B01

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Open-Minded

Scientific Questions

- What is key to understand charge carrier dynamics under non-equilibrium conditions?
- Are local relaxation and non-local / transport effects determined by identical or different microscopic processes?
- What is happening at interfaces?
- What is the influence on the excited charge carrier density *n*?
- How does the static electronic structure *E*(**k**) affect the non-equilibrium dynamics
- How does a particular optical or THz stimulus modify the dynamics and what are the respective processes activated?

Goals

Work package 1

- Analysis of scattering processes and propagation in non-equilibrium transport along the interface normal direction
- Separation of phononic and electronic contributions
- Analysis of transport in Bloch bands in comparison to transport by hopping in staples of 2D materials

Work package 2

- Time- and angle-resolved photoemission using THz field transients
- Analysis of in-plane charge carrier transport dynamics in real time
- Linear and non-linear response
- Manipulation of the electron dynamics by modifications of the static electronic structure using alkali surface doping

Expected Results

Cross Linking and Collaboration

Lattice dynamics and ultrafast diffraction

C01 — Sokolowski-Tinten | B04 C03 — Horn-von Hoegen

Samples of 2D materials, e.g., TaS_2, MoS_2

C05 — Schleberger, Sokolowski-Tinten, Wucher

Generation of THz field transients

B09 — Mittendorff

Complementary experiments on electron dynamics at interfaces

A07 — Eschenlohr, Ollefs | A06 — Campen, Hasselbrink, Tong | B06 — Meyer zu Heringdorf

Model calculations of propagating electronic excitations

A02 — König, Sothmann

Dynamics by *ab initio* methods

B02 — Gruner, Pentcheva | C02 — Kratzer

Modeling non-equilibrium dynamics

B03 B07 — König, Kratzer, Schützhold

Work Program

Work package 1
Time-resolved photoemission of charge carriers propagating in layered materials

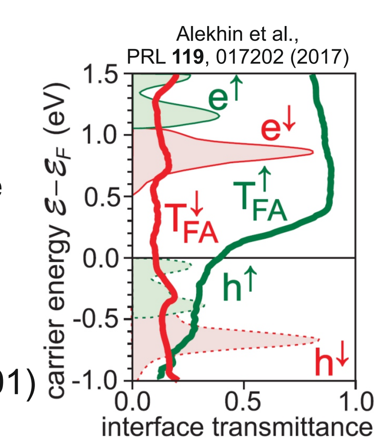
<u>Metallic heterostructures Au/Fe/MgO(001)</u>

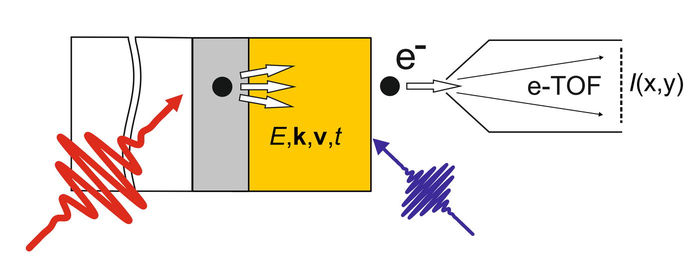
2020

- time-resolved linear photoemission using back side pumping to analyze transport effects in the vicinity of the Fermi energy (→ preliminary results)
- first steps regarding temperature dependent experiments employing back side pumping to identify phonon contributions (→ reduce sample vibration)

2021

- temperature dependent experiments (continutation)
- angle-resolved experiments for ballistic electron propagation to probe elastic scattering by a position sensitive anode, see figure for interface trans-mittance at Fe/Au(001)

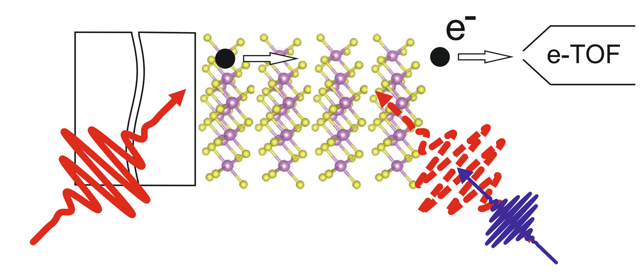


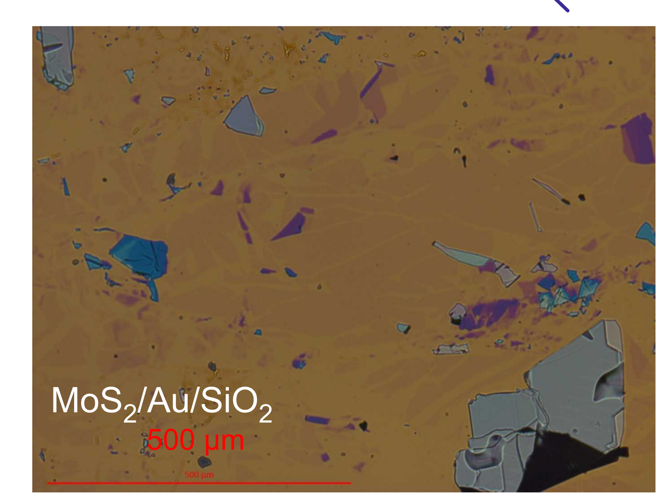


2022

<u>Stacks of two-dimensional materials MoS_2, $MoSe_2$, and TaS_2 upon back side pumping</u>

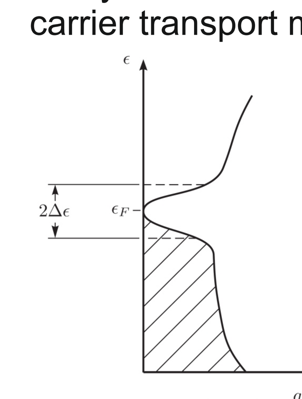
- Sample preparation by exfoliation on thin Au films on transparent substrates (collaboration with project C05)
- Temperature dependent experiments to analyze hopping propability





2023

- Analysis of thermally activated charge carrier transport mediated by hopping, e.g. electric conductivity σ for temperature dependent variable range hopping with a parabolic energy gap



$$\sigma = \sigma_0\left[-\left(\frac{\tilde{T}_0}{T}\right)^{1/2}\right]$$

Baranowski, Wiley (2006)

- Energy and temperature dependent transport of non-equilibrium carriers

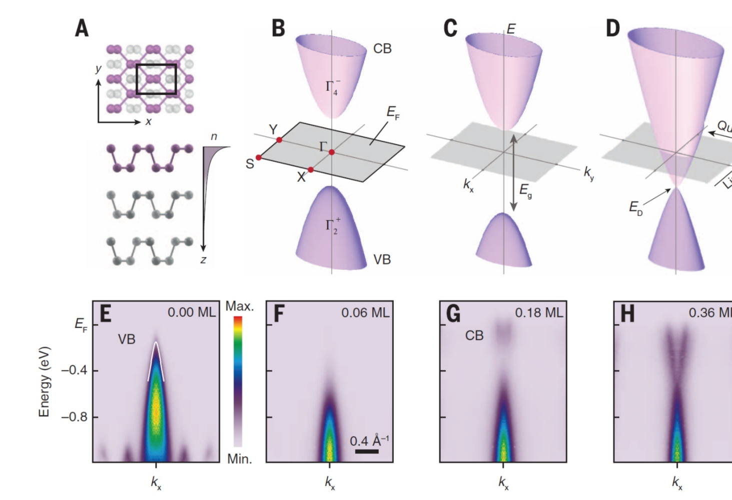
Work package 2
Real-time analysis of THz driven charge currents in surfaces of black phosphorus

2020

- Optimization of THz generation using photoconductive antenna emitter and Ti:sapphire RegA @ 100 kHz
→ gating of bias voltage to limit dc heating of antennae, reach peak field >50 kV/cm
→ funding application for gated pulse generator and voltage amplifier
- Laser ARPES of black phosphorus

2021

- Alkali surface doping of black phosphorus Band inversion by giant Stark effect

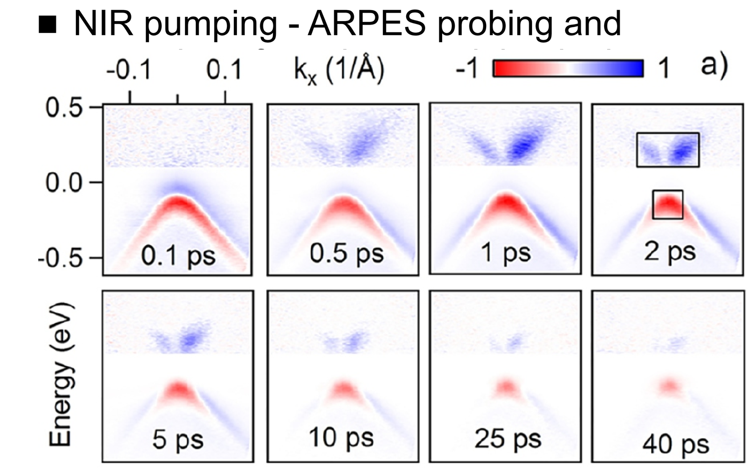


Kim et al., Science **349**, 723 (2015)

- NIR pumping - ARPES probing of black phosphorus for different doping

2022

- Alkali surface doping of black phosphorus
- NIR pumping - ARPES probing and



Chen et al., Nano Letters **19**, 488 (2019)

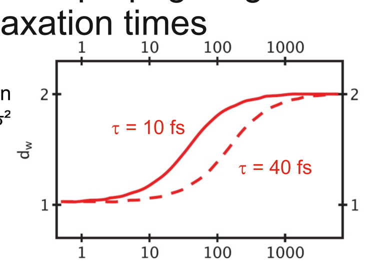
2023

- THz pump ARPES probe experiments
- Analysis of electron dynamics near E_F for variation of *E*(**k**) by alkali doping along armchair and zigzag directions
- Manipulation of non-equilibrium electron dynamics by alkali doping
- Linear to non-linear crossover in response to THz driving the electron system

Preliminary Work

- Results obtained by 2PPE presents **superdiffusive propagation** behaviour of hot e's due to fast scattering processes at energies $E-E_F$= 0.6 - 2.0 eV
- **Idea:** exciting hot e's close to Fermi edge E_F to see mainly ballistic propagating hot e's due to longer relaxation times
  - time dependent anomalous diffusion constant d_w governed by variance σ^2 of the displacement of a single particle distribution:

$$d_w(t) = \frac{2}{t}\frac{\sigma^2}{d\sigma^2/dt}$$

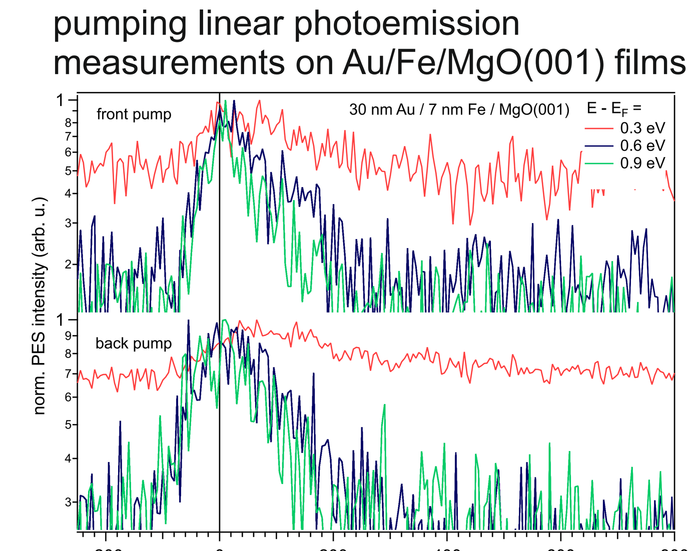


Battiato et al., PRB **86**, 024404 (2012)

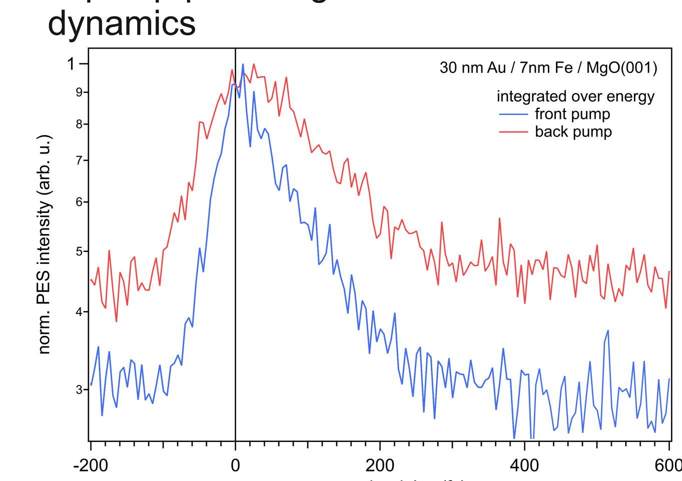
  - d_w= 1: ballistic propagation
  - 1≤ d_w< 2: superdiffusive regime
  - d_w= 2: diffusive regime

For short times, t << τ, motion is mainly ballistic and the particle experiences approximately no scatterings. On times,t >> τ, many scatterings appear and the overall motion is well approximated by standard diffusion.

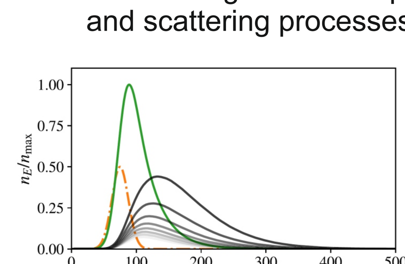
- **First** results with front and back side pumping linear photoemission measurements on Au/Fe/MgO(001) films



- Observation of differences in population dynamics at a given energy $E-E_F$ hardly to see
- transport effects less obvious due to time independent signal background
- **But**: integration over an energy range of $E-E_F$= 0 - 1.55 eV reveals a clear contrast in pump-probe signal and relaxation dynamics

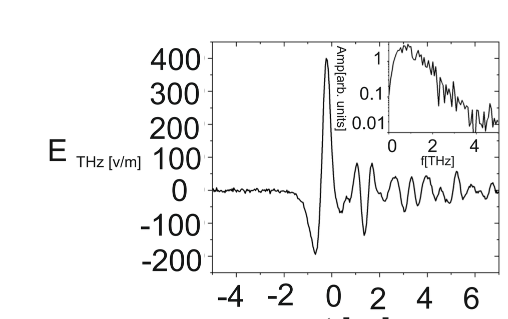


- back pump: small delayed shift in peak intensity and broadening in population dynamics → signature for transport?
  - Nenno et al.,PRB **98**, 224416 (2018): delayed shift and broadening of particle densities at lower energies due to spatiotemporal transport and scattering processes

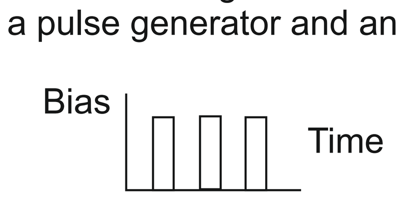


Simulation after excitation with a Gaussian pulse: Density n_E over time for carriers of discrete energies (black line, darker for lower energies) and at the injection energy (green line).

- Generation of THz pulse to investigate realtime charge transport using a THz-pump photoemission-proBiasbe experiment by using a photoconductive (PC) antenna. This part is implemented by collaboration with project B09.
- The generated THz field with 400 V/m amplitude with the near-IR fiber laser (780 nm, 80 MHz) of energy per pulse of 1.4 nJ:



- Next steps:
1- Increasing energy per pulse to 4 uJ by using RegA 9040 pulsed laser (800 nm, 250 kHz).
2- Increasing bias voltage.
- Challenges and solutions:
1- Excessive heating of the PC (solution: using a pulse generator and an amplifier).



- 2- Increasing the size of the PC emitters (solution: optimization of the THz pulse by using different PCs).