Femtosecond time resolved pump-probe spectroscopic ellipsometry – some applications – and some words about long-lasting coherent phonon oscillations in GaP

Rüdiger Schmidt-Grund

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thanks to all those who were also strongly involved in development of TSE method and physical understanding of processes: (alphabetically)

J. Andreasson, C. Emminger, S. Espinoza, D. Franta, O. Herrfurth, K. Hingerl, A. Horn, M. Kloz, J. Leveillee, M. Olbrich, T. Pflug, M. Rebarz, S. Richter, E. Runge, A. Schleife, Y. Slimi, N. Stiehm, C. Sturm, S. Zollner, M. Zahradník

main cooperation:

UNIVERSITÄT LEIPZIG

Semiconductor Physics Group

Ellipsometry and Polariton Physics Group

eli | beamlines

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Why ultrafast dynamics?

(photo-induced) dynamics of the electronic (and phonon) system of materials for understanding and improving of:

- fundamental understanding
- application in ultra-fast devices
- lightning and optical data transport and processing
- opto-electro-magnetic coupling and switching
- solar energy harvesting

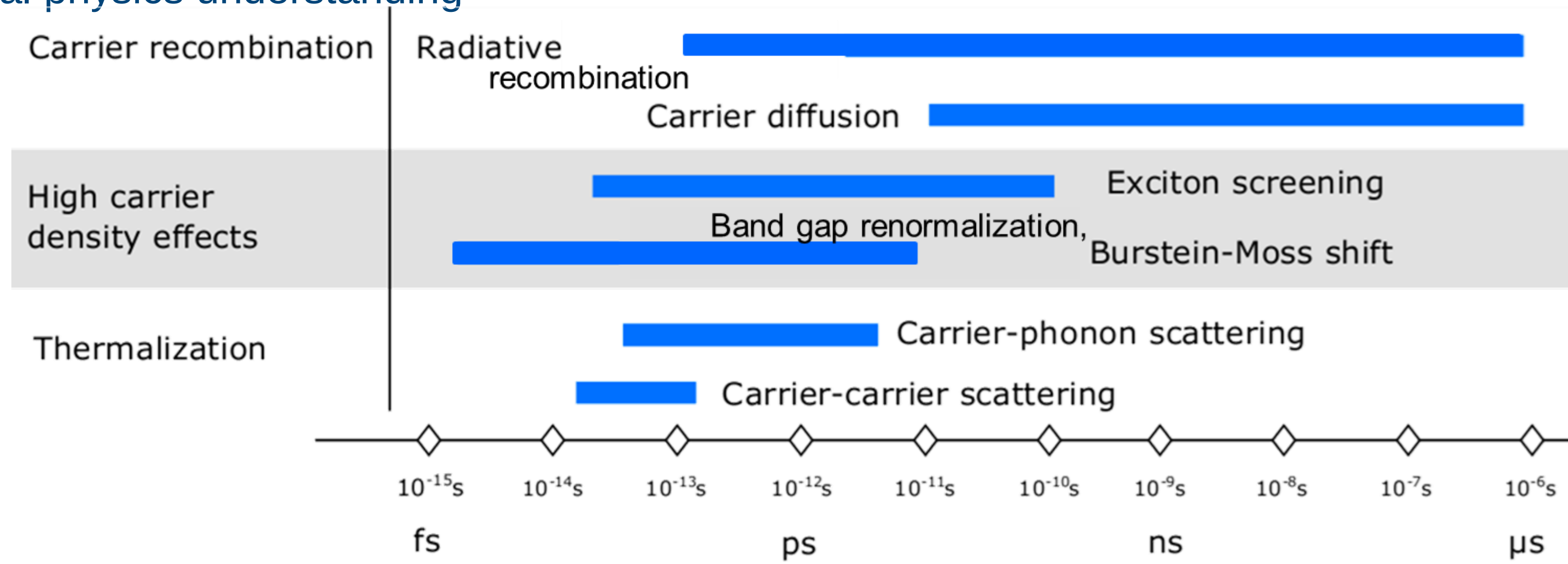
for these applications, knowledge is needed of:

- ➢ band structure as well as (joint) density of states and transition matrix elements as function of excited charge carriers
- ➢ excitation, relaxation, scattering, tunnelling and recombination dynamics
- ➢ mechanism which can lead to loss or trapping of excited charge carriers

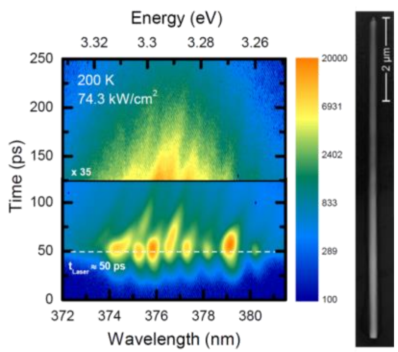
→ time resolved studies – here for example of the dielectric function dynamics after laser excitation

Ultrafast processes

fundamental physics understanding



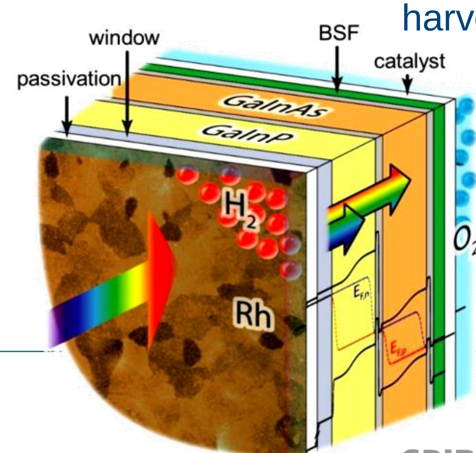
performance of opto-electronic devices, e.g.:



dynamics of lasing modes

M. Wille, RSG et al.: Nanotechnology ***27*** *(2016) 225702*

light-to-current conversion efficiency in light-harvesting applications



III-V semiconductors

GaP as one of the basic materials

ACS Energy Lett. ***3*** *(2018) 1795*

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after being hit by a strong laser pulse.

We have observed their fate by
fs-time resolved spectroscopic pump-probe ellipsometry
and understood it (tried to) by fundamental theory.

What happens to the charge carriers under absorption of light?

What is their dynamics?

Where do they go to?

Rüdiger Schmidt-Grund

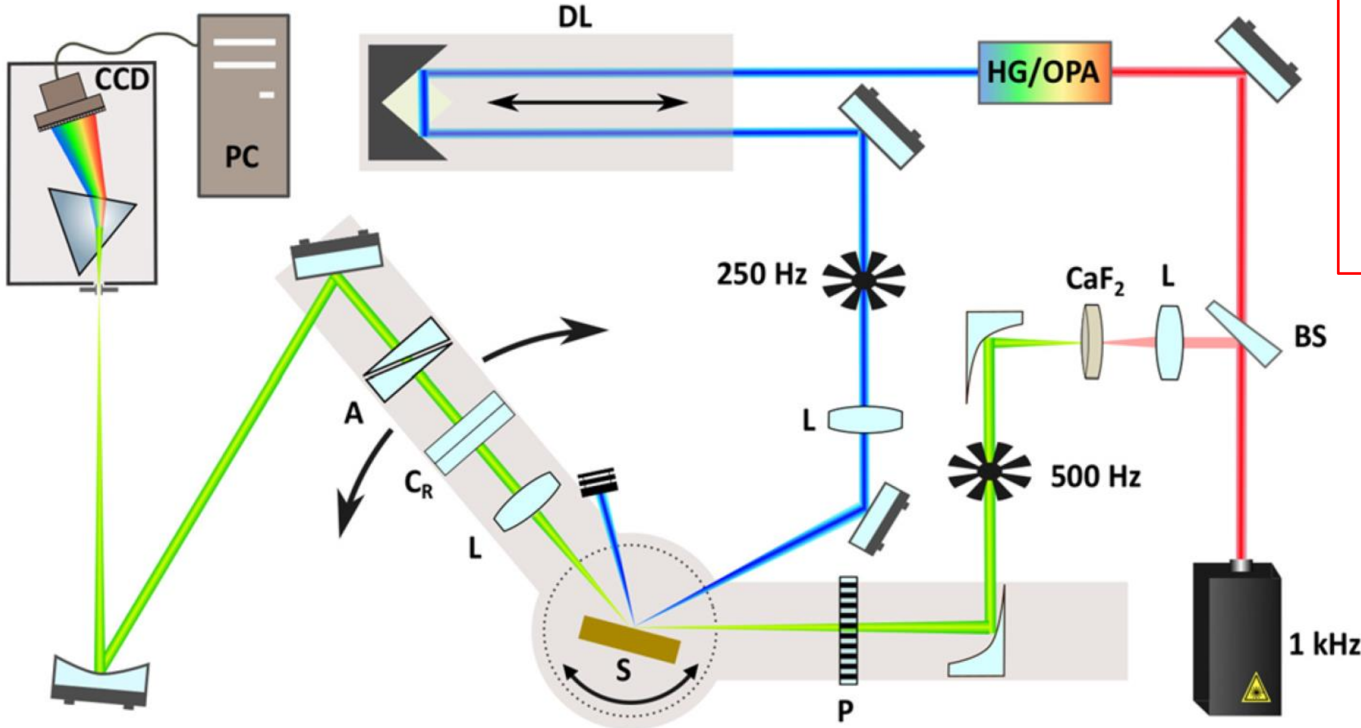
main work in method development and implementation:

Steffen Richter, Mateusz Rebarz, Oliver Herrfurth

but indeed, many others contributed considerably

Steffen Richter, Mateusz Rebarz, Oliver Herrfurth, Shirly Espinoza, Rüdiger Schmidt-Grund, and Jakob Andreasson, Rev. Sci. Instrum. ***92****, 033104 (2021)*

Broadband femtosecond spectroscopic ellipsometry



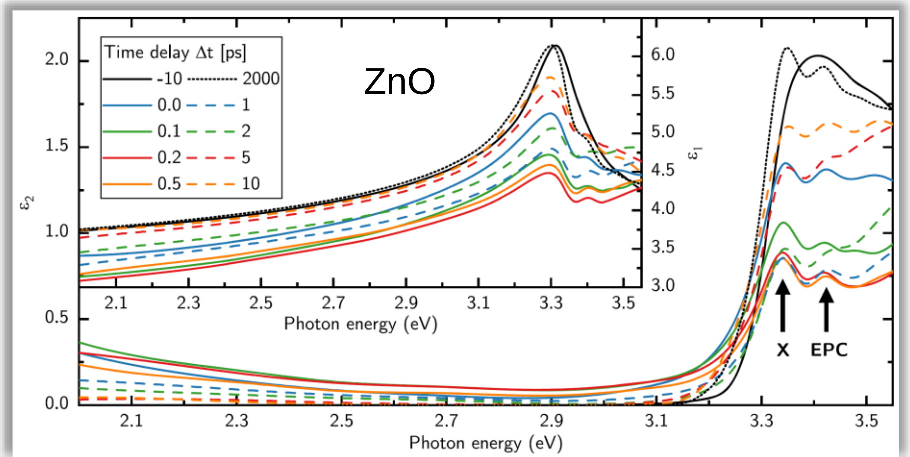
setup build up and located @ ELI beamlines facility in Dolní Břežany (Prague), Czech Republic

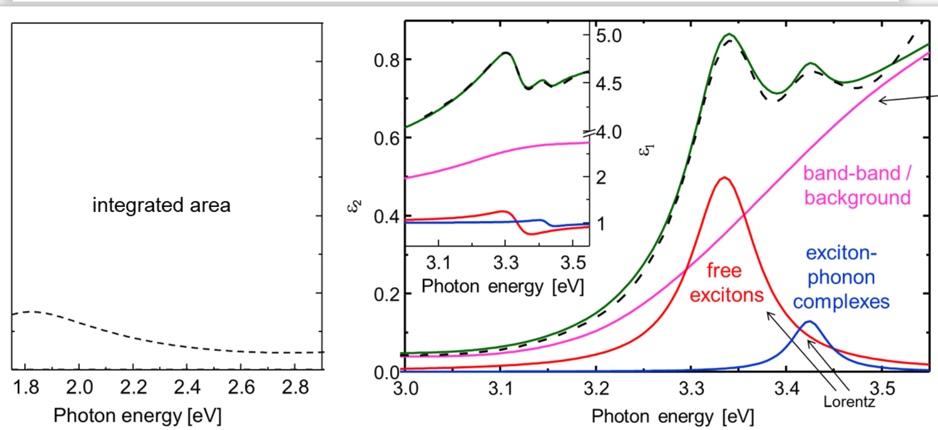
Method

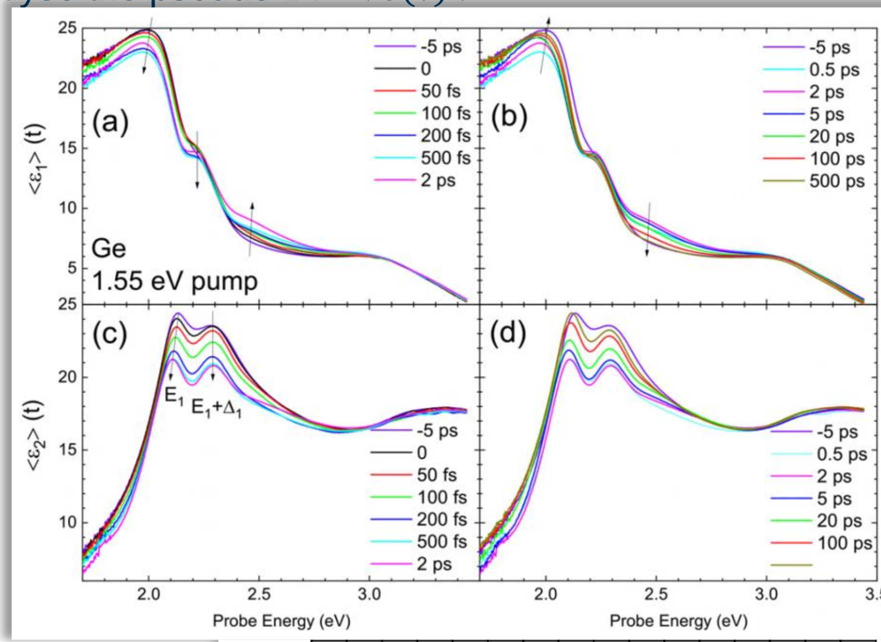
Data analysis

we model data for each $\Delta t = \tau$ to obtain the complex DF $\varepsilon(\tau)$

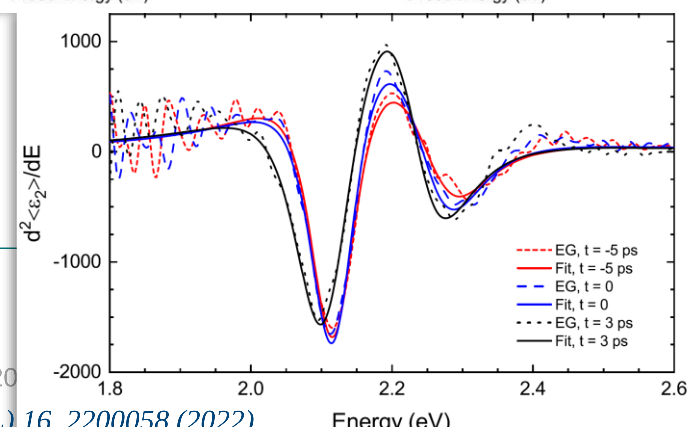
we model either $\varepsilon(\tau)$ or $\Delta\varepsilon(\tau) = \varepsilon(\tau) - \varepsilon(\tau = 0)$ or analyse the pseudo-DF $< \varepsilon(\tau) >$







Ge



New J. Phys. 22, 083066 (2020) *Appl. Phys. Lett. 115, 052105 (2019)* *physica status solidi (RRL) 16, 2200058 (2022)*

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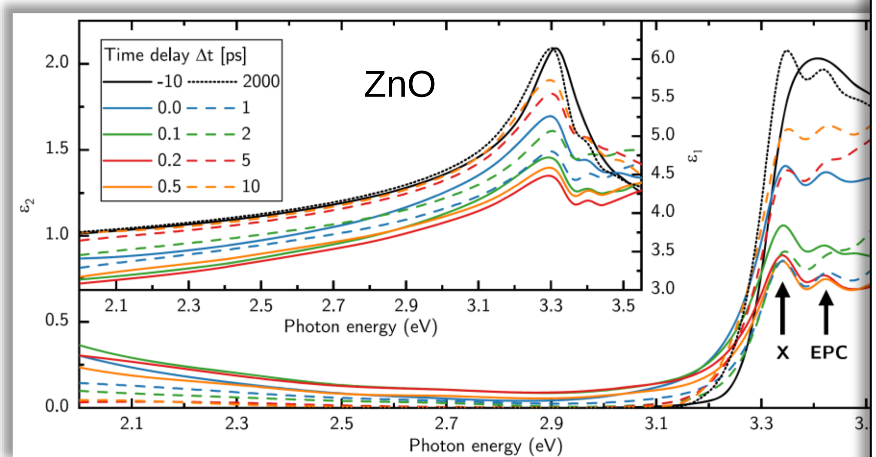

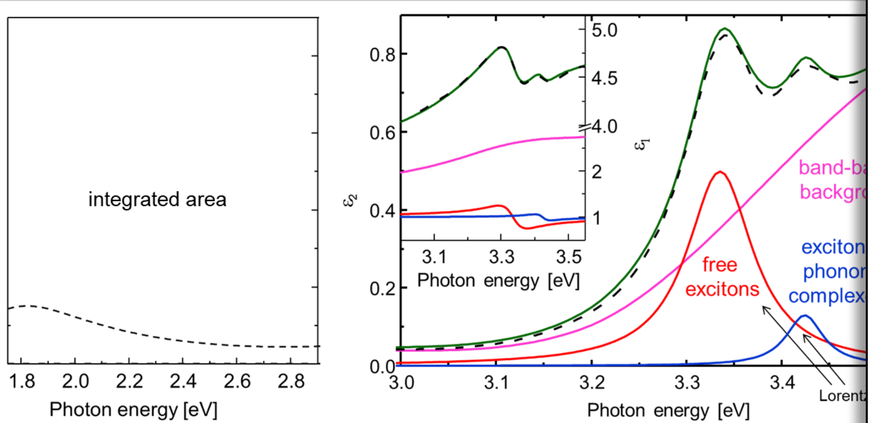

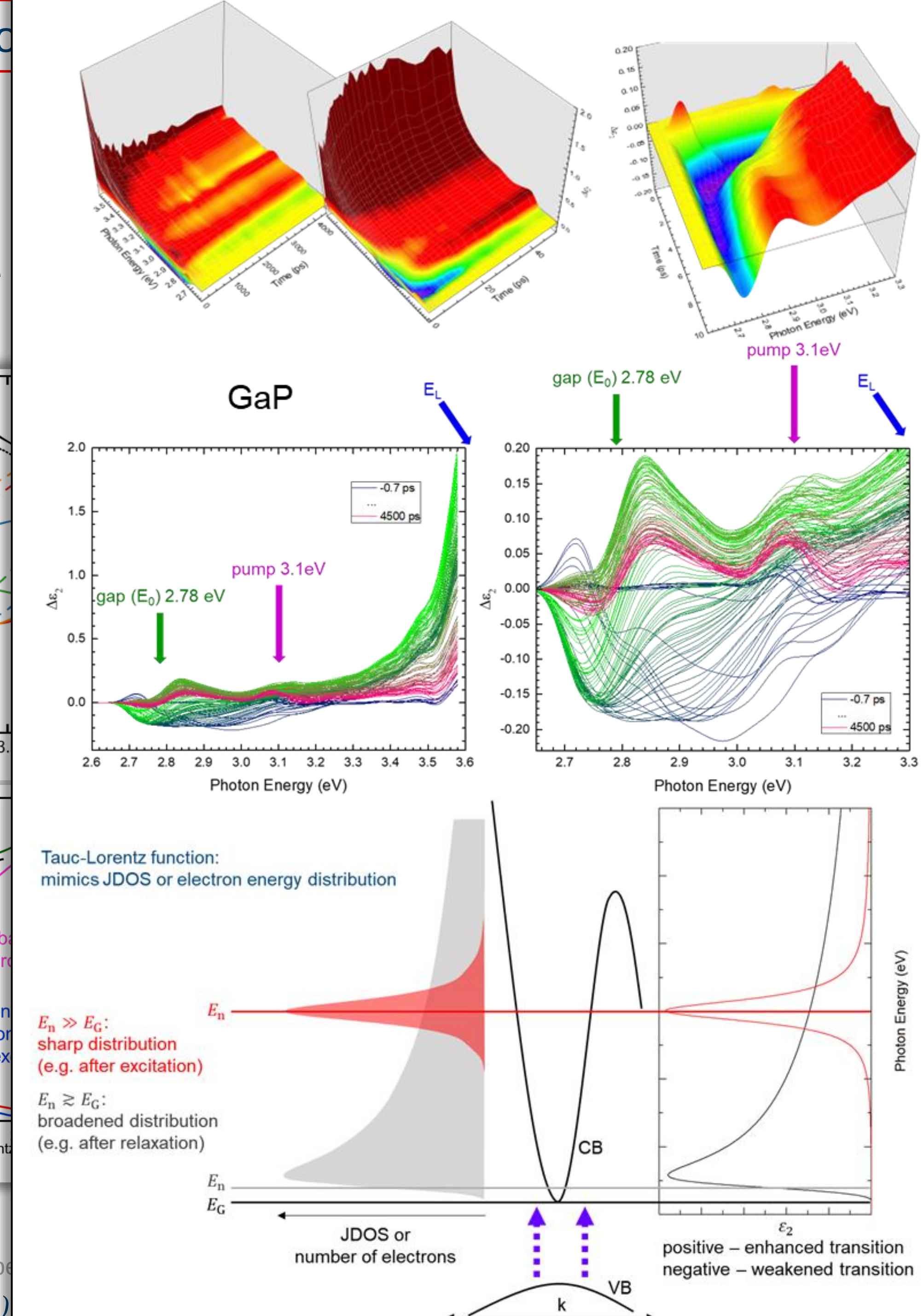

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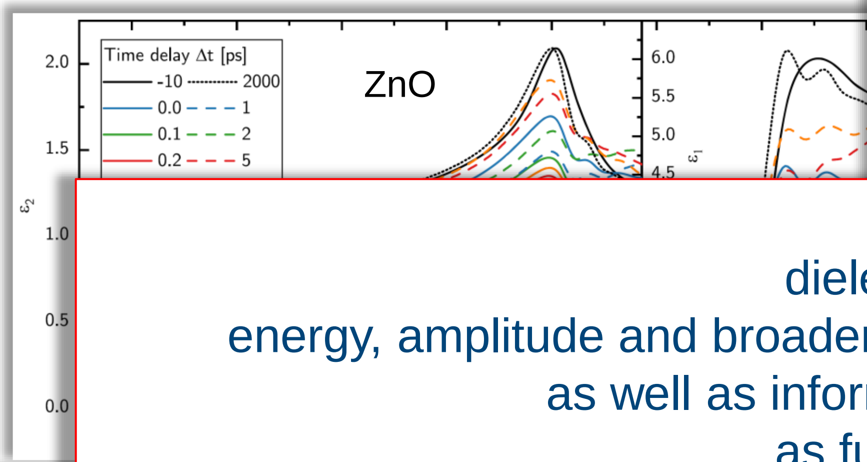

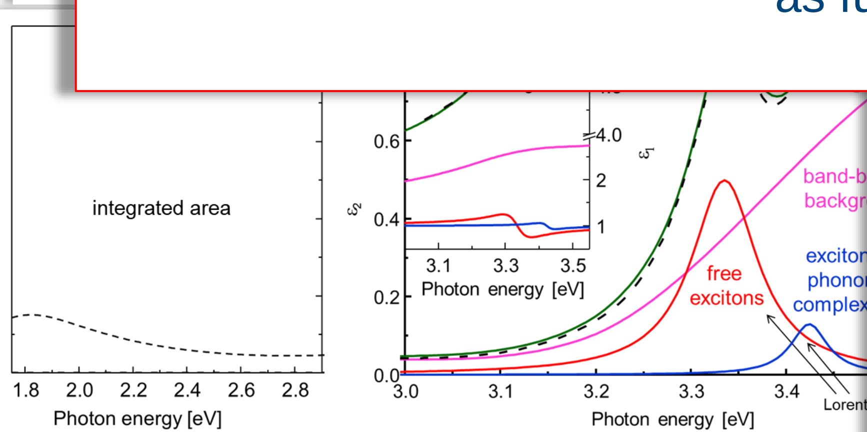

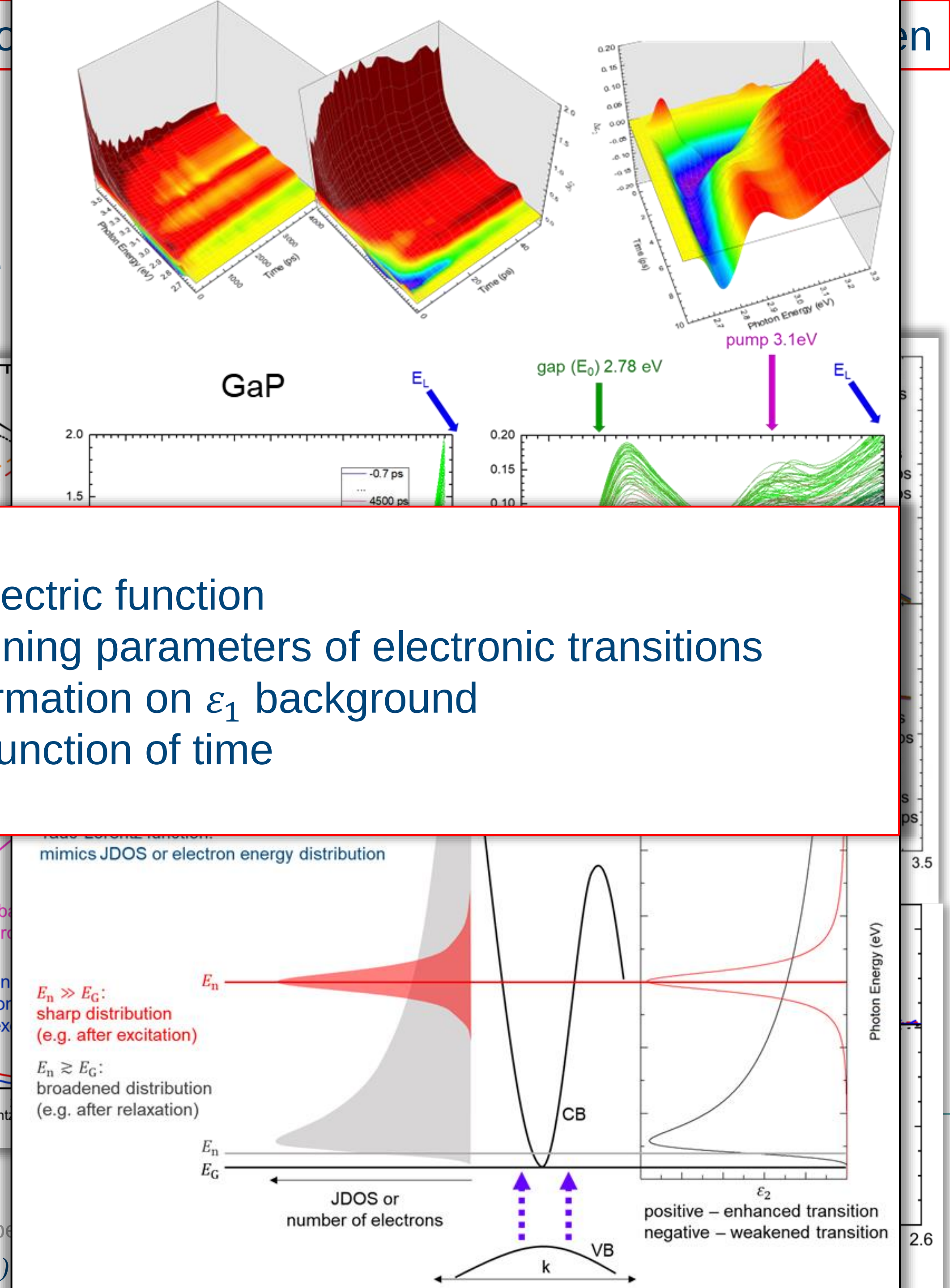

dielectric function

energy, amplitude and broadening parameters of electronic transitions

as well as information on ε_1 background

as function of time

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André, Noah

Method

Compare to fundamental theory

band structure and DF by ground-state plane-wave density functional theory:

transition probability (rate):

$$w_{nm}\left(\vec{k},\hbar\omega\right)=\frac{2\pi}{\hbar}\int_{\vec{k}}\left|\widehat{\mathbf{M}}_{nm}\right|^2\delta\left(E_m\left(\vec{k}_m\right)-E_n\left(\vec{k}_n\right)\pm\hbar\omega\right)\mathrm{d}\vec{k}$$

with joint density of states

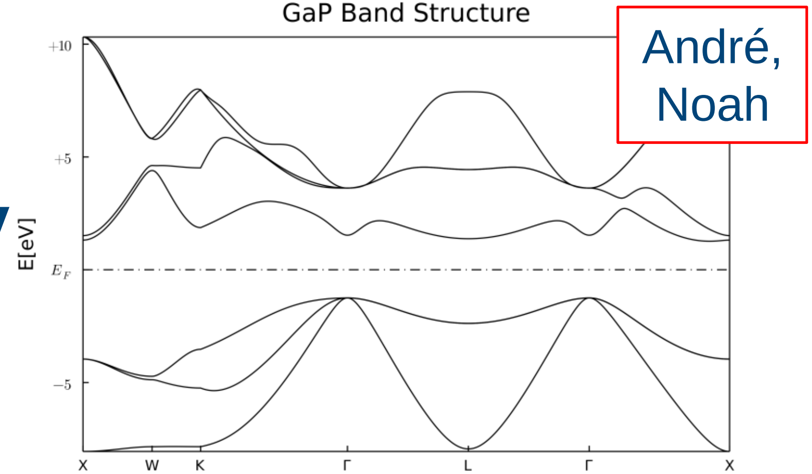
$$\rho_{nm}=\int_{\vec{k}}\delta\left(E_m\left(\vec{k}_m\right)-E_n\left(\vec{k}_n\right)\pm\hbar\omega\right)\mathrm{d}\vec{k}$$

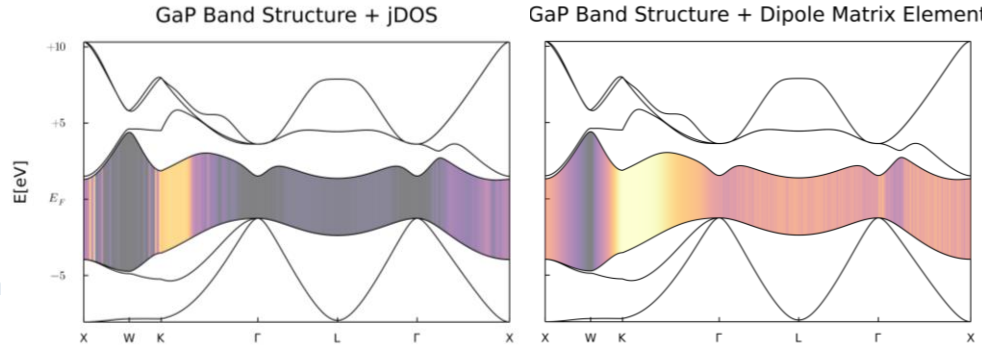
and transition dipole matrix element

$$\widehat{\mathbf{M}}_{nm}=\left\langle u^{(i)}_{n,\vec{k}}\middle|\vec{p}\middle|u^{(f)}_{n,\vec{k}}\right\rangle$$

(JDOS and Matrix elements between all bands)

$\Rightarrow$ dielectric function





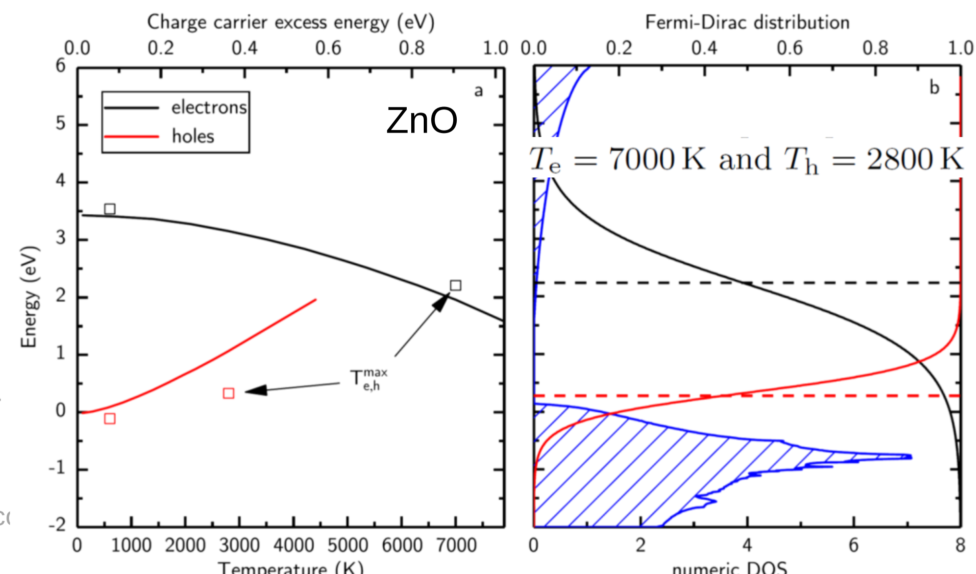
transition matrix elements from Kohn-Sham eigenvectors by norm-conserving pseudo-potentials

also band symmetries analysed and considered

Charge carrier statistics and Fermi-distributions (carrier temperature, excess-energy)

$$\Delta E_{\mathrm{e/h}}=\frac{3}{2}k_{\mathrm{B}}T_{\mathrm{e/h}}\qquad\Delta E_{\mathrm{e}}=\frac{E_{\mathrm{pump}}-E_{\mathrm{gap}}}{1+m_{\mathrm{e}}/m_{\mathrm{h}}}$$

$$\Delta E_{\mathrm{h}}=\frac{E_{\mathrm{pump}}-E_{\mathrm{gap}}}{1+m_{\mathrm{h}}/m_{\mathrm{e}}}$$



New J. Phys. 22, 083066 (2020)

The tale of electrons

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What happens to the charge carriers under absorption of light?

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Where do they go to?

let's go through the story – in the world of

optoelectronic (Si, Ge, InP, GaP, ZnO) materials

The tale of electrons

exciting with high-intense laser beam: electrons and holes occupy formerly “empty” conduction (CB) and valence (VB) band states → hot charge carriers with temperatures up to several thousands of Kelvin

- → carriers spread within the entire Brillouin zone (BZ) to energetically matching bands
- → band gap renormalization (BGR) → redshift of transition energies between VB and CB
- → transitions between now occupied states are Pauli-blocked → reduction of absorption at the respective energies
- → excess electrons and holes can be excited by the probe light → arising of new intra VB and/or intra CB transitions → increase of absorption at the respective energies
- → when the material exhibits excitons: they are screened by the excited carriers, but may also form so-called Mahan excitons → energy redshift and absorption bleaching
- → collective free charge carrier oscillations (Drude)

in the following time (sub-ps … ps), excited carriers scatter amongst each other and with the lattice (phonons):

- → energy dissipation as heat → redshift of transition energies
- → relaxation to band minima → redshift of the Pauli blocked absorption features
- → formation of hot-electron–hot-phonon states (hot exciton-phonon complexes)
- → hot electrons can propagate ballistically over several µm
- → phonons oscillate coherently

finally, the system relaxes back to equilibrium either due to radiative or non-radiative carrier recombination within, depending on the material, some tens of ps (e.g. ZnO) or up to several ns (e.g. GaP)

André, Carola, Noah, Oliver, Shirly, Stefan, Steffen

The tale of electrons – Excitation

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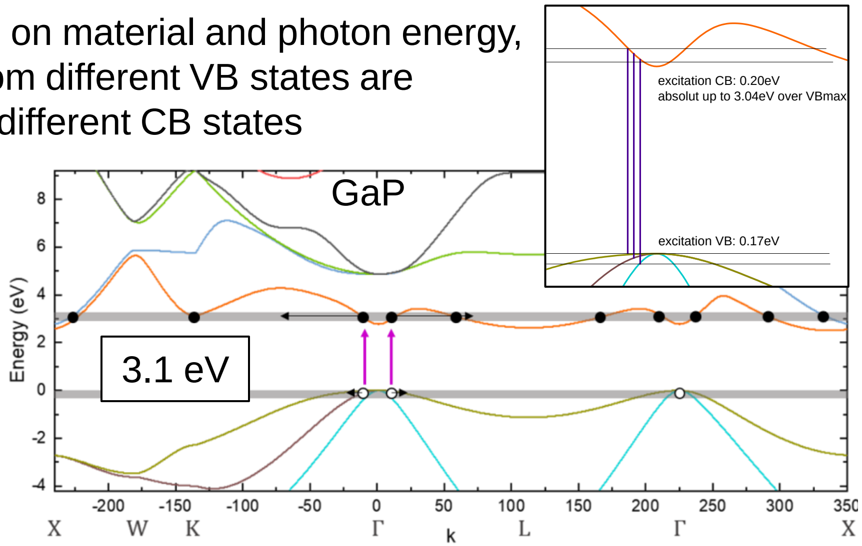
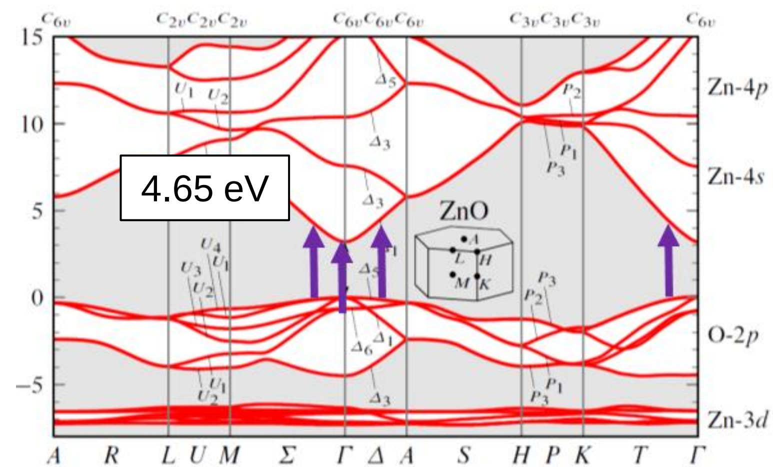
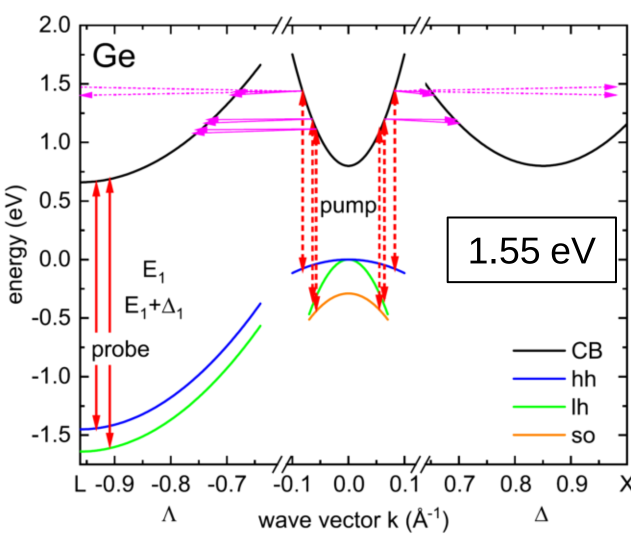
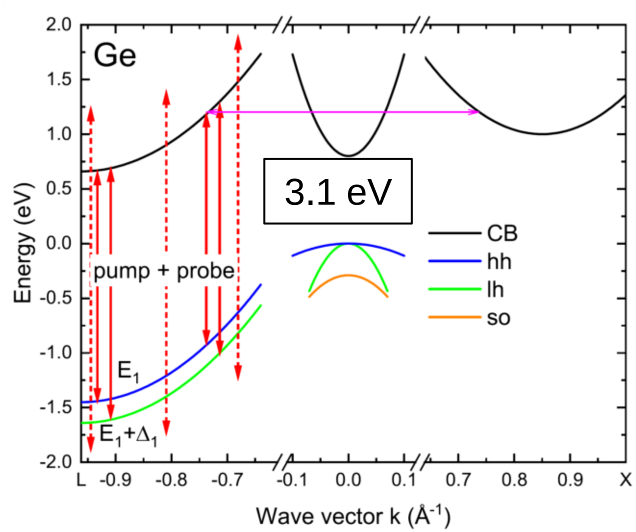
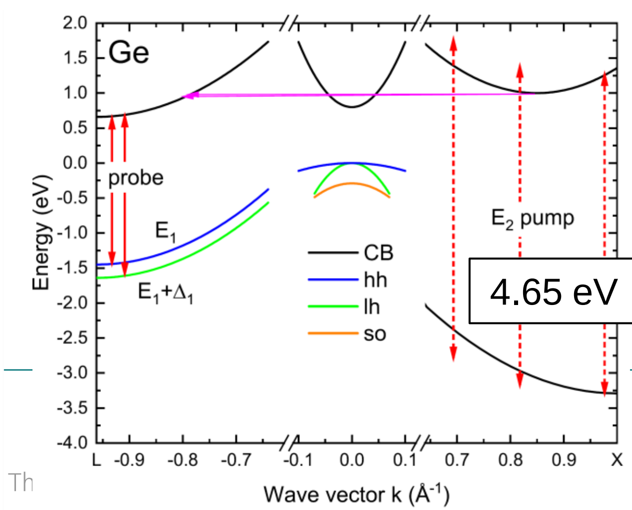
depending on material and photon energy, carriers from different VB states are excited to different CB states











Appl. Phys. Lett. 115, 052105 (2019) *New J. Phys. 22, 083066 (2020)*

Noah

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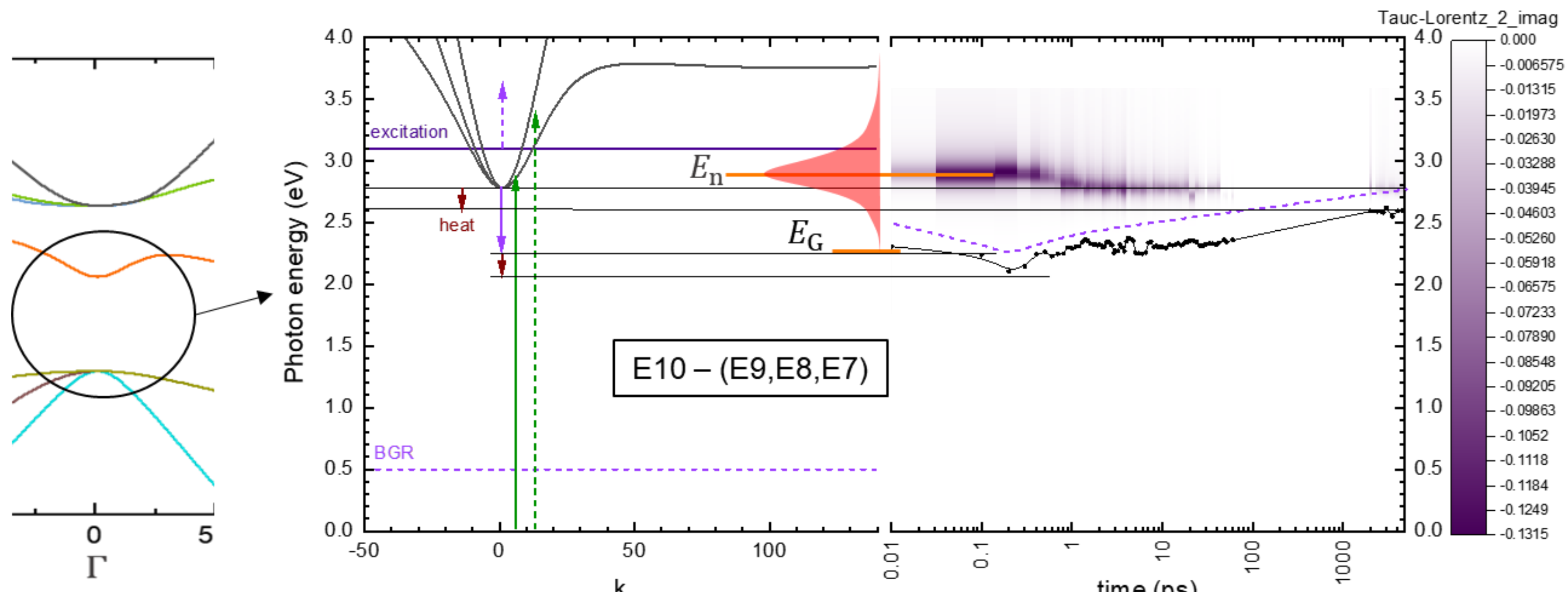
- → band gap renormalization (BGR) → redshift of transition energies between VB and CB by ΔE_{BGR}

scattering into entire Brillouin zone within energy range $\Delta E_{\mathrm{excitation}} + \Delta E_{\mathrm{BGR}}$

example GaP: we obtain ΔE_{BGR} from time evolution of E_{G} of the TcL describing blocking at Γ

in the graph the energy difference of the lowest CB to the uppermost VBs is displayed



additional red-shift contribution for later time by heating due to energy transfer to the lattice

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The tale of electrons – Excitation and shortly after

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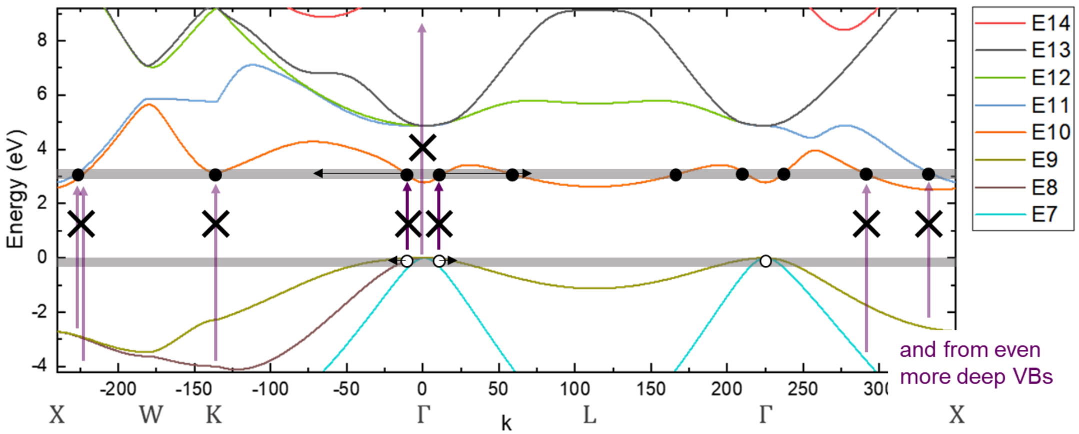
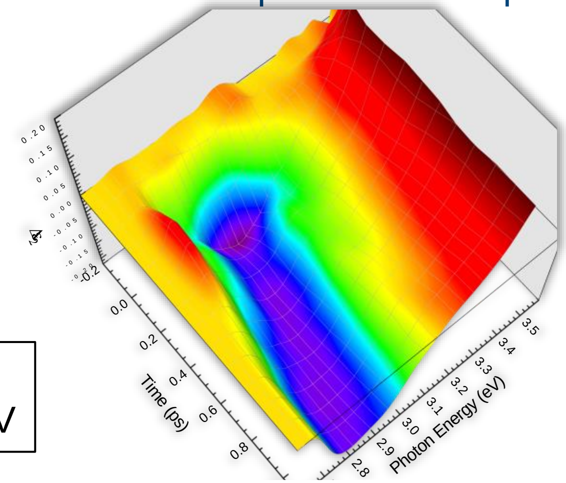
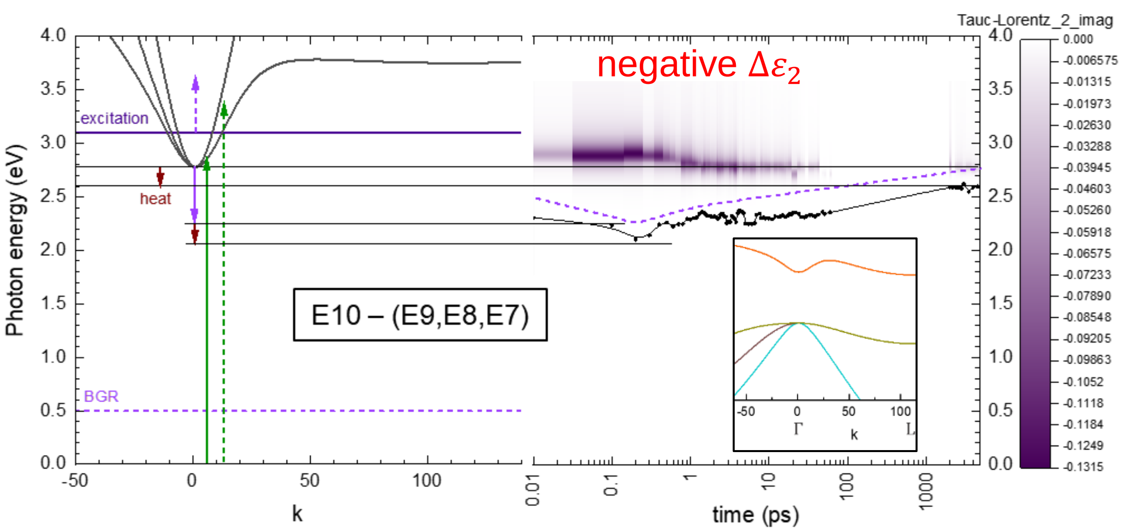
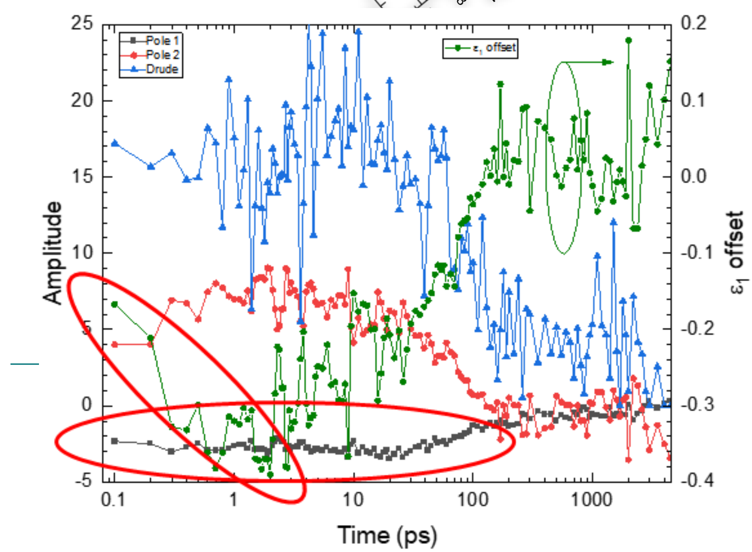




GaP
3.1 eV





André, Oliver, Steffen

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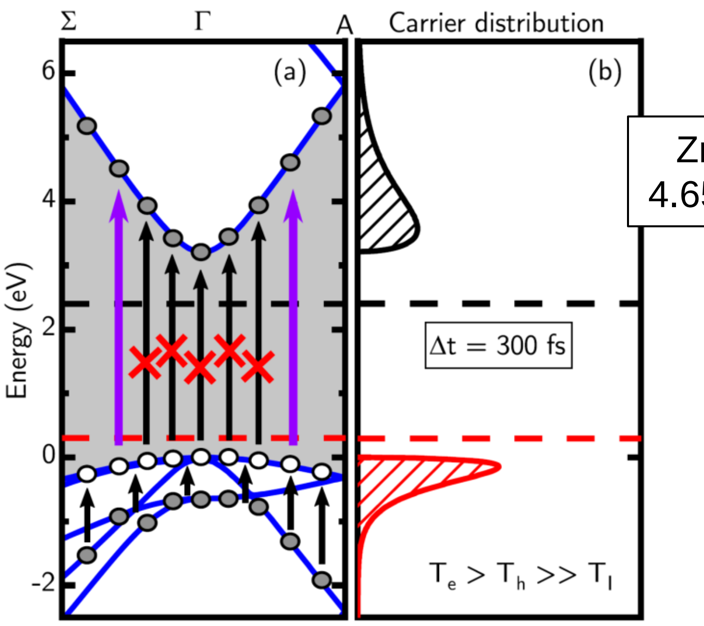
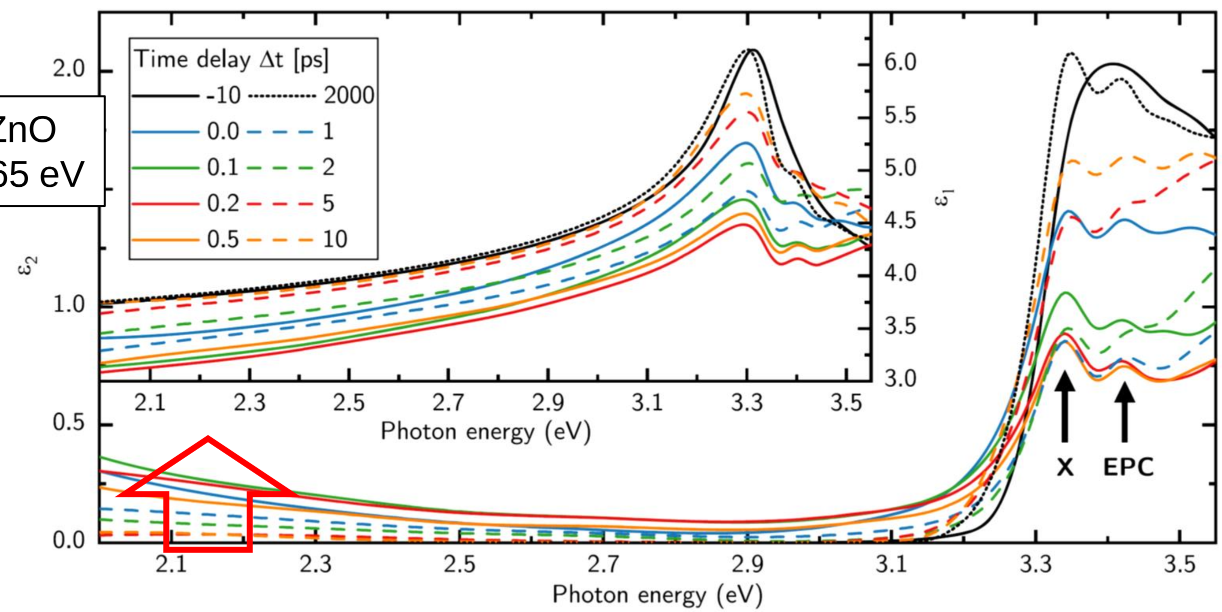
1.: intra-VB transition near Γ (electrons: lower VB to topmost VB; holes: lowest to higher hole bands)



ZnO
4.65 eV



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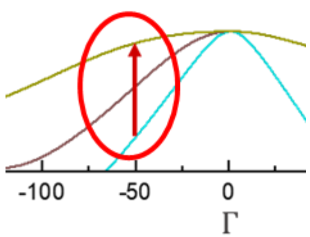
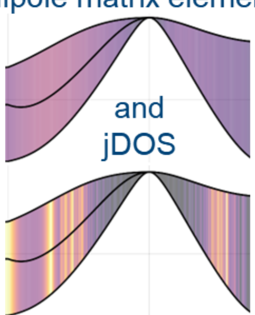


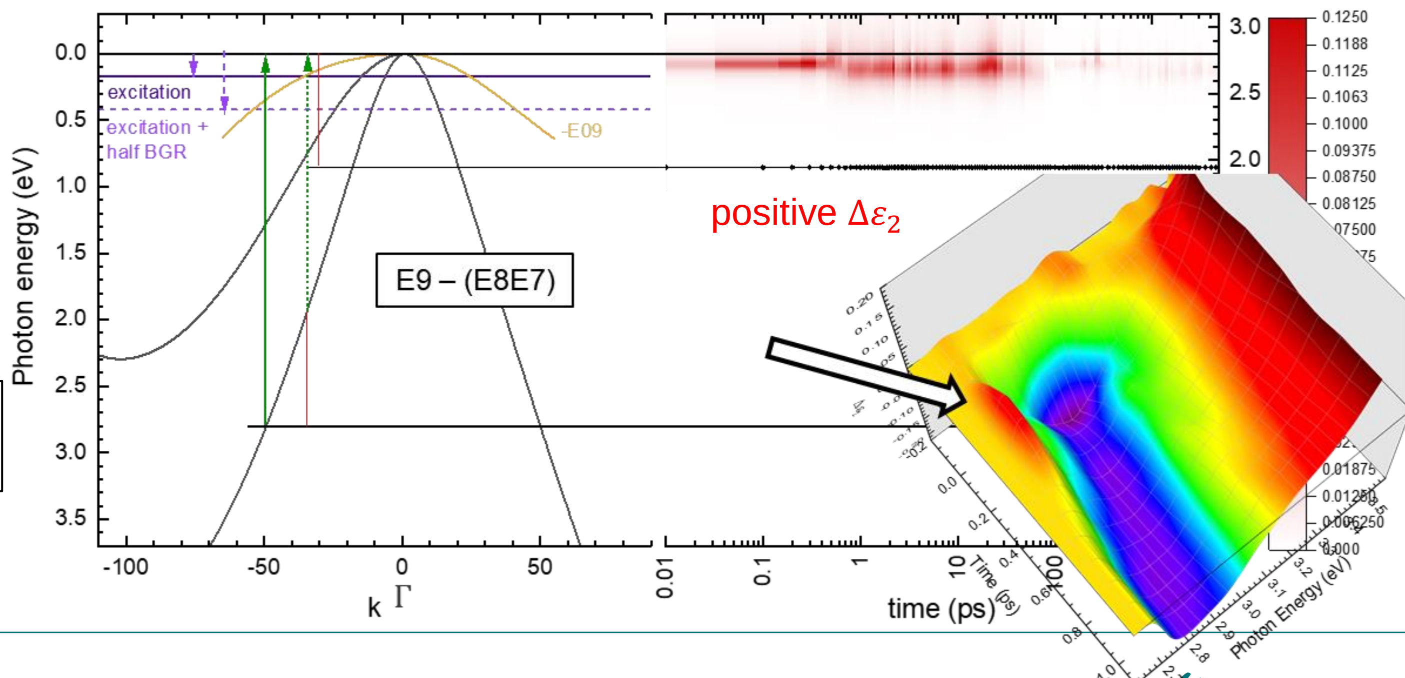
dipole matrix elements



and jDOS

have finite values

GaP
3.1 eV



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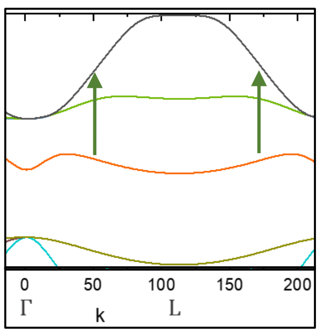
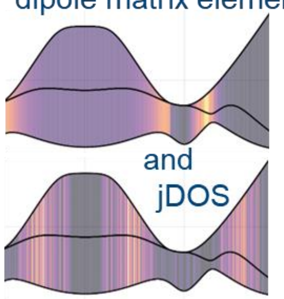
2.: intra-CB transitions along L (hot electrons from lowest CB to higher conduction bands)



dipole matrix elements

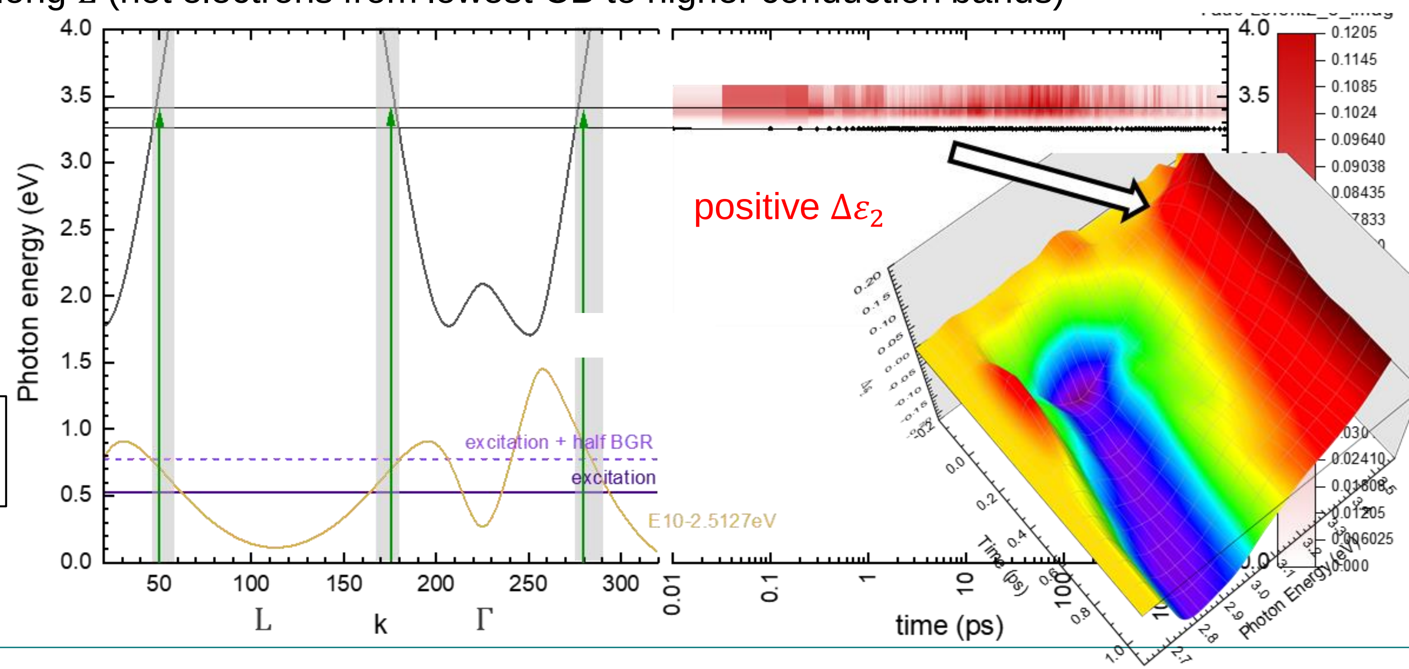


and jDOS

have finite values

but:
DPM quite weak
→ stationary theory correct?

GaP
3.1 eV



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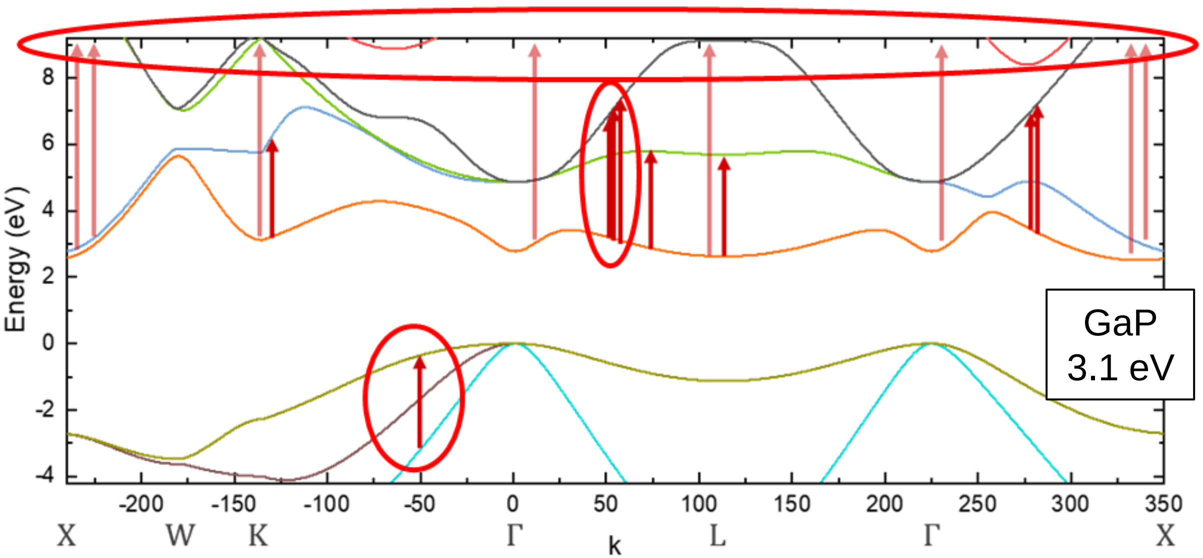
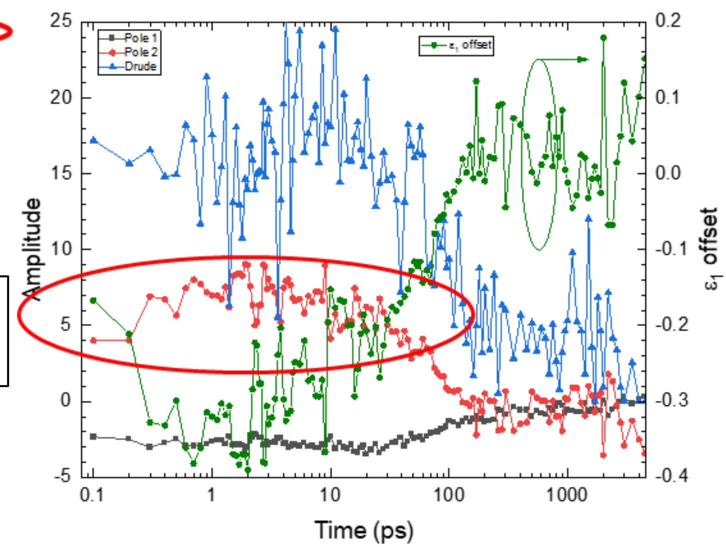
… and to higher bands (outside spectral range) expressed by positive pole





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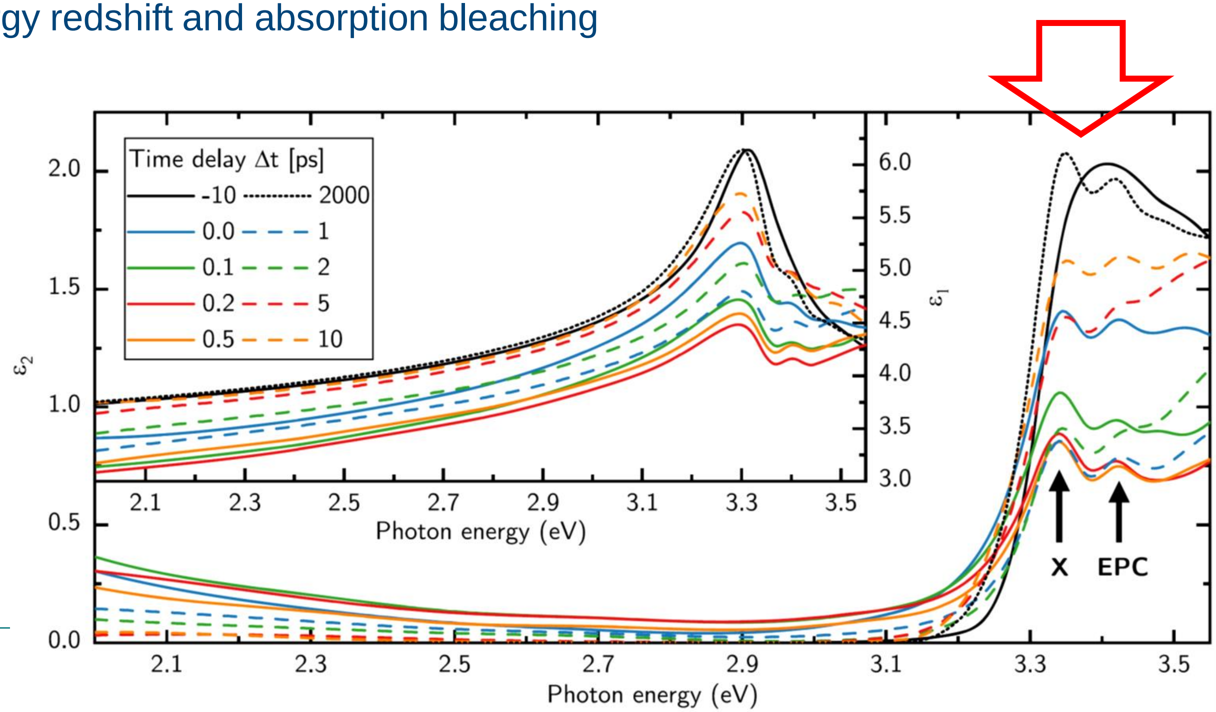
ZnO
4.65 eV

excitons persisted at any time although exceeding Mott density → not only screening length matters, but also wavevector – at high excess-energy, strong non-parabolicity → misfit → reduced screening
→ Mahan excitons

Schleife et al, Phys. Rev. Lett. ***107*** *236405*



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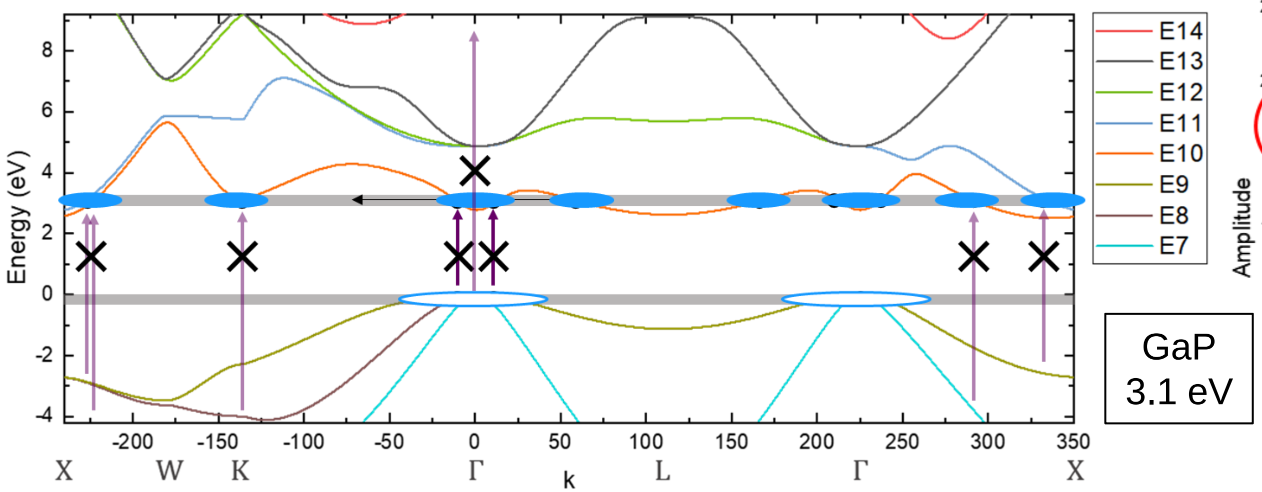
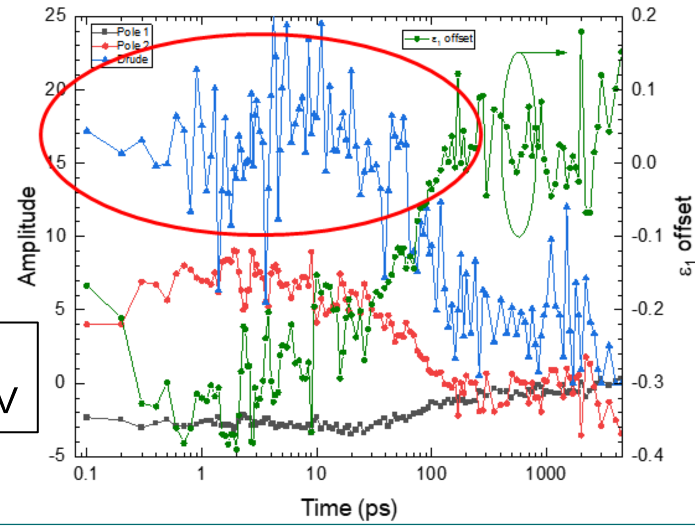
- → when the material exhibits excitons: they are screened by the excited carriers, but may also form so-called Mahan excitons → energy redshift and absorption bleaching
- → collective free charge carrier oscillations (Drude) – electron and hole lakes in CB and VB





Rüdiger Schmidt-Grund
12th WSE Satellite Meeting on Time-Resolved Spectroscopic Ellipsometry, 09/2023

The tale of electrons – in the following ps

in the following time (sub-ps … ps), excited carriers scatter amongst each other and with the lattice (phonons):

- → energy dissipation as heat → redshift of transition energies
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- → formation of hot-electron–hot-phonon states (hot exciton-phonon complexes)
- → hot electrons can propagate ballistically over several µm
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finally, the system relaxes back to equilibrium either due to radiative or non-radiative carrier recombination within, depending on the material, some tens of ps (e.g. ZnO) or up to several ns (e.g. GaP)

André, Carola, Noah, Oliver, Shirly, Stefan, Steffen

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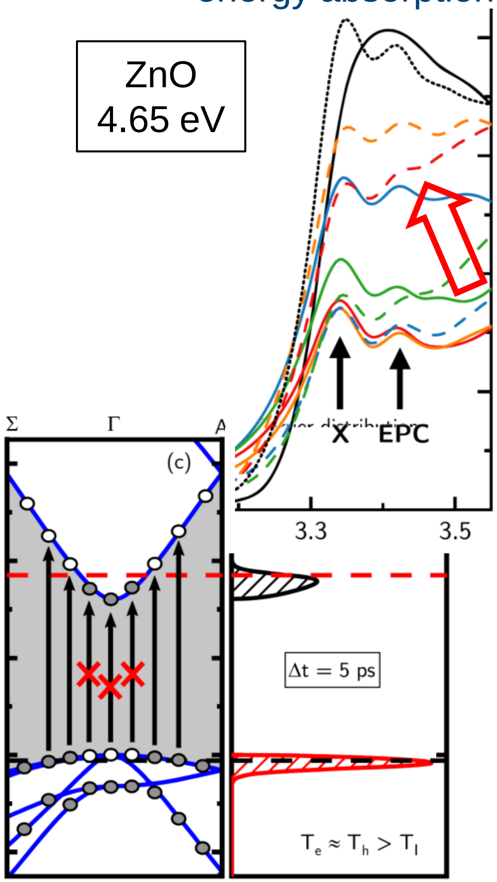
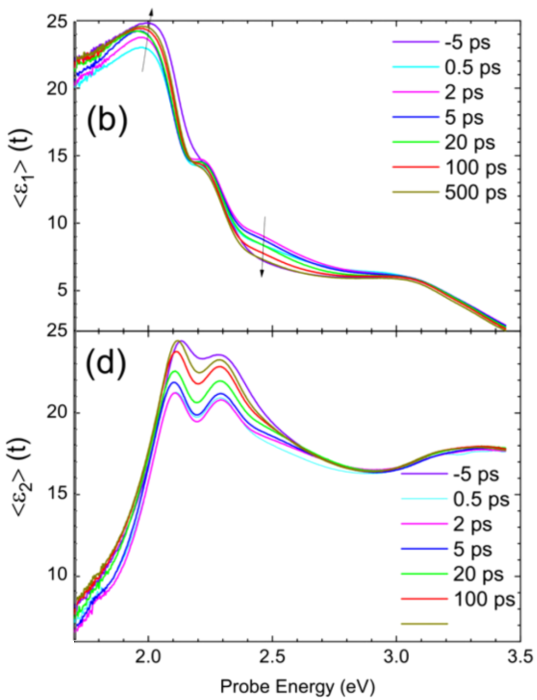
in the following time (sub-ps … ps), excited carriers scatter amongst each other and with the lattice (phonons):

- → energy dissipation as heat → redshift of transition energies
- → relaxation to band minima → redshift of the Pauli blocked absorption features and recovery of high-energy absorption

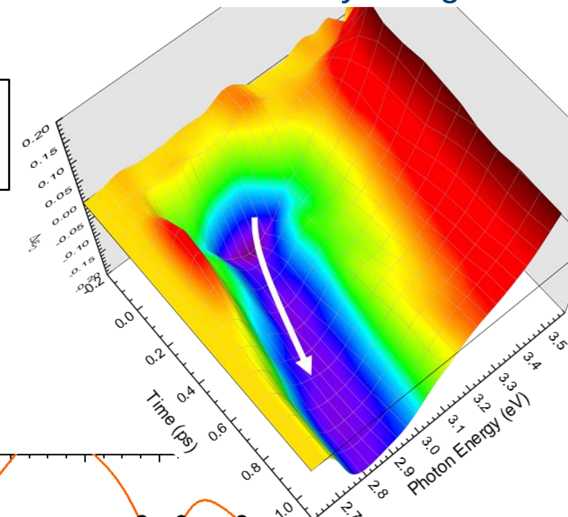
ZnO
4.65 eV

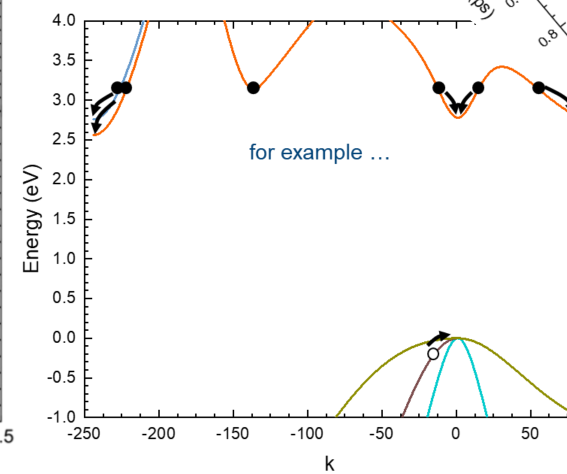


Ge
1.55 eV



GaP
3.1 eV





New J. Phys. 22, 083066 (2020) *Appl. Phys. Lett. 115, 052105 (2019)*

André, Oliver, Steffen

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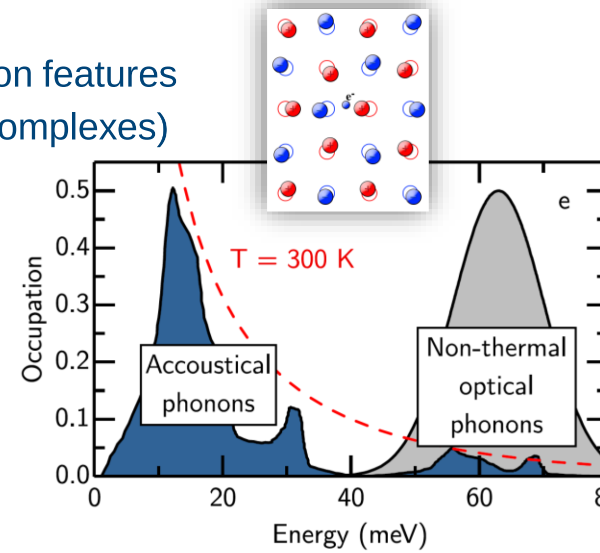


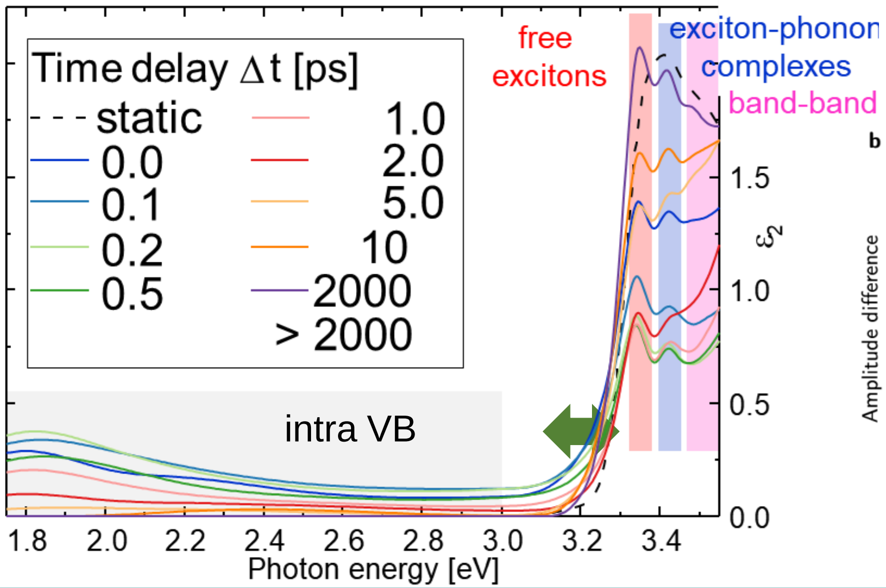
electron thermalization slowed down by hot-phonon effect:

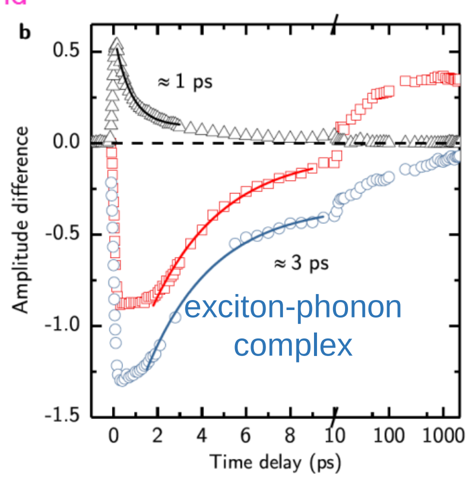
electron-phonon scattering: creation of hot phonons
→ in turn hot electrons created by phonon decay

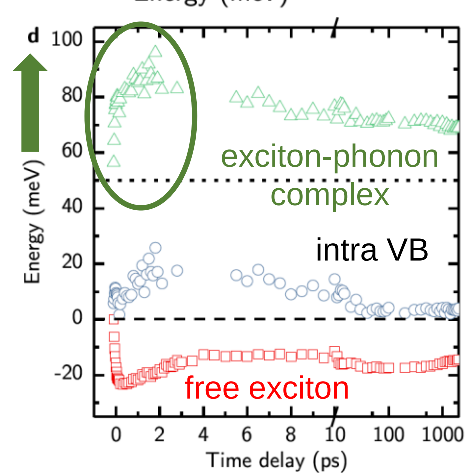
exciton-phonon binding energy increases

high-energy exciton-phonon complex (exciton-polaron)

ZnO
4.65 eV





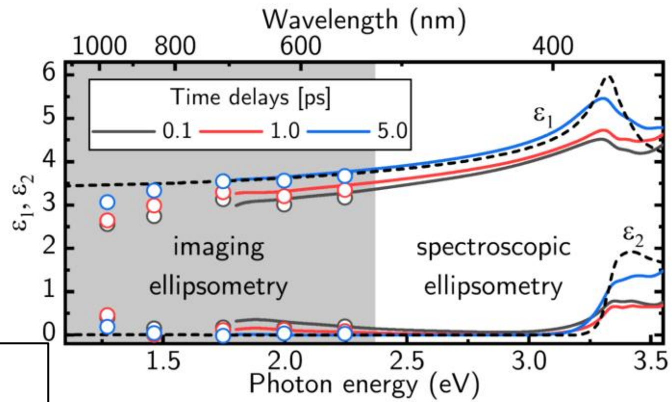


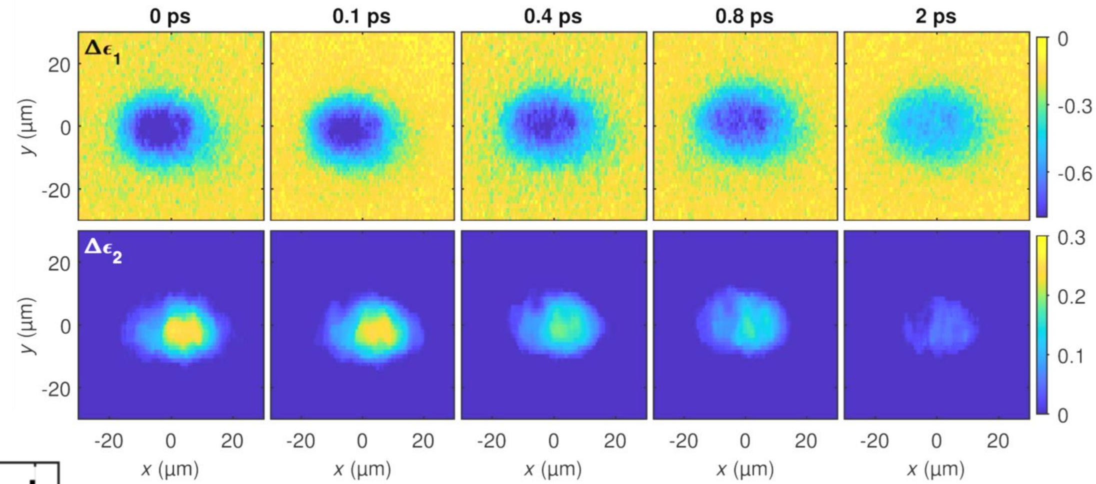
New J. Phys. 22, 083066 (2020)

Oliver, Marcus, Theo

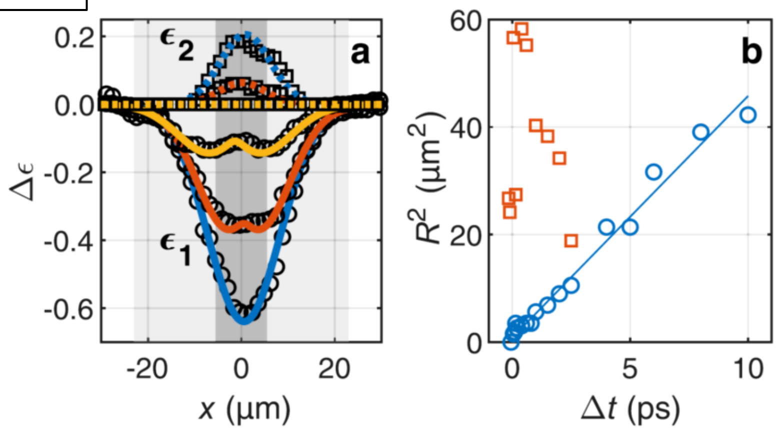
The tale of electrons – in the following ps

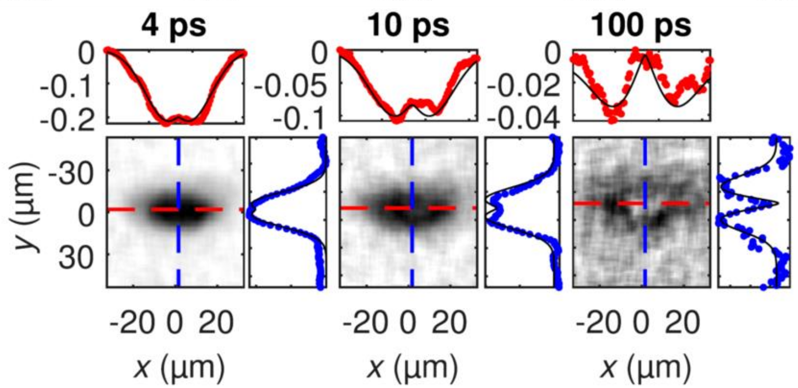
→ hot electrons can propagate ballistically over several µm





ZnO
4.67 eV





→ combined effect of carrier cooling and fast carrier transport: effective diffusion coefficient $1.1 \times 10^4\ \mathrm{cm^2/s}$

→ ring structure: can be explained by a random-walk model including ballistic transport due to the thermal gradient induced by the hot-phonon effect (speed several 10^5 m/s, electron by 2.5 times faster than holes)

Appl. Phys. Lett. 115, 212103 (2019)

Carola, Stefan

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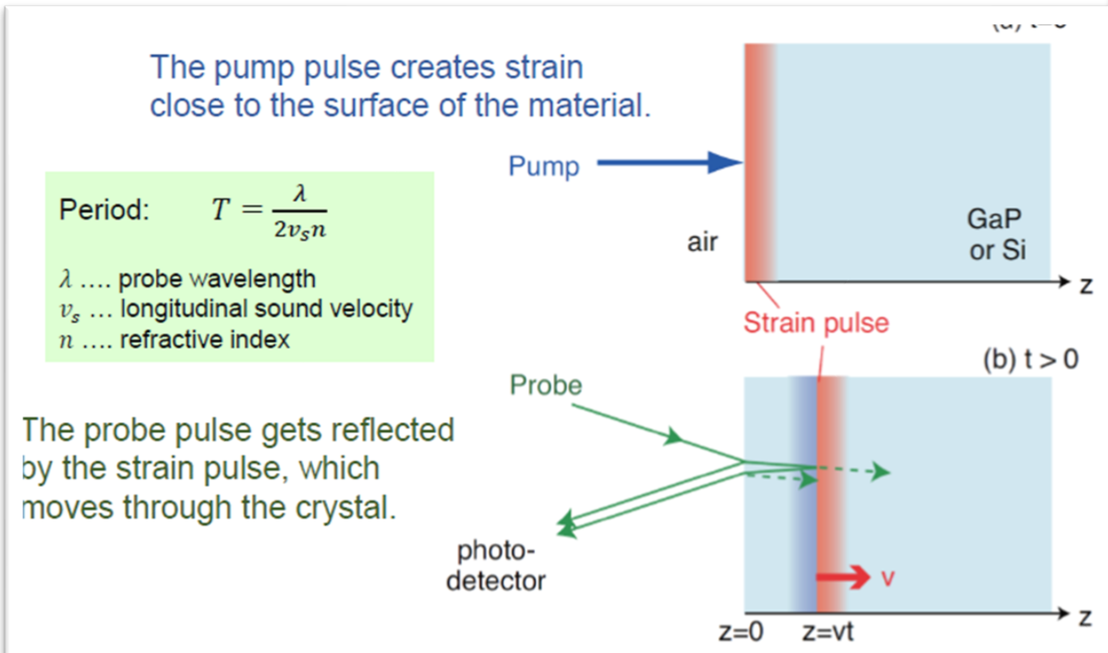
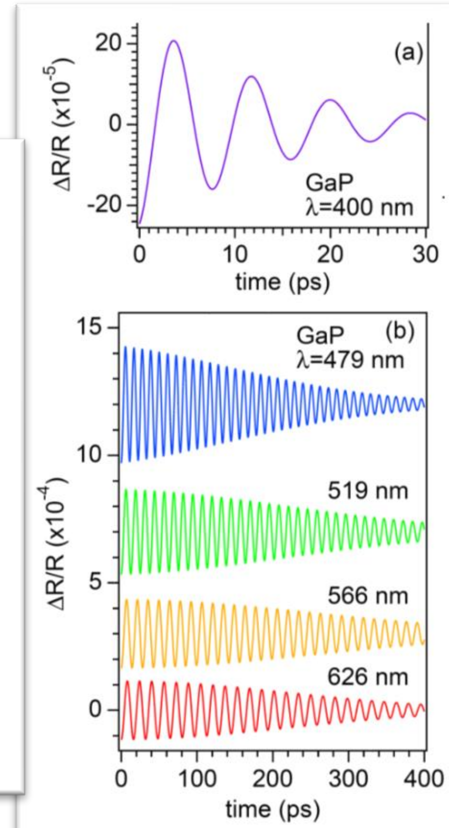




K. Ishioka, V. Rustagi, U. Höfer, H. Petek, and C. J. Stanton, Phys. Rev. B 95, 035205 (2017).

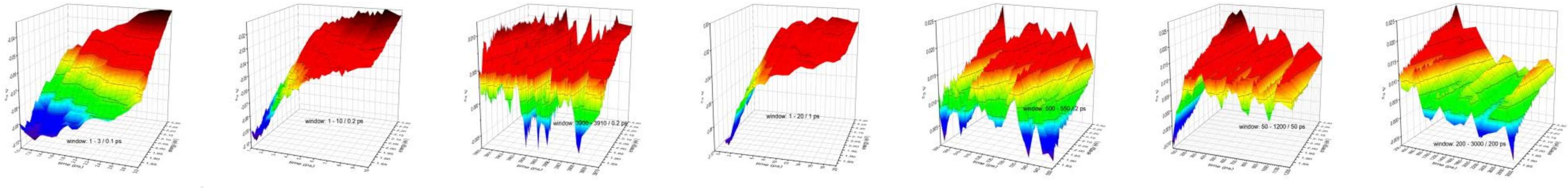
Phys. Status Solidi RRL 16, 2200058 (2022)

Noah, Younes

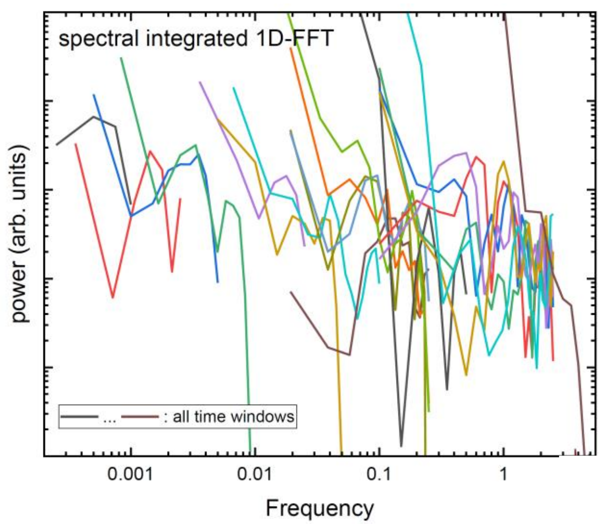
Long-lasting coherent phonon oscillations in Gallium Phosphide

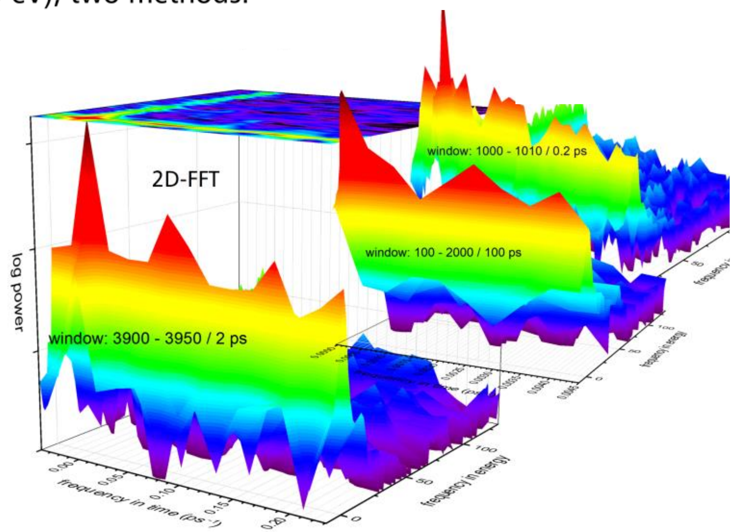
time-delayed spectra recorded in various windows with different time steps from 0.1 to 500ps distributed in the full time range up to 4500ps

examples for $\Delta\langle\varepsilon_2\rangle$-spectra for some windows:



Fourier-analysis of $\Delta\langle\varepsilon_2\rangle$ -spectra (1.8 – 2.3 eV), two methods:





Noah, Younes

Long-lasting coherent phonon oscillations in Gallium Phosphide

⇒ yields a large bunch of frequencies:

- allow only those who have at least 3 data points (step width) per period and 3 periods per window
- averaging of frequencies bunching together
- interferences of probe-light within a cavity, where one mirror is formed by the propagating sound wave (density wave) – varying cavity length in time
- related sound velocity according: $T = \frac{1}{\nu} = \frac{\lambda}{2nv_s}$

André, Oliver, Steffen

The tale of electrons – long-time story

in the following time (sub-ps … ps), excited carriers scatter amongst each other and with the lattice (phonons):

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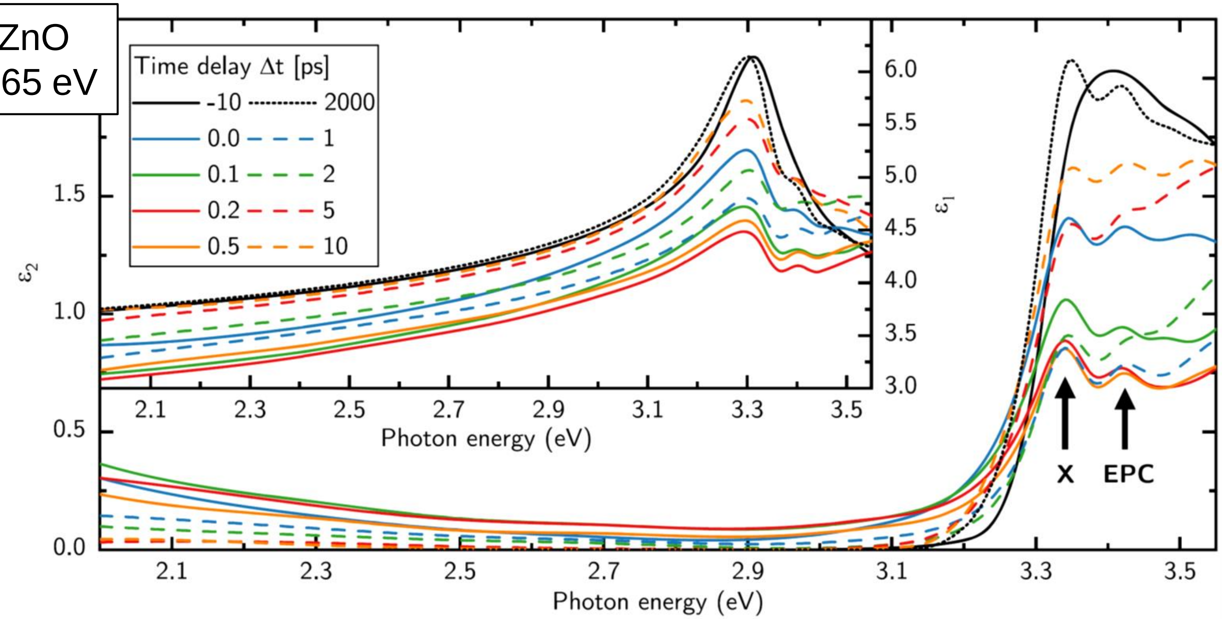
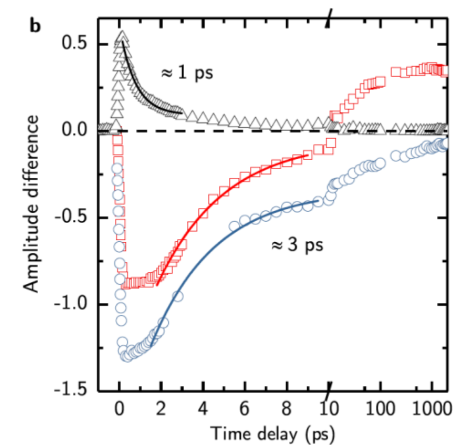
4.65 eV



fast radiative recombination (emission dynamics: photoluminescence / lasing)



New J. Phys. 22, 083066 (2020)

Noah

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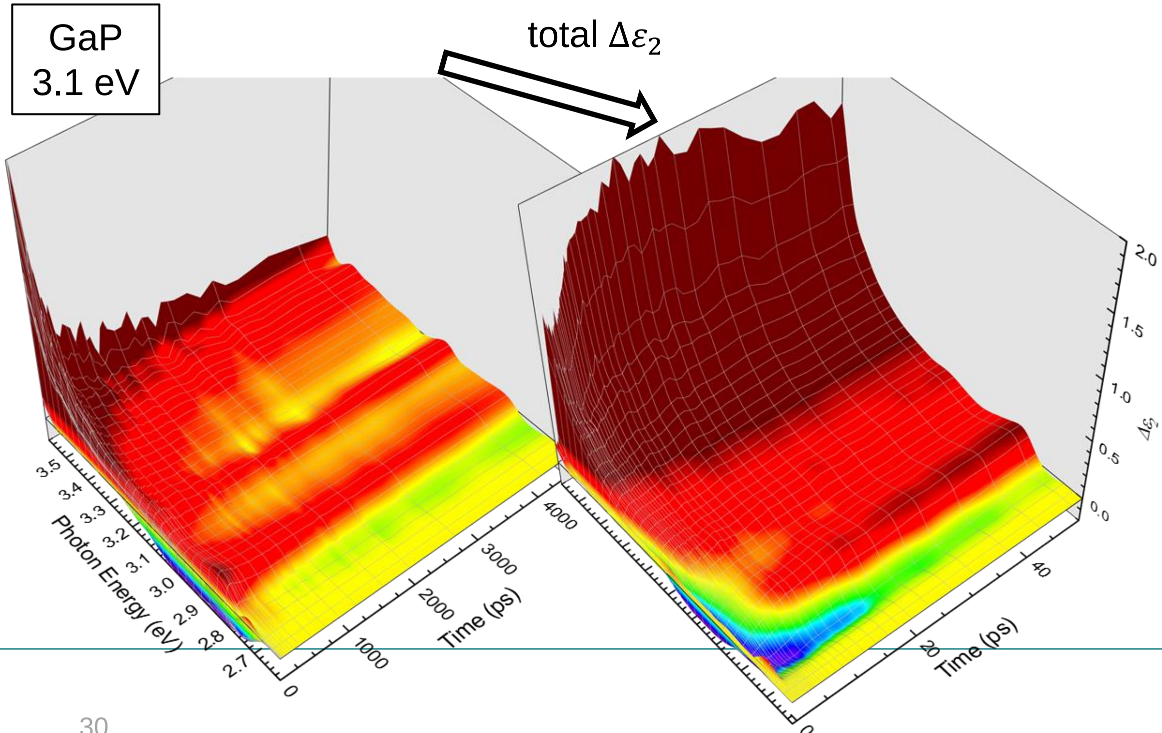
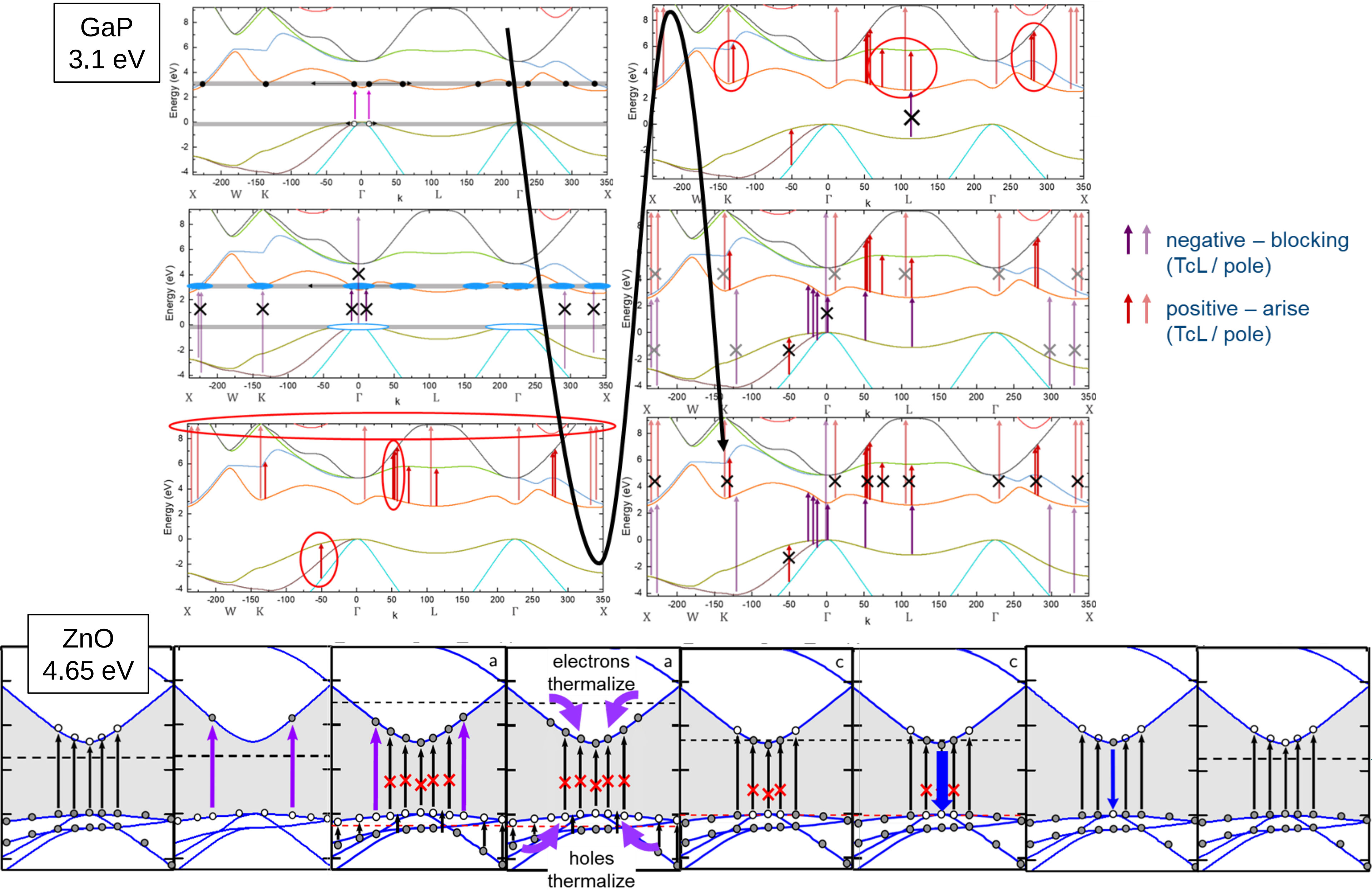


- → additional total oscillator strength due to intra-CB transitions peaks at around 20…40 ps
- → no complete recovery after 4 ns (next pump only after 1 ms)

The tale of electrons



The tale of electrons

exciting with high-intense laser beam: electrons and holes occupy formerly “empty” conduction (CB) and valence (VB) band states → hot charge carriers with temperatures up to several thousands of Kelvin

- → carriers spread within the entire Brillouin zone (BZ) to energetically matching bands
- → band gap renormalization (BGR) → redshift of transition energies between VB and CB
- → transitions between now occupied states are Pauli-blocked → reduction of absorption at the respective energies
- → excess electrons and holes can be excited by the probe light → arising of new intra VB and/or intra CB transitions → increase of absorption at the respective energies
- → when the material exhibits excitons: they are screened by the excited carriers, but may also form so-called Mahan excitons → energy redshift and absorption bleaching
- → collective free charge carrier oscillations (Drude)

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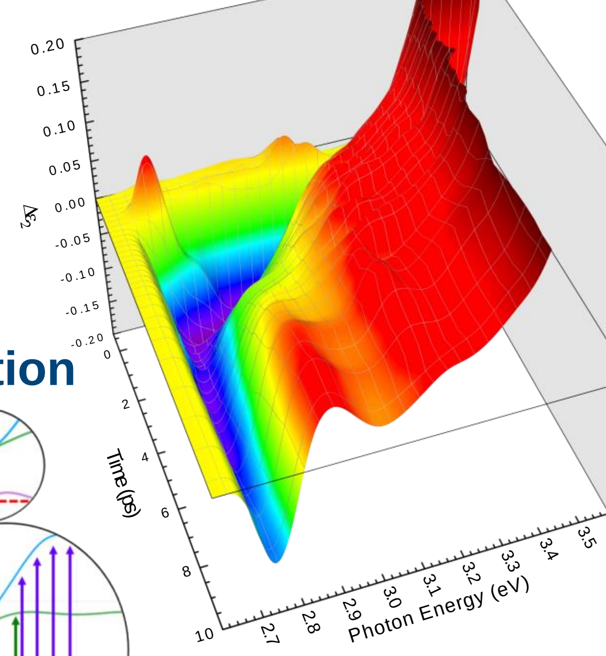
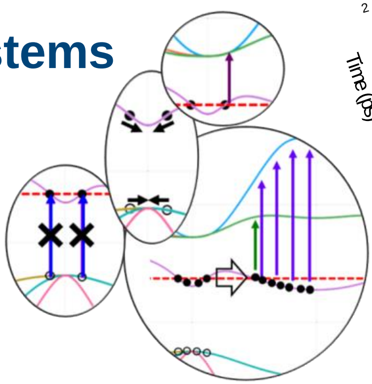
Summary

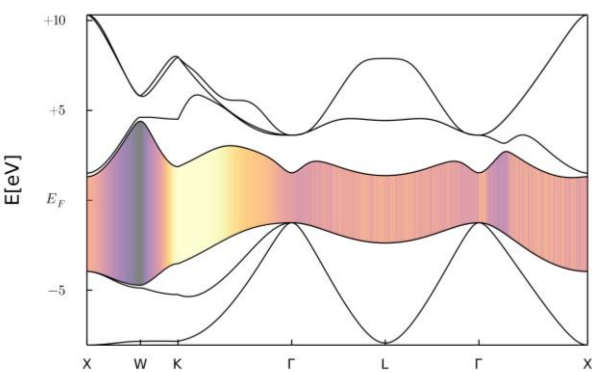
<u>Pump-probe fs-TSE</u>

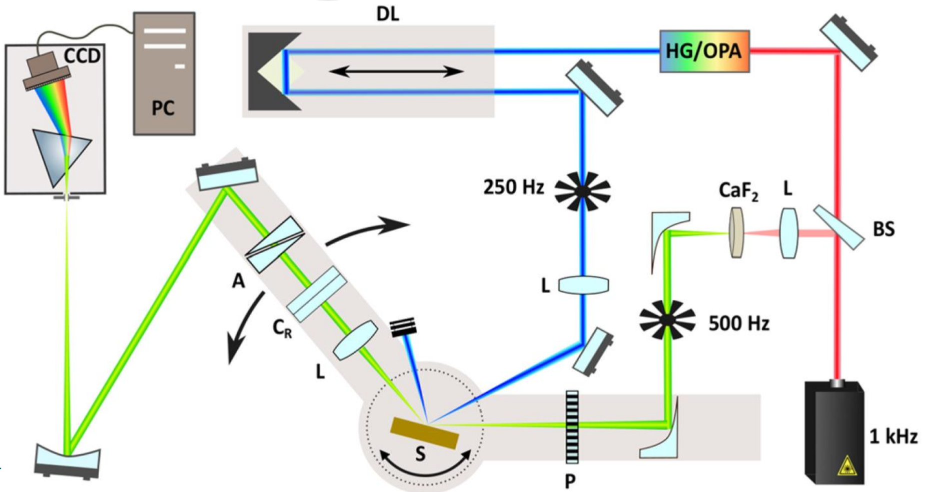
André, Carola, Marcus, Mateusz, Noah, Oliver, Shirly, Stefan, Steffen, Theo, Younes

- **method recently developed by us**
- **time evolution of the <u>full</u> complex dielectric function**
- **already applied to several material systems**
  - intra-band transitions (VB-VB and CB-CB)
  - hot-phonon scattering
  - ballistic carrier propagation
  - transient birefringence changes
  - coherent acoustic phonon oscillations
  - spectral weight transfer
  - …









Summary

Review of Scientific Instruments ARTICLE

*Steffen Richter, Mateusz Rebarz, Oliver Herrfurth, Shirly Espinoza, Rüdiger Schmidt-Grund, and Jakob Andreasson Rev. Sci. Instrum. **92**, 033104 (2021)*

Broadband femtosecond spectroscopic ellipsometry

Femtosecond-time-resolved imaging of the dielectric function of ZnO in the visible to near-IR spectral range

Cite as: Appl. Phys. Lett. **115**, 212103 (2019); https://doi.org/10.1063/1.5128069
Submitted: 16 September 2019 . Accepted: 05 November 2019 . Published Online: 19 November 2019

O. Herrfurth, T. Pflug, M. Olbrich, M. Grundmann, A. Horn, and R. Schmidt-Grund

New Journal of Physics
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IOP Institute of Physics

PAPER *New J. Phys.* **22** (2020) 083066

Ultrafast dynamics of hot charge carriers in an oxide semiconductor probed by femtosecond spectroscopic ellipsometry

Steffen Richter[1,2,7,8], Oliver Herrfurth[2,8], Shirly Espinoza[1], Mateusz Rebarz[1], Miroslav Kloz[1], Joshua A Leveillee[3], André Schleife[3], Stefan Zollner[4,5], Marius Grundmann[2], Jakob Andreasson[1] and Rüdiger Schmidt-Grund[2,6]

PHYSICAL REVIEW RESEARCH **3**, 013246 (2021)

Transient birefringence and dichroism in ZnO studied with fs-time-resolved spectroscopic ellipsometry

O. Herrfurth,[1,*] S. Richter,[2,3] M. Rebarz,[3] S. Espinoza,[3] J. Zúñiga-Pérez,[4] C. Deparis,[4] J. Leveillee,[5] A. Schleife, M. Grundmann,[1] J. Andreasson,[3] and R. Schmidt-Grund[7]

RESEARCH ARTICLE *Phys. Status Solidi RRL* **2022**, 16, 2200058
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Coherent Acoustic Phonon Oscillations and Transient Critical Point Parameters of Ge from Femtosecond Pump–Probe Ellipsometry

Carola Emminger, Shirly Espinoza, Steffen Richter, Mateusz Rebarz, Oliver Herrfurth, Martin Zahradník, Rüdiger Schmidt-Grund, Jakob Andreasson, and Stefan Zollner*

Transient dielectric functions of Ge, Si, and InP from femtosecond pump-probe ellipsometry

Cite as: Appl. Phys. Lett. **115**, 052105 (2019); https://doi.org/10.1063/1.5109927
Submitted: 14 May 2019 . Accepted: 01 July 2019 . Published Online: 01 August 2019

Shirly Espinoza, Steffen Richter, Mateusz Rebarz, Oliver Herrfurth, Rüdiger Schmidt-Grund, Jakob Andreasson, and Stefan Zollner

Thank You for Attention!

and thanks to all those who were also strongly involved in development of TSE method and physical understanding of processes: (alphabetically)

J. Andreasson, C. Emminger, S. Espinoza, D. Franta, O. Herrfurth, K. Hingerl, A. Horn, M. Kloz, J. Leveillee, M. Olbrich, T. Pflug, M. Rebarz, S. Richter, E. Runge, A. Schleife, Y. Slimi, N. Stiehm, C. Sturm, S. Zollner, M. Zahradník

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