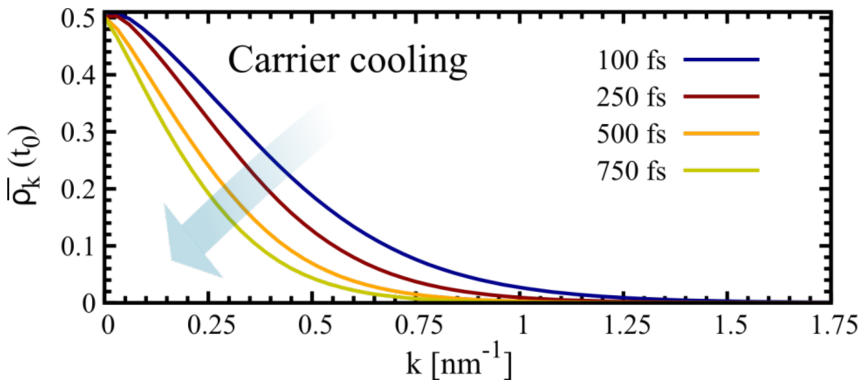


- **Carrier cooling** takes place on a **picosecond time scale**
- **Optical phonons** (in particular ΓLO, ΓTO and *K* phonons) are more **efficient** than acoustic phonons

PRB 84, 205406 (2011)

Carrier cooling

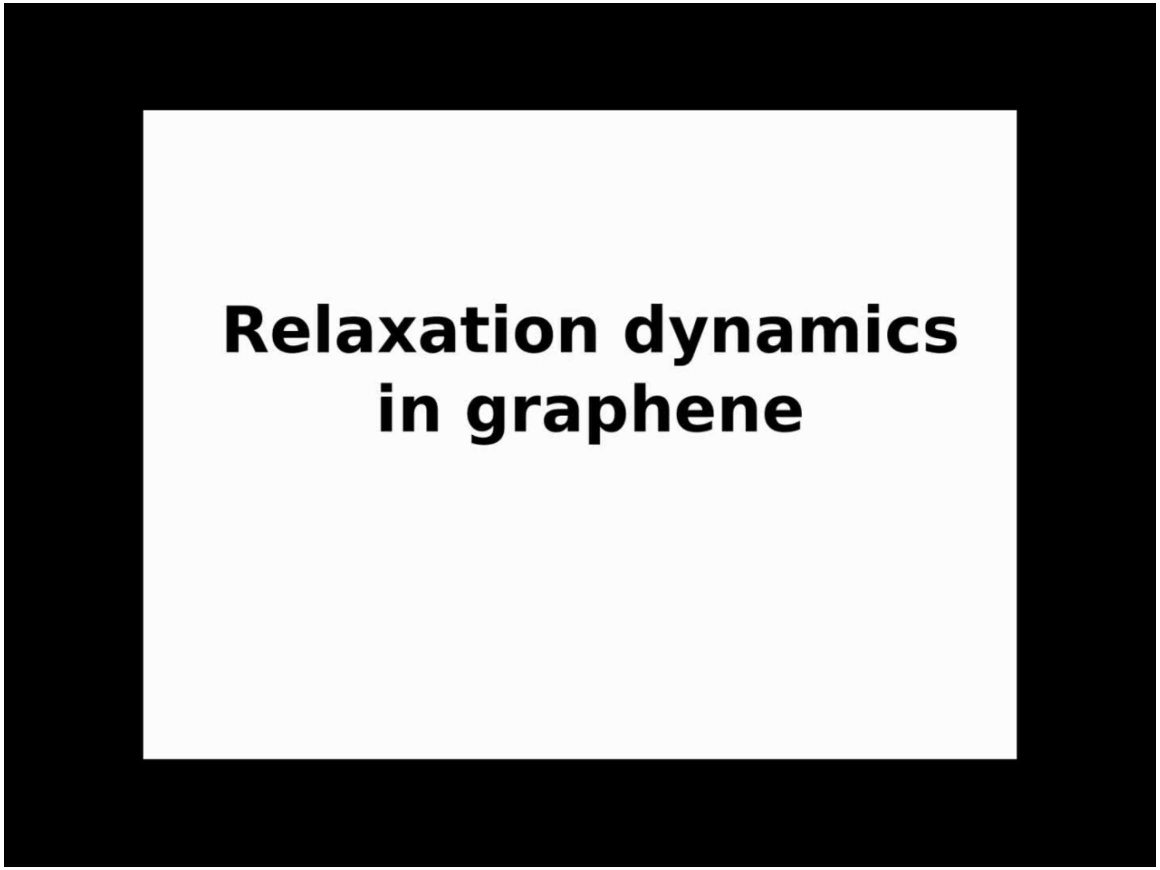




Carrier dynamics is characterized by **two processes**:

- ➞ Carrier-carrier and carrier-phonon scattering leads to **thermalization** on **fs time scale**
- ➞ Phonon-induced **carrier cooling** occurs on **ps time scale**

Relaxation dynamics in graphene



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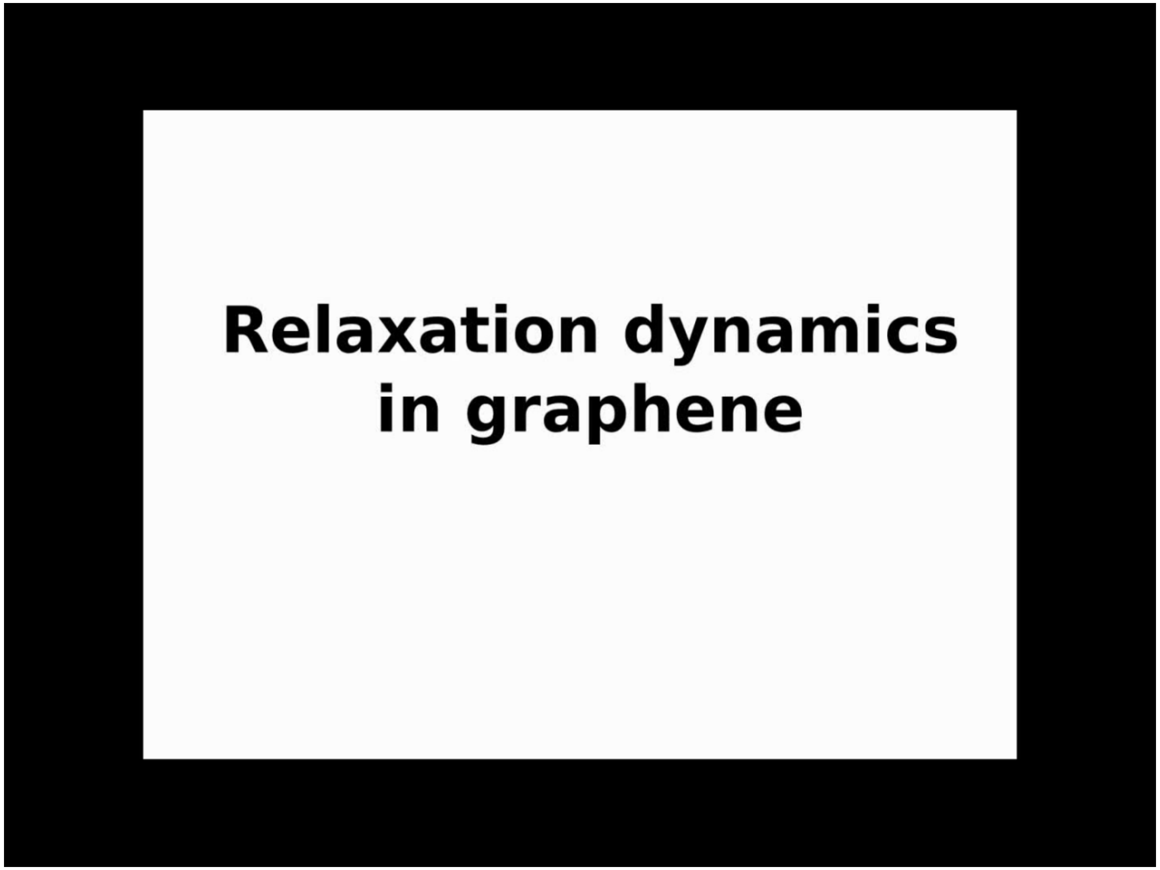
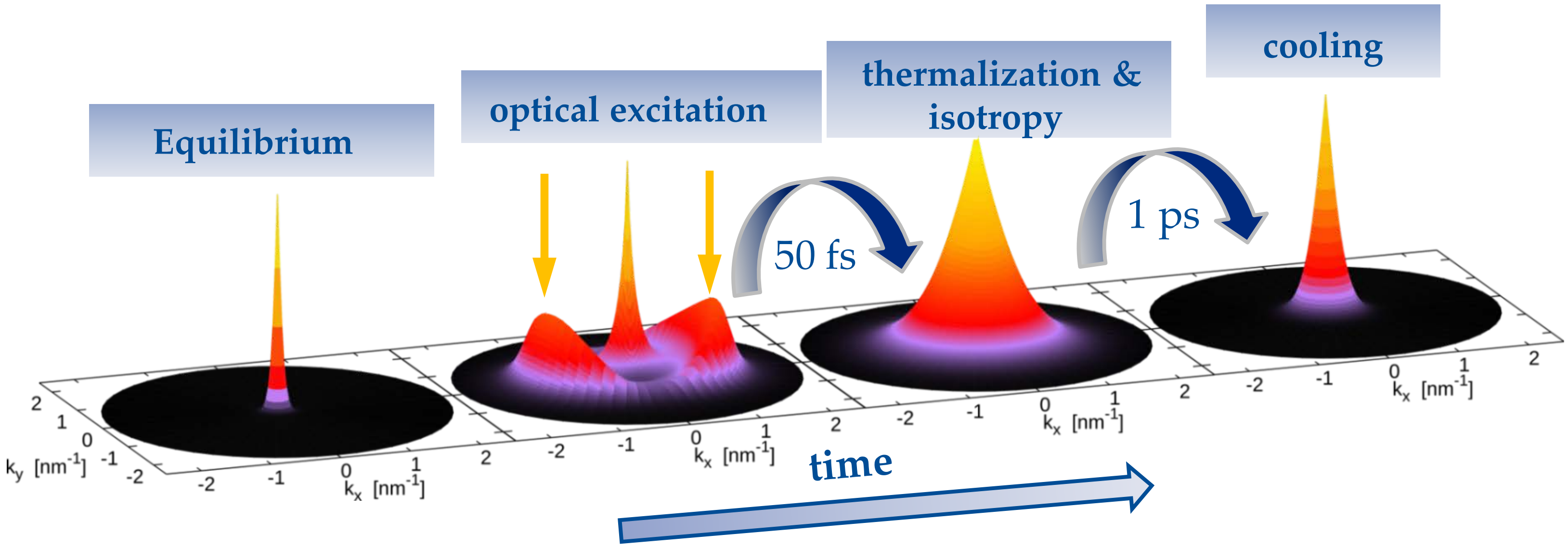
Relaxation dynamics in graphene



Steps during relaxation dynamics



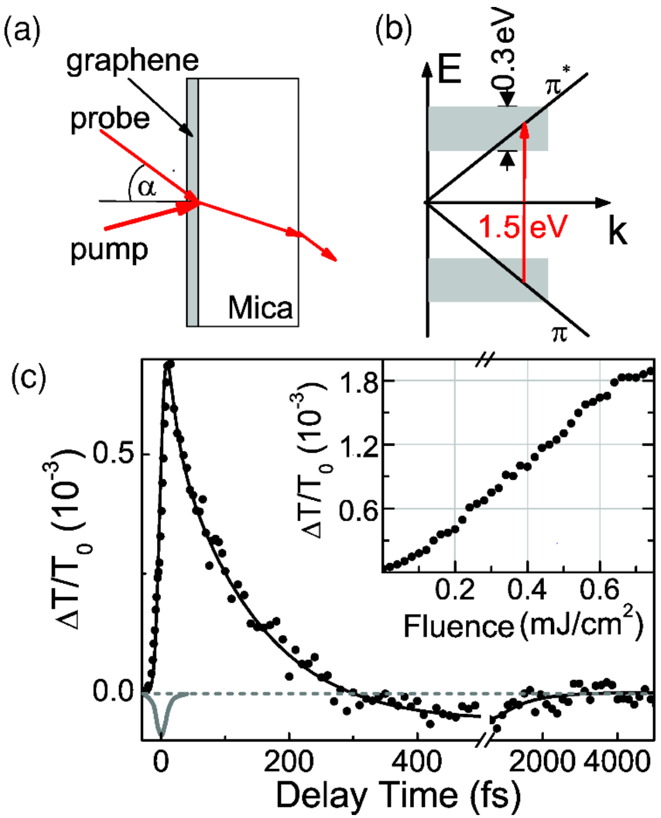
- Optically generated strongly **anisotropic non-equilibrium** carrier distribution
- **Carrier-phonon** scattering accounts for **isotropy,** while **carrier-carrier** scattering leads to a spectrally broad **thermalized** distribution within the first **50 fs**
- **Carrier-phonon** scattering gives rise to carrier **cooling** on **ps time scale**

PRB 84, 205406 (2011)

Experiment in the infrared regime

- Pump-probe-experiment measuring **differential transmission** in graphene
- **Excitation energy** in the infrared region at **1.5 eV**
- Temporal **resolution** is **10 fs**
- Initial increase of transmission is due to the **absorption bleaching**
- Following **decay** is characterized by **two time constants**:

$$\tau_1 = 140\ \text{fs}; \quad \tau_2 = 0.8\ \text{ps}$$



collaboration with **Thomas Elsaesser** (Max-Born Institut, Berlin)

PRB 83, 153410 (2011)

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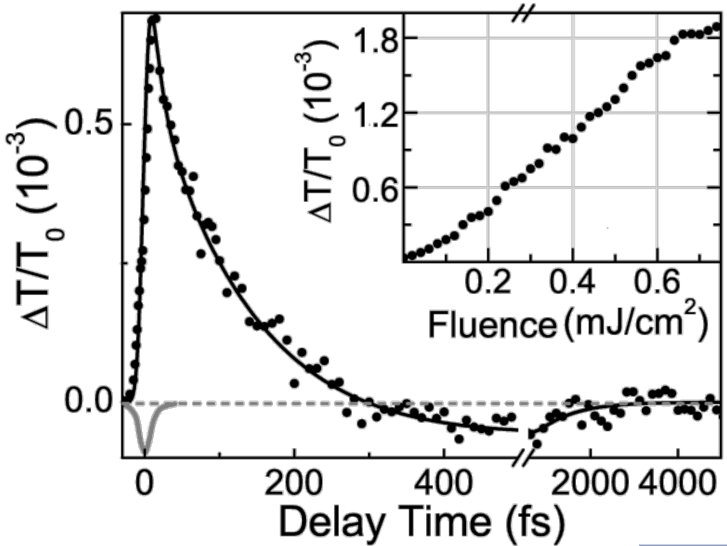
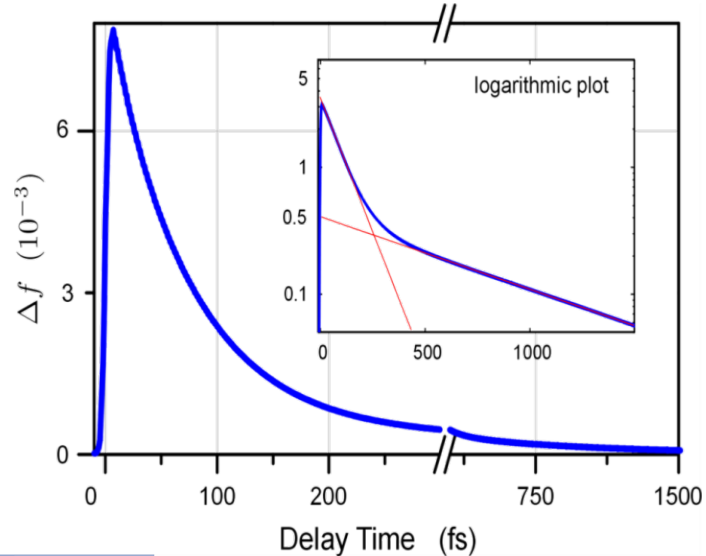
Experiment-theory comparison

Experiment



Theory



τ_1 = 140 fs, τ_2 = 0.8 ps

two decay times

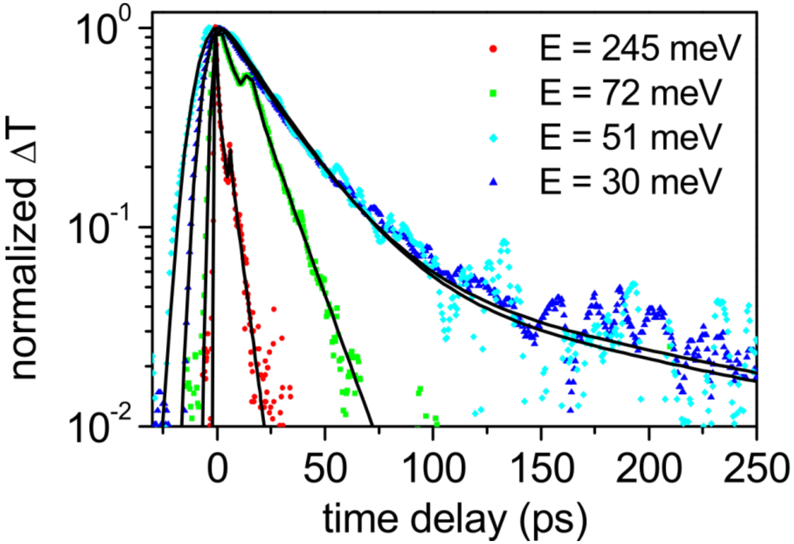
τ_1 = 104 fs, τ_2 = 0.7 ps

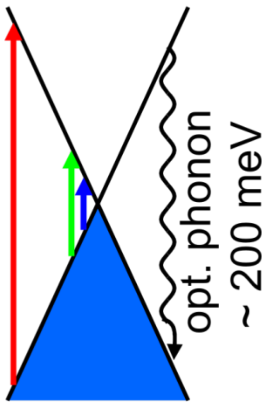
- Theory is in **good agreement** with experiment:

→ τ_1 corresponds to **thermalization**, τ_2 describes **carrier cooling**

PRB 83, 153410 (2011)

Experiment close to the Dirac point





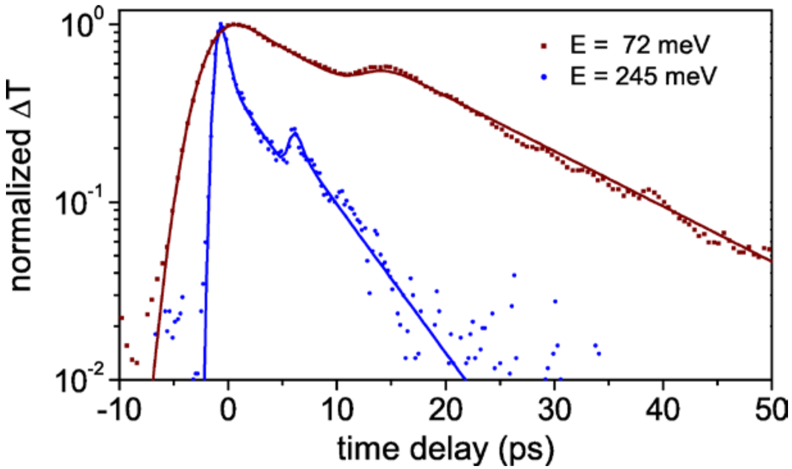
collaboration with **Manfred Helm** (Helmholtz-Zentrum Dresden-Rossendorf)

- Transmission in the vicinity of Dirac point and **below** the **energy** of **optical phonons** (~ 200 meV) ➞ acoustic phonons dominant?
- Relaxation **dynamics** is **slowed down** (5 ps at 245 meV, **25 ps** at 30 meV)

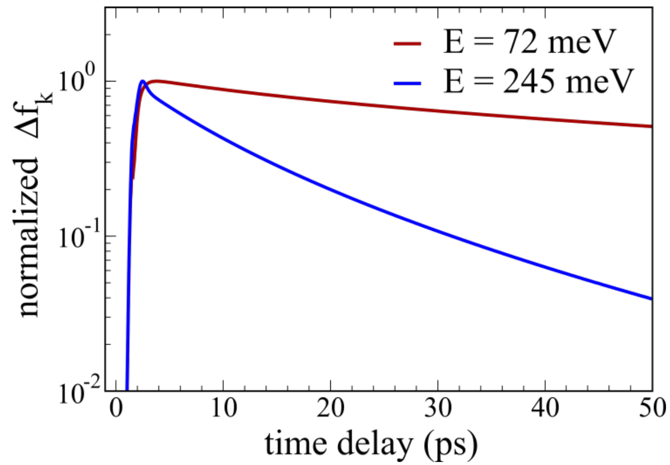
PRL 107, 237401 (2011)

Experiment-theory comparison

Experiment



Theory



- Theory in **good agreement** with experiment (slowed-down dynamics):

→ **Optical phonons** remain the **dominant** relaxation channel, since carrier-carrier scattering leads to a **spectrally broad distribution**

PRL 107, 237401 (2011)

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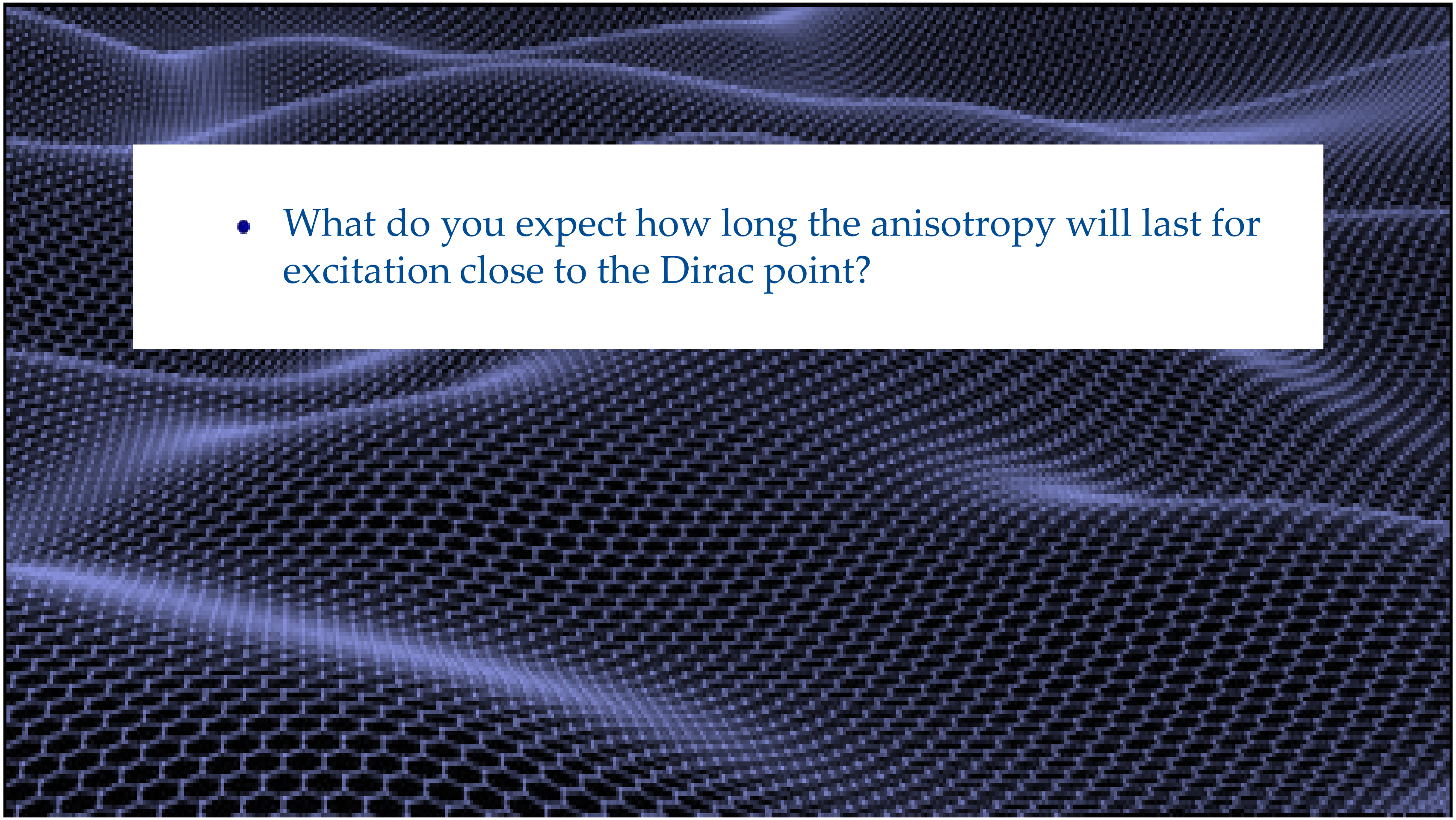
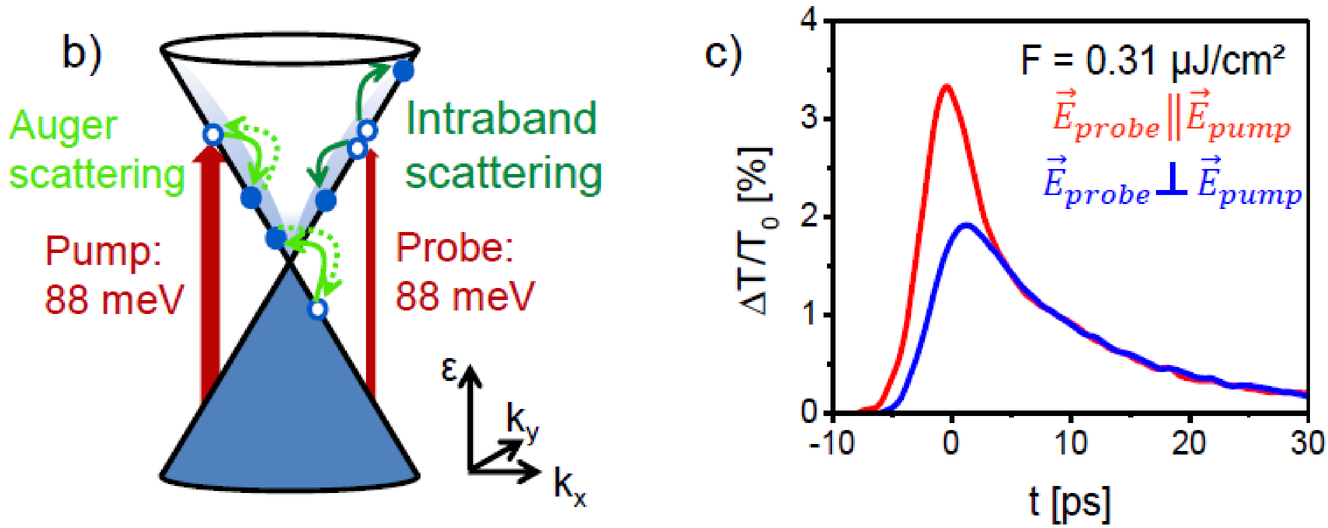
Think-pair-share: Anisotropy close to Dirac point



- What do you expect how long the anisotropy will last for excitation close to the Dirac point?

Anisotropy close to the Dirac point

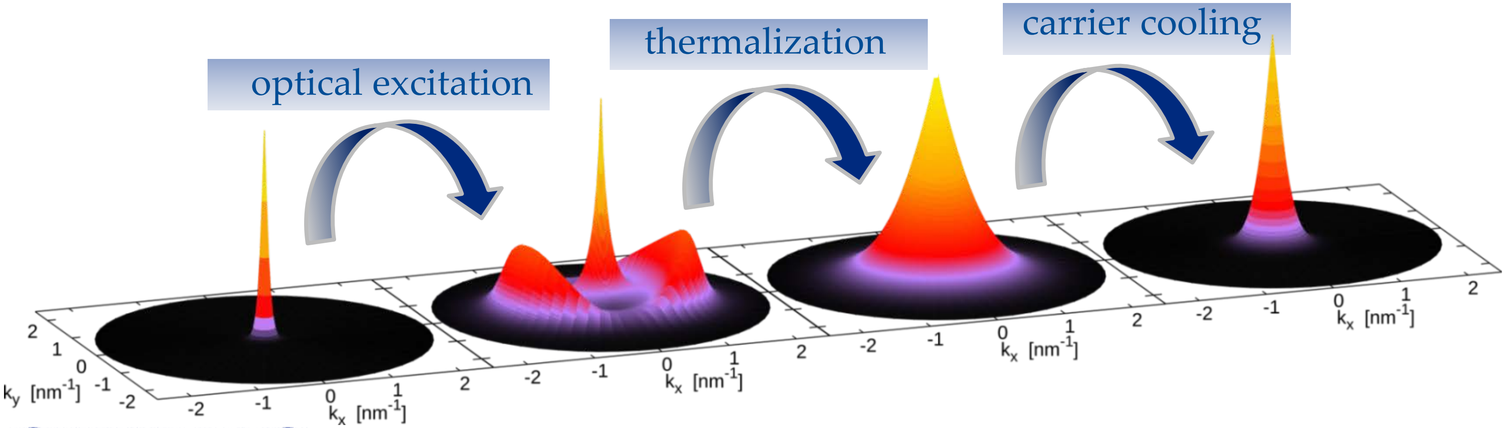


- Optical excitation at energies lower than the optical phonon energy of 200meV strongly **suppress carrier-phonon scattering**

→ **Isotropic carrier** distribution is reached via **carrier-carrier scattering** on a much smaller **ps time scale**

Outline

- Motivation
- Microscopic modelling
- Carrier dynamics
- **Many-particle phenomena**



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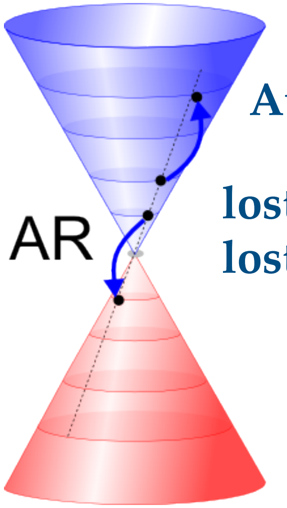
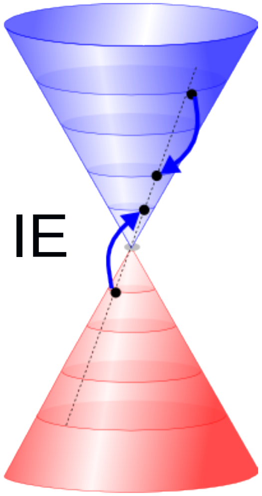
Auger scattering

- Auger scattering **changes** the number of charge **carriers in the system**



Auger recombination (AR)

lost electron in cond.band
lost hole in valence band

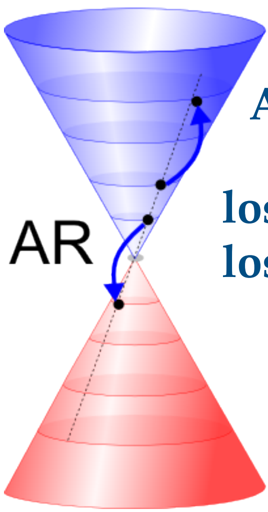


Inverse Auger recombination or **impact excitation (IE)**

gained electron in cond. band
gained hole in valence band

Impact excitation

- Auger scattering **changes** the number of charge **carriers in the system**



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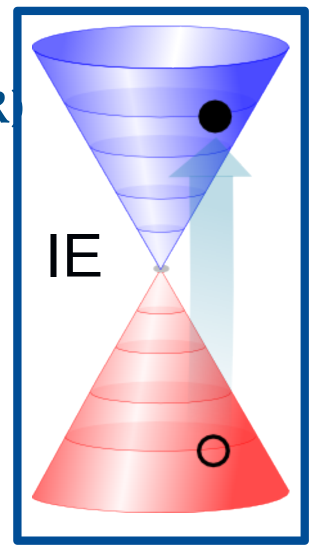

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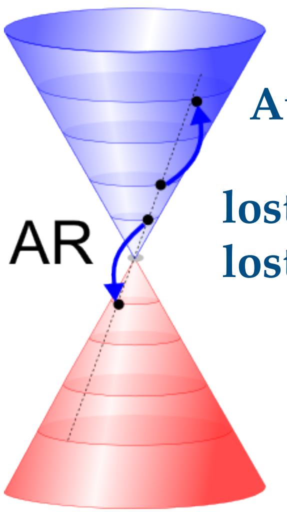

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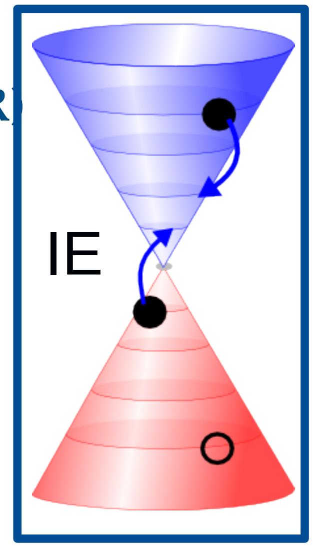

Inverse Auger recombination or **impact excitation (IE)**

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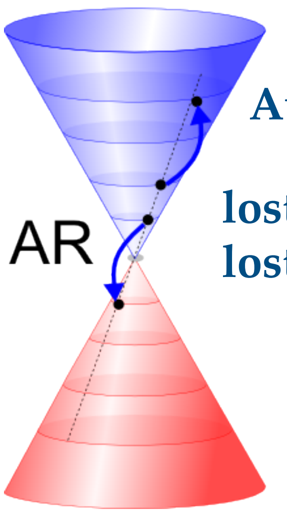
Carrier multiplication

- Auger scattering **changes** the number of charge **carriers in the system**



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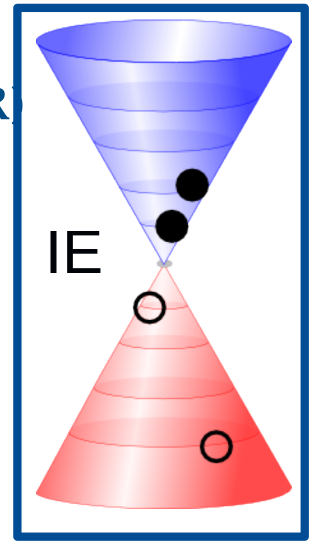


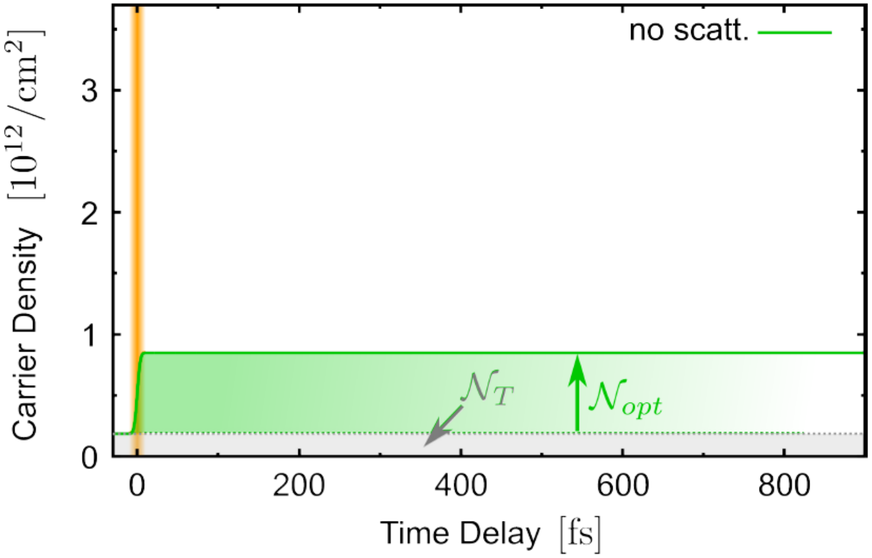
Inverse Auger recombination or **impact excitation (IE)**

gained electron in cond. band
gained hole in valence band

→ **carrier multiplication**

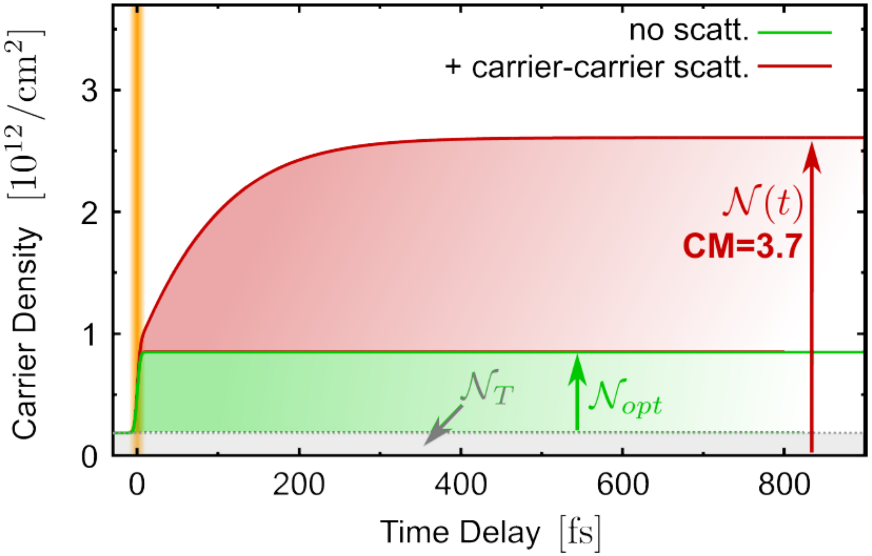
- In **conventional semiconductors** (band gap, parabolic band structure) Auger scattering is **inefficient** due to energy and momentum conservation

Carrier density



- **Carrier density increases** during the excitation pulse

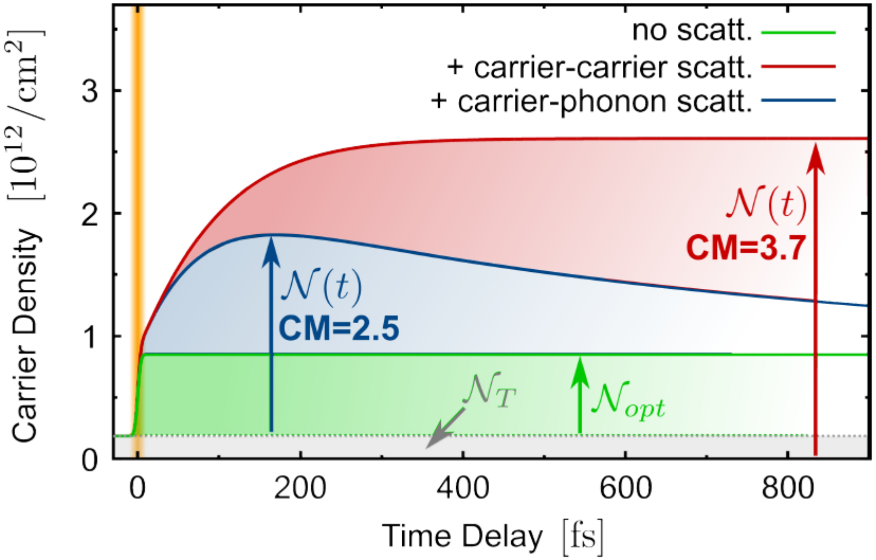
Carrier multiplication



- **Carrier density increases** during the excitation pulse
- Auger scattering leads to **carrier multiplication (CM)**

$$CM = \frac{\mathcal{N}(t) - \mathcal{N}_T}{\mathcal{N}_{opt}}$$

Carrier multiplication



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- **Carrier-phonon** scattering **reduces CM** on a picosecond time scale

Nano Lett. 10, 4839 (2010)

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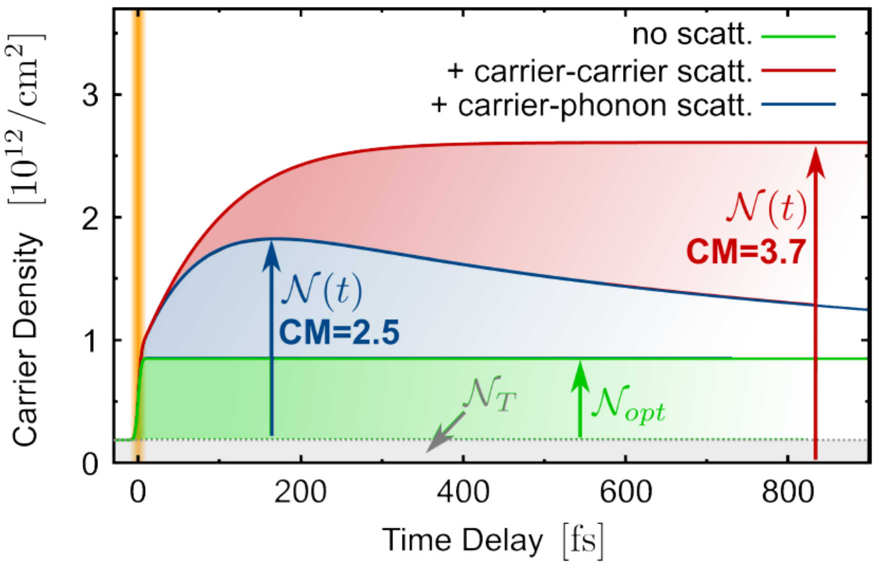
Microscopic mechanism



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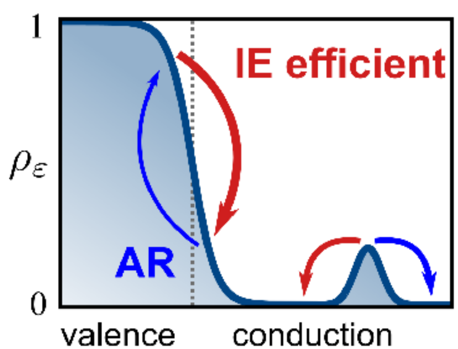
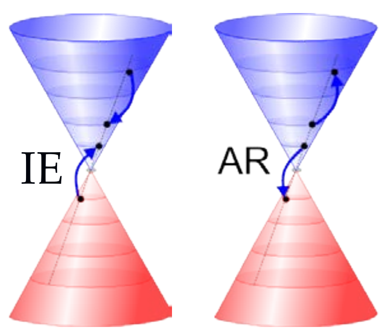
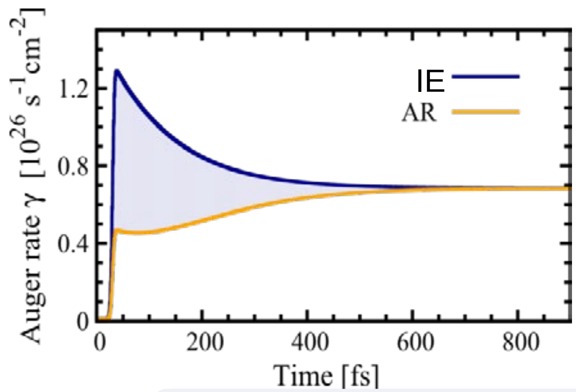
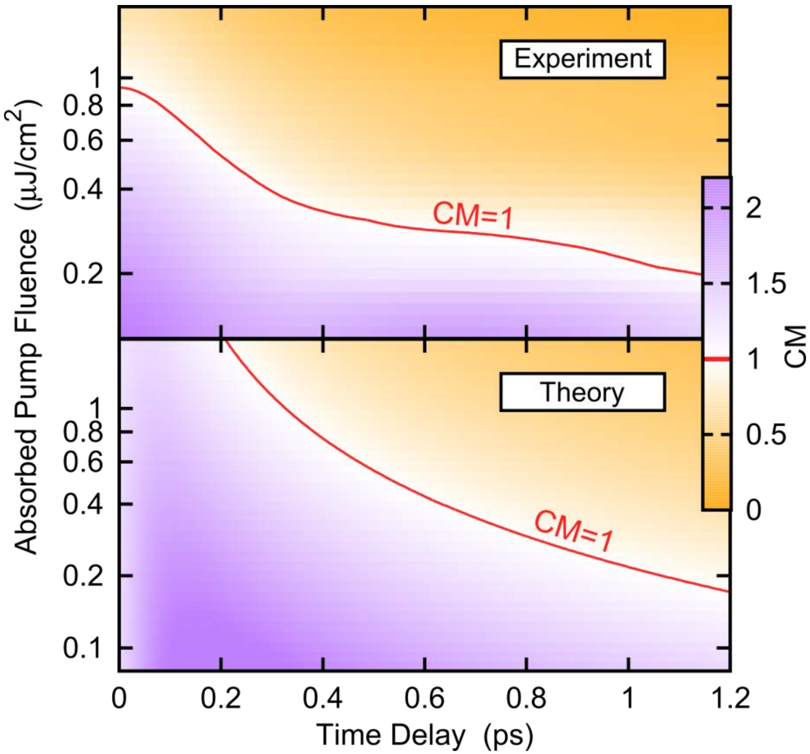






Nano Lett. 10, 4839 (2010)

Experiment-theory comparison



- High-resolution **multi-color pump-probe** spectroscopy (**Daniel Neumaier** and **Heinrich Kurz,** RWTH Aachen)

  → monitor temporal evolution of the carrier density

- Theoretical prediction is in **excellent agreement** with experiment:

  → Appearance of **CM** with **distinct fluence dependence**

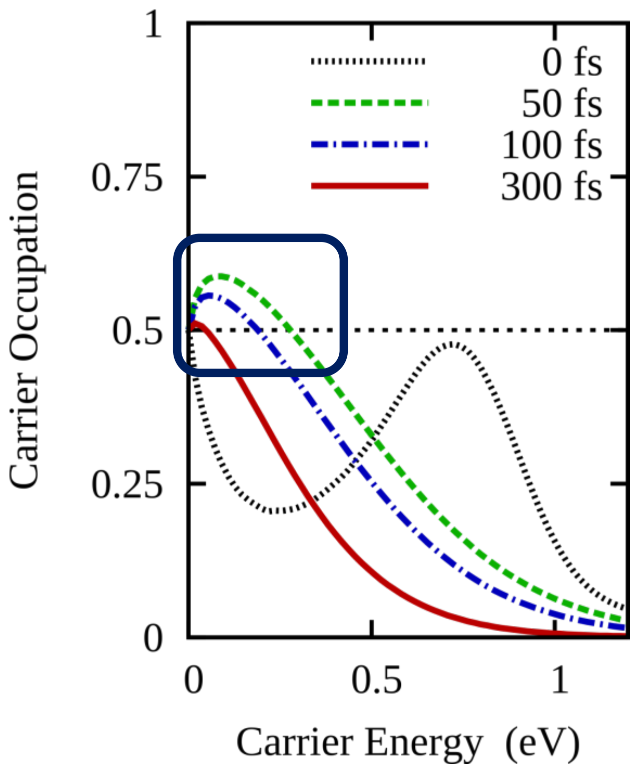
Nano Lett. 14, 5371 (2015)

Think-pair-share: Carrier multiplication

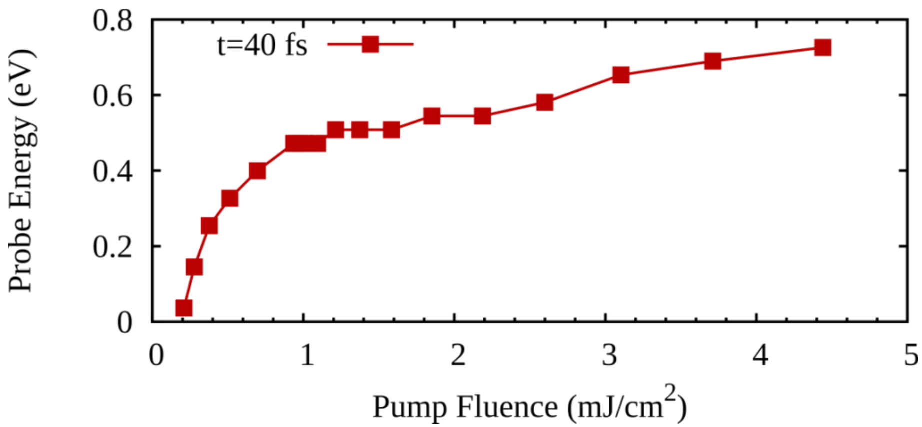
- In which technological devices could carrier multiplication be of advantage?

- Would you expect the CM to be more efficient at low excitation energies, low temperatures and low doping?

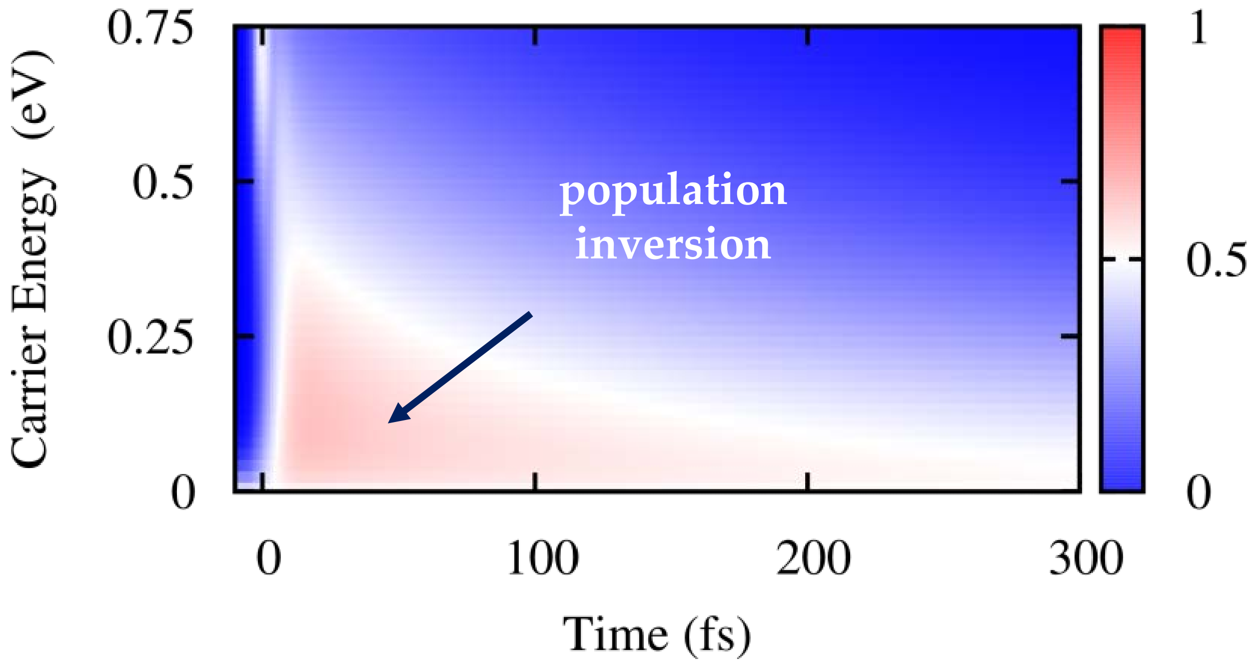
II. Population inversion



- **Population inversion** occurs in the **high-excitation** regime (>0.2 $mJcm^{-2}$)
- **Spectrally** and **temporally limited** depending on pump fluence

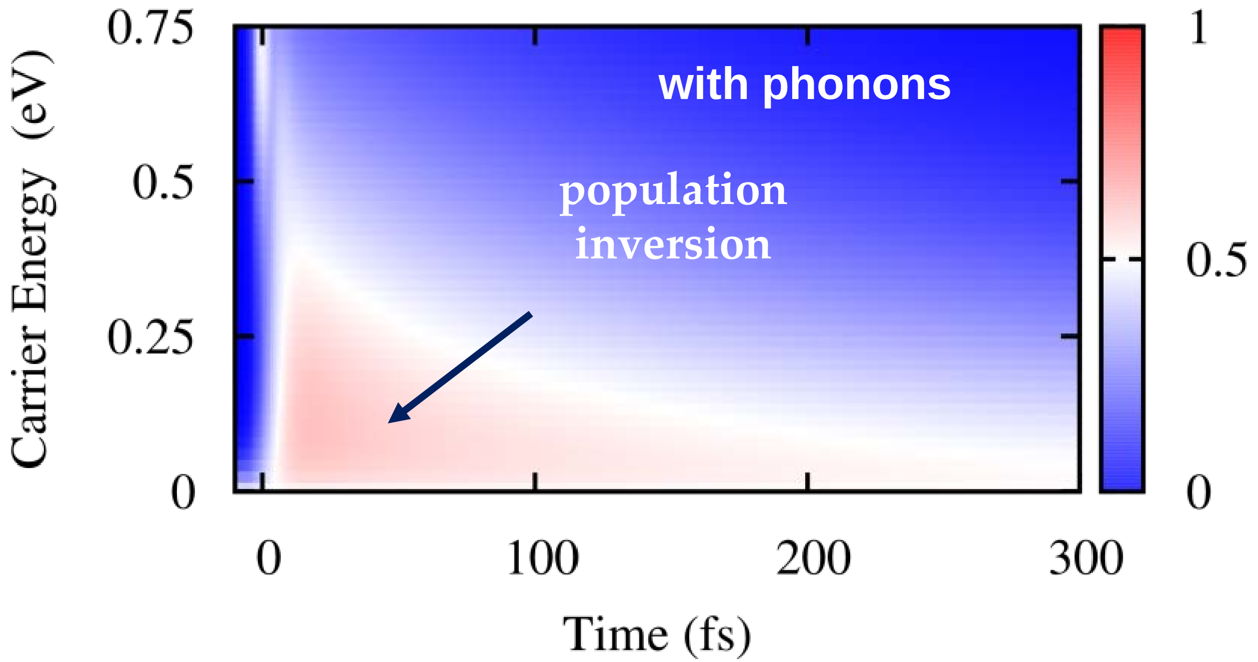


Build-up and decay



- Population inversion is **built up** within the **first 10 fs** during optical excitation
- The generated population inversion **decays** on a time scale of **few 100 fs**

Build-up and decay



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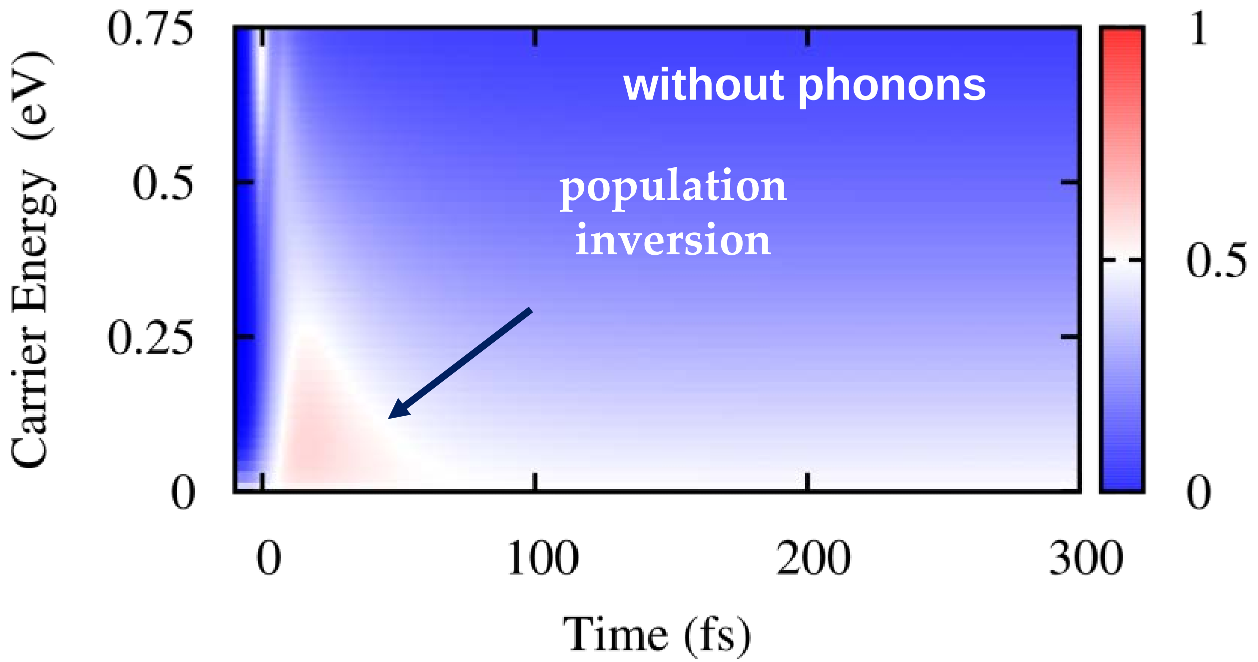


- Population inversion is **built up** within the **first 10 fs** during optical excitation
- The generated population inversion **decays** on a time scale of **few 100 fs**
- **Intraband** scattering with **phonons** plays a crucial role: the gain region is strongly reduced without phonons

PRB 87, 165413 (2013)

Microscopic mechanism

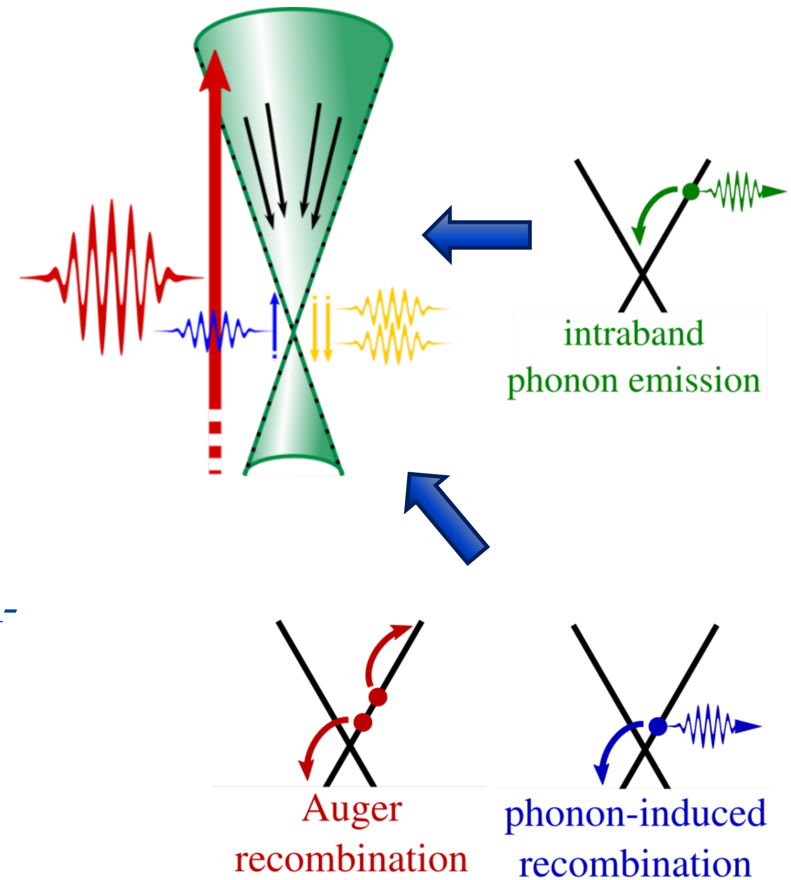
- Excited carriers scatter down via **phonon-induced intraband** processes

- **Vanishing density of states** at the Dirac point gives rise to a **relaxation bottleneck**

→ **build-up of population inversion**

- Efficient **Auger recombination** and phonon-induced interband scattering reduce the carrier accumulation

→ **decay of population inversion**



PRB 87, 165413 (2013)

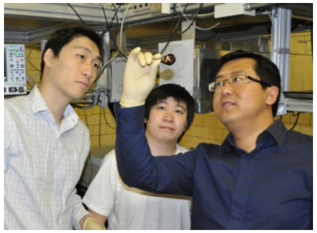
Population inversion in experiment

Selected for a Viewpoint in *Physics*

PRL **108,** 167401 (2012) PHYSICAL REVIEW LETTERS week ending 20 APRIL 2012

Femtosecond Population Inversion and Stimulated Emission of Dense Dirac Fermions in Graphene

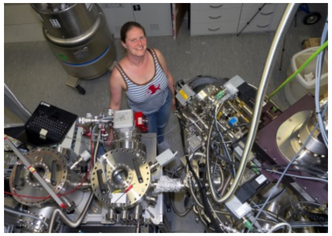
T. Li,[1] L. Luo,[1] M. Hupalo,[1] J. Zhang,[1,2] M. C. Tringides,[1] J. Schmalian,[1,3] and J. Wang[1]



- In **strongly excited** graphene, quasi-instantaneous build-up of **broadband population inversion** is found manifesting itself in **negative conductivity**

Nature Materials **12**, 1119–1124 (2013) | doi:10.1038/nmat3757

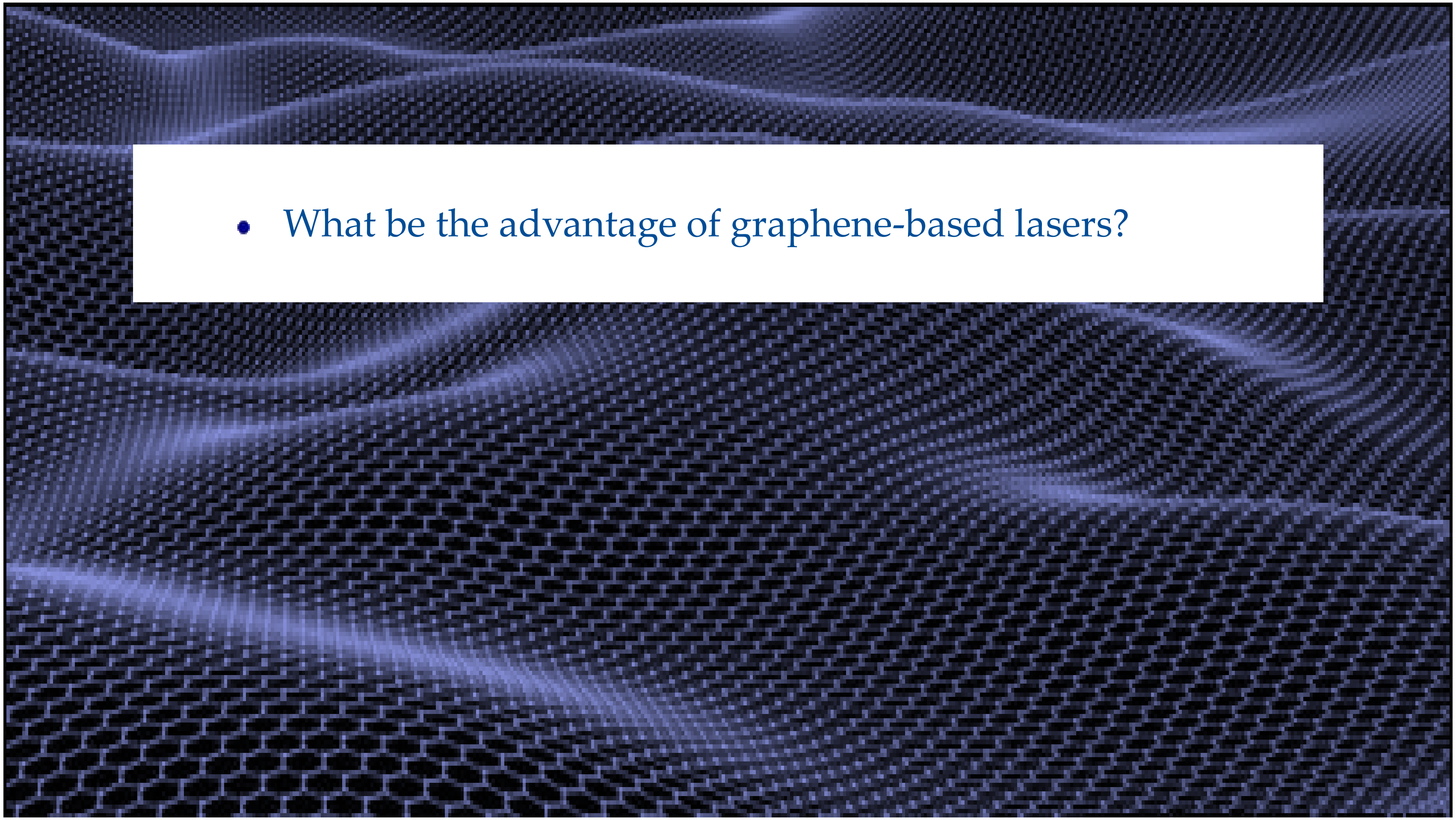
Snapshots of non-equilibrium Dirac carrier distributions in graphene



Isabella Gierz, Jesse C. Petersen, Matteo Mitrano, Cephise Cacho, I. C. Edmond Turcu, Emma Springate, Alexander Stöhr, Axel Köhler, Ulrich Starke & Andrea Cavalleri

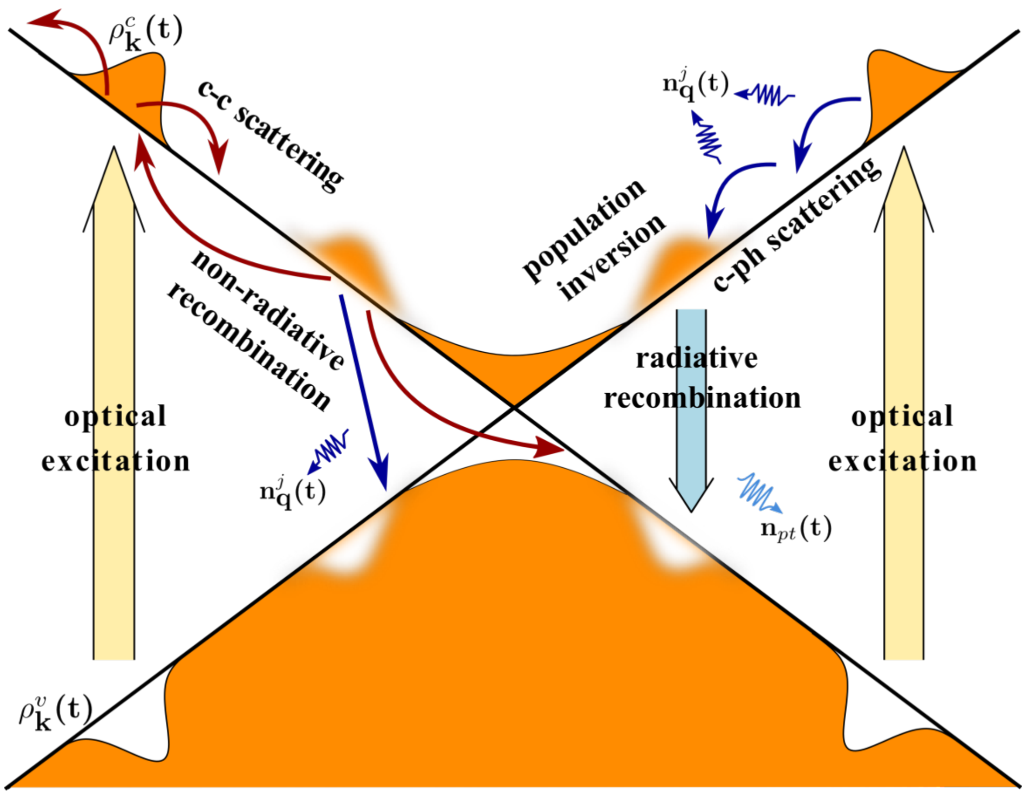
- The appearance of **a transient population inversion** is directly confirmed in a **time-resolved ARPES** experiment

Think-pair-share: Population inversion



- What be the advantage of graphene-based lasers?

Graphene-based lasers



- To achieve **long-lived gain** and **coherent laser light emission**, non-radiative recombination channels need to be suppressed and radiative coupling enhanced

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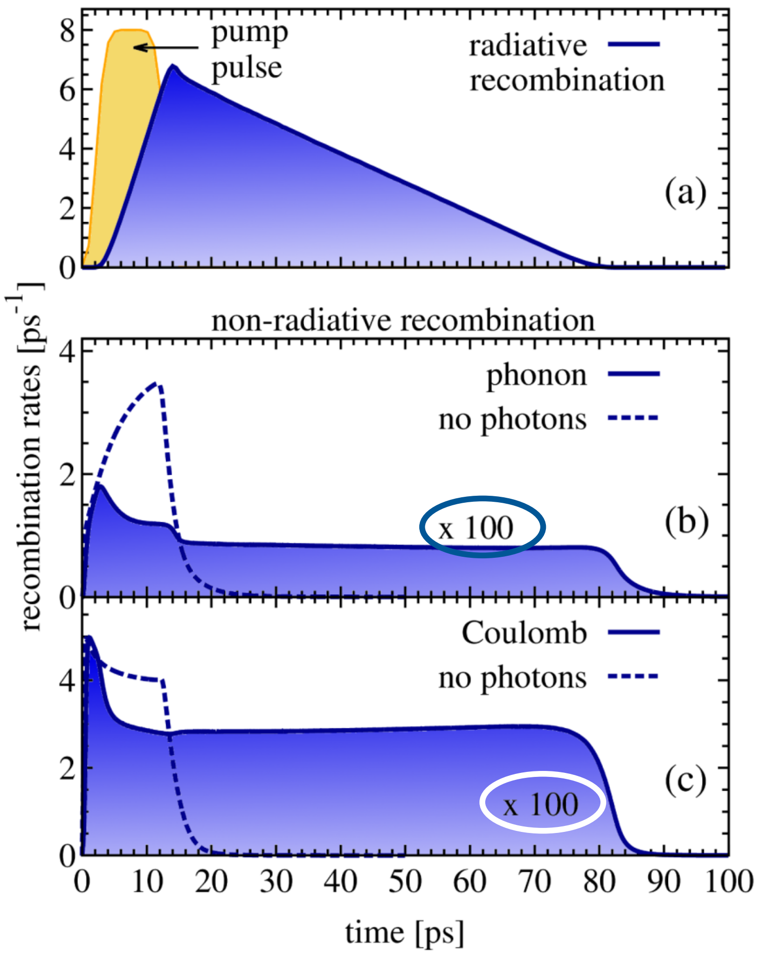
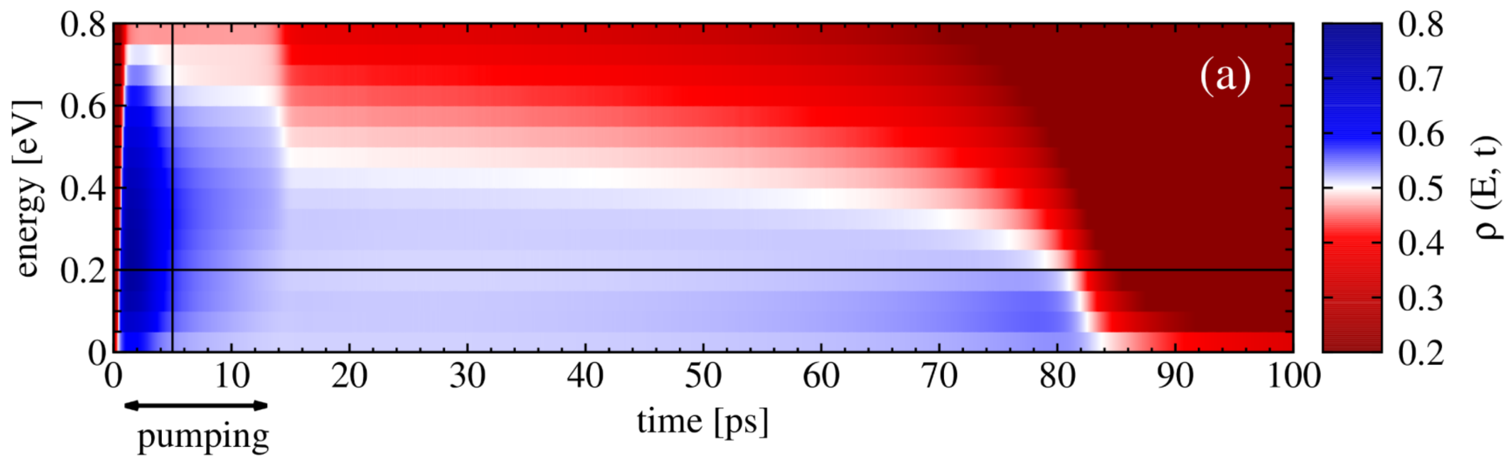
Radiative vs. non-radiative recombination

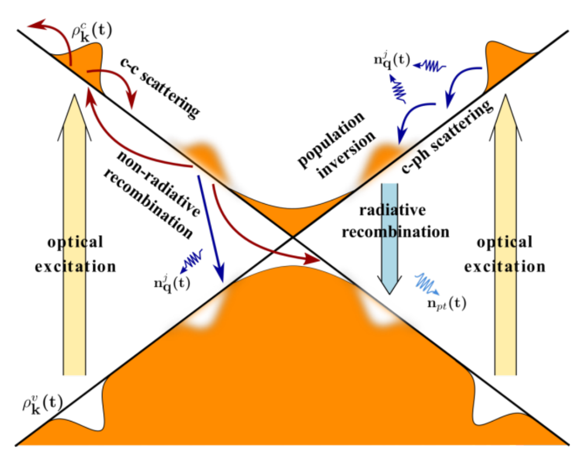


- Implementing graphene into a **cavity**, carrier-light coupling is enhanced giving rise to **strong radiative recombination**
- Including a **high-dielectric substrate, non-radiative recombination** channels are strongly **suppressed**
- **Radiative recombination** of excited carriers **prevails** over non-radiative channels

Dyamics of electrons



- **Long-lived gain** is achieved on a time scale of **100 ps** depending on the excitation strenght and duration



- **Quasi-equilibrium** is reached between radiative and non-radiative **recombination processes** and **intraband scattering** refilling the depleted states

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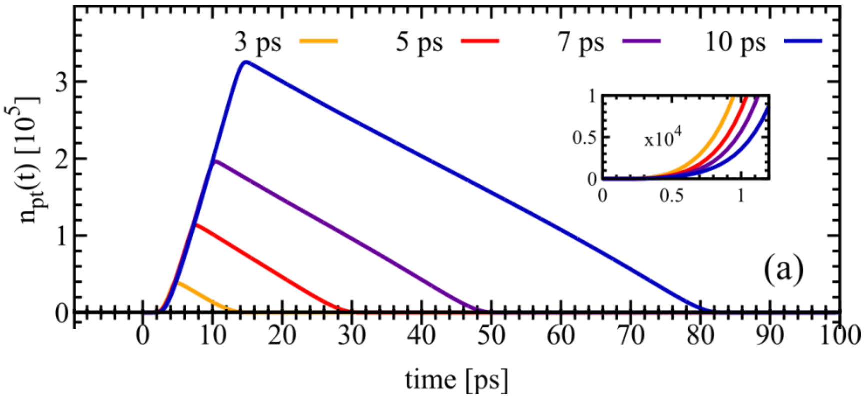
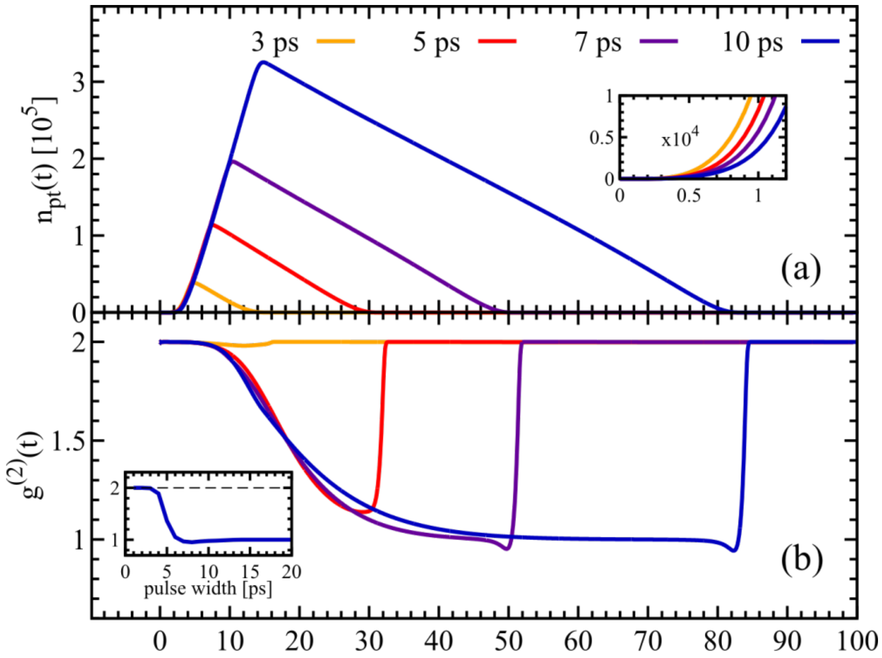
Dynamics of photons

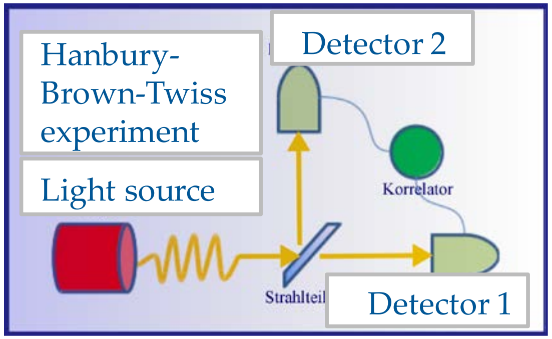


Photon dynamics

- Initially, only **spontaneous emission** contributes to dynamics of photons
- Exponential increase is due to the processes of **induced emission**

Emission of coherent laser light





Photon dynamics

- Initially, only **spontaneous emission** contributes to dynamics of photons
- Exponential increase is due to the processes of **induced emission**

Photon statistics

- **Laser threshold** is surpassed for excitations longer than 5ps resulting in emission of **coherent laser light**
- Second-order autocorrelation function:

thermal light: $g^{(2)} = 2$

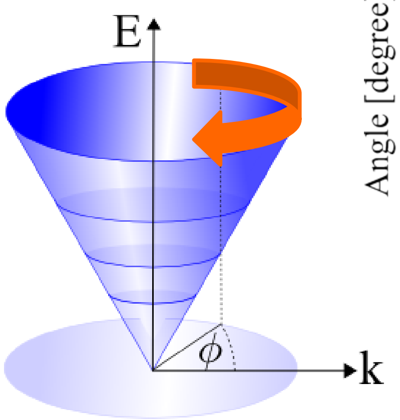
laser light: $g^{(2)} = 1$

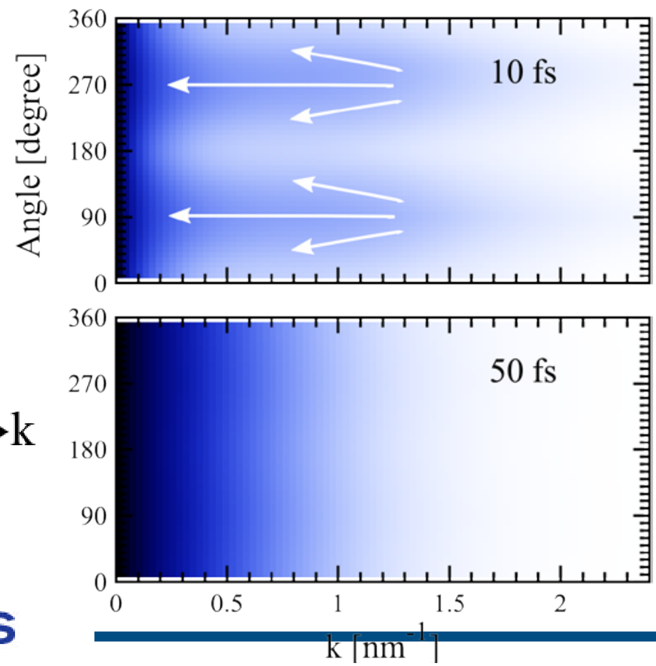
PRB 92, 085407 (2015)

Conclusions

Density matrix theory offers microscopic access to time-, momentum-, and angle-resolved relaxation dynamics:

- **Thermalized, isotrope distribution** is reached already **after 50 fs** – followed by phonon-induced **carrier cooling** on a **ps time scale**

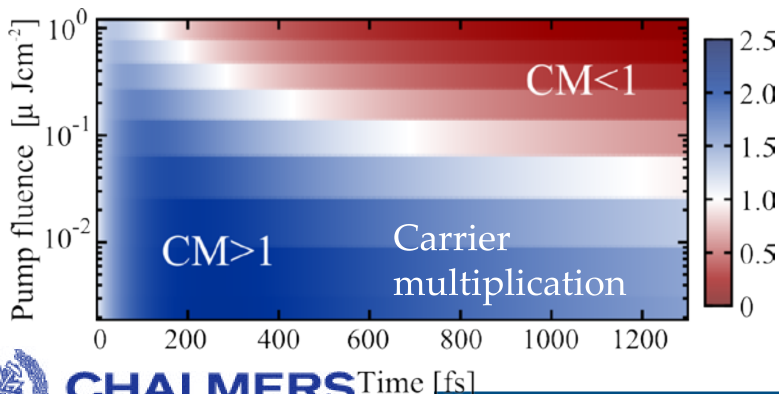


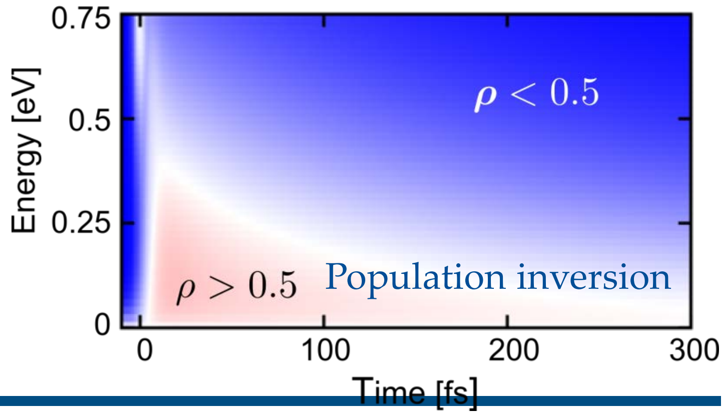


Conclusions

Density matrix theory offers microscopic access to time-, momentum-, and angle-resolved relaxation dynamics:

- **Thermalized, isotrope distribution** is reached already **after 50 fs** – followed by phonon-induced **carrier cooling** on a **ps time scale**
- Efficient **Auger scattering** (impact ionization) gives rise to a significant **carrier multiplication**
- Spectrally broad, **transient population inversion** can be obtained in **strong excitation regime**





Learning Outcomes

- Recognize the **potential of graphene** for fundamental science and technological applications
- Understand how optical and electronic properties of graphene can be **microscopically modelled** (tight-binding, second quantization, Bloch equations)
- Explain how ultrafast **carrier dynamics** in graphene works
- Realize the importance of **carrier multiplication** and its relevance for highly efficient photodetectors
- Demonstrate the importance of **population inversion** for highly tunable graphene-based lasers

Think-pair-share: What is graphene?

- What do you associate with the material graphene?

- What do you think makes graphene different from conventional materials, such as silicon?