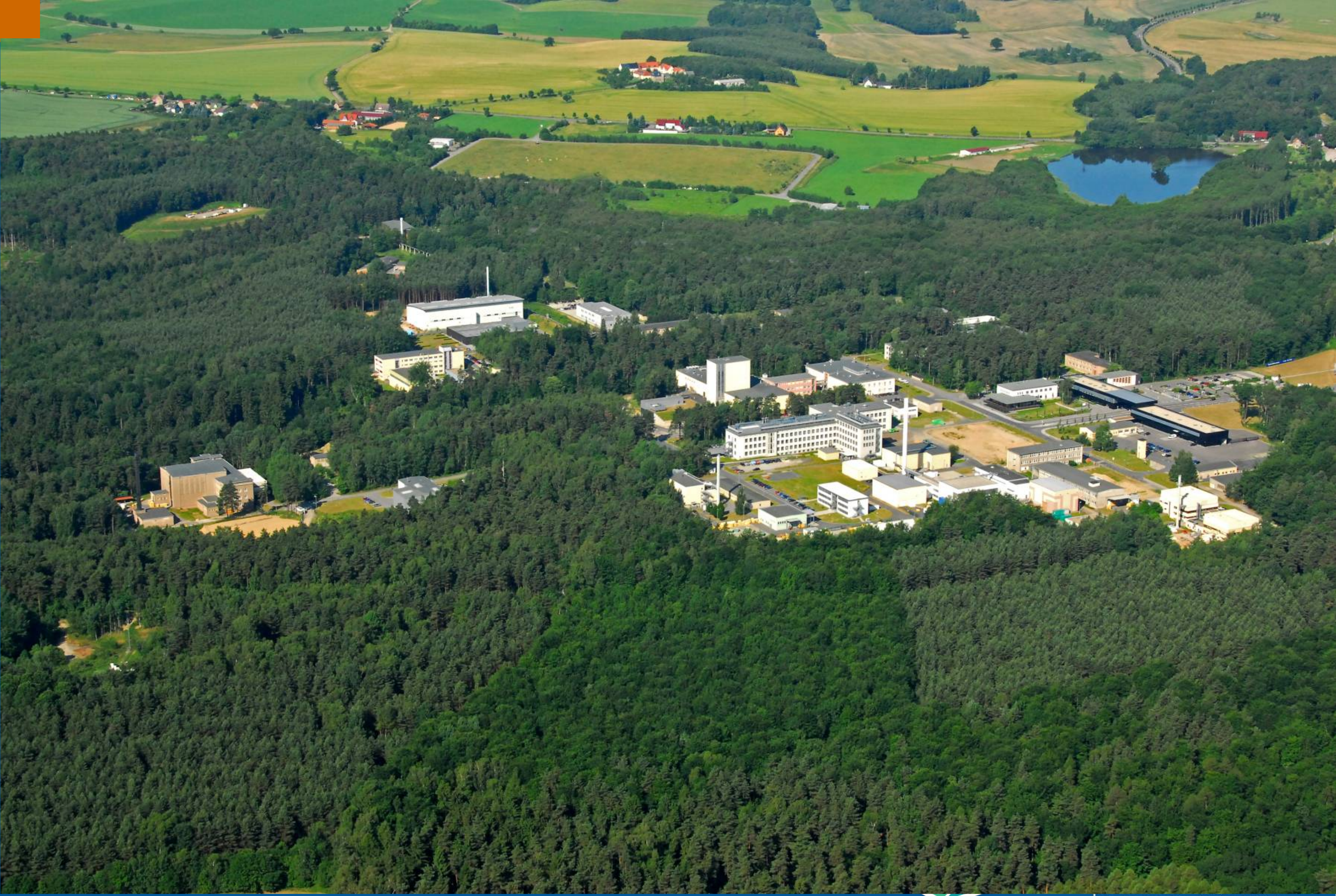
Synchrotron and Free Electron Laser Radiation: Generation and Application (SFR-2016), Budker INP, Novosibirsk, 4-7 July 2016

Semiconductor spectroscopy with infrared and THz free-electron lasers

H. Schneider

In collaboration with: **M. Fehrenbacher, D. Stephan, J. Bhattacharyya, S. Zybell, S. Winnerl, M. Mittendorff, J. König-Otto, M. Helm**

Institute of Ion-Beam Physics and Materials Research
Helmholtz-Zentrum Dresden-Rossendorf
Dresden, Germany



ROSSENDORF

Overview

- **Research infrastructure & Motivation**
- **Graphene**
  pump-probe spectroscopy of Landau levels
- **Exciton dynamics in GaAs quantum wells (QW)**
  intra-exciton transitions
- **Transient photoluminescence in *single* InGaAs quantum dots (QD)**
  inter-sublevel dynamics
- **Superlens**
  infrared/THz near-field microscopy
- **Conclusion**

The Helmholtz-Zentrum Dresden Rossendorf

Ca. **1000 employees** from **45 nations**

Research Sites in **Dresden**, Freiberg, Leipzig, Grenoble

Institutes | **Research programs** | **Research facilities**

Institutes

- **Ion Beam Physics and Materials Research**
- **Dresden High Magnetic Field Laboratory**
- **Radiation Physics**
- **Resource Ecology**
- **Radiopharmaceutical Cancer Research**
- **Radiooncology**
- **Fluid Dynamics**
- **Helmholtz Institute Freiberg for Resource Technology**

Research programs

- **Matter**
- **Energy**
- **Health**

Research facilities

- **Radiation Source ELBE**
- **High Magnetic Field Laboratory**
- **Ion Beam Centre**
- **Rossendorf Beamline**
- **PET Centre**
- **TOPFLOW Facility**

red: solid state physics

DRESDEN concept HZDR

Elbe in Dresden

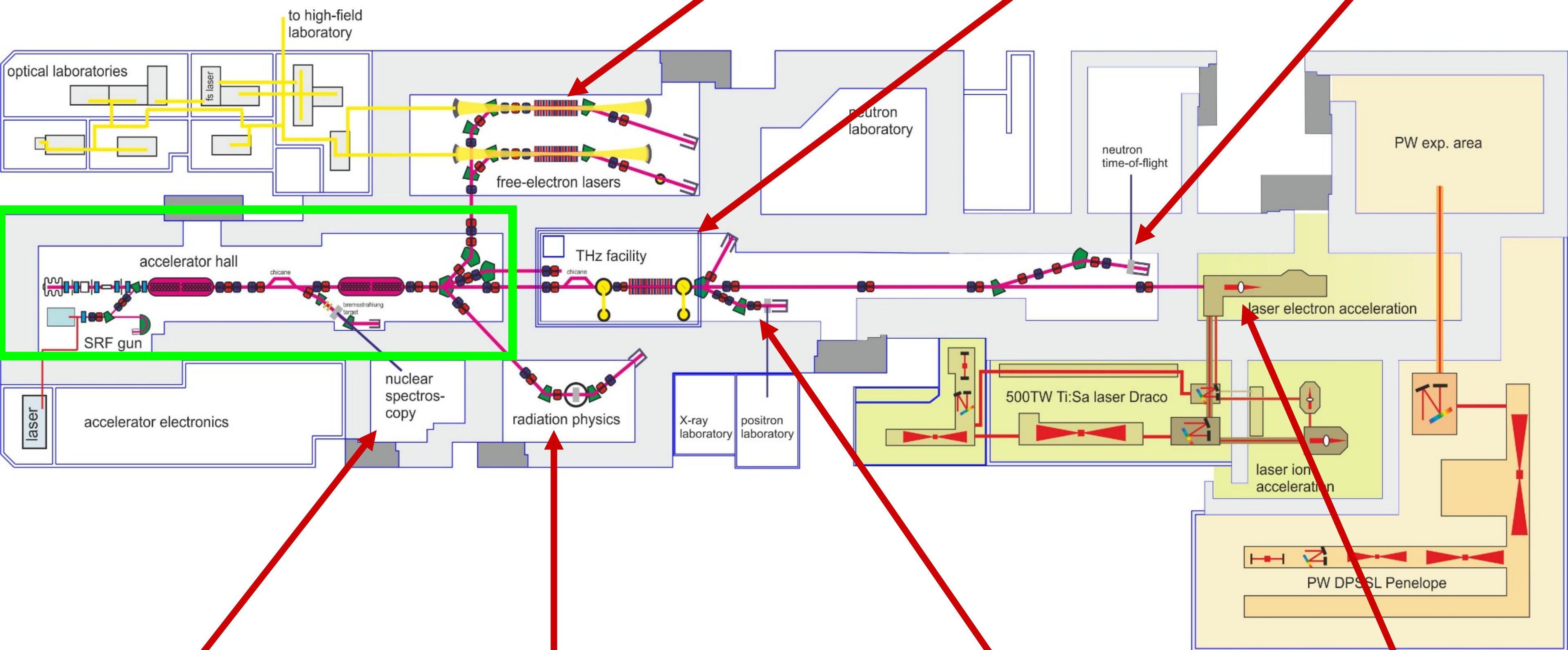
ELBE = **E**lectron **L**inear accelerator with high **B**rilliance and **L**ow **E**mittance

1 mA, 40 MeV CW electron accelerator

free-electron laser
4 – 250 µm

THz radiation
100 µm – 3 mm

neutron time of flight
0 – 10 MeV



Bremsstrahlung
0 – 17 MeV

ELBE electrons/ monochromatic X-rays
30 – 34MeV/10 – 100 keV

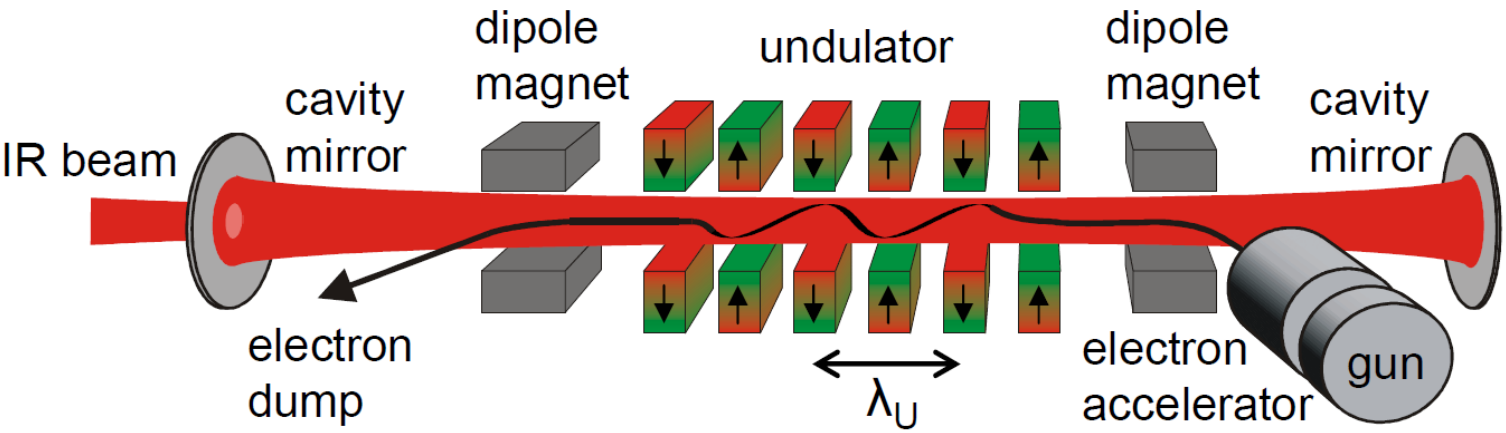
pulsed, mono-energetic positrons 0.2 – 30 keV

electron laser interaction

DRESDEN concept HZDR

Free-electron laser FELBE

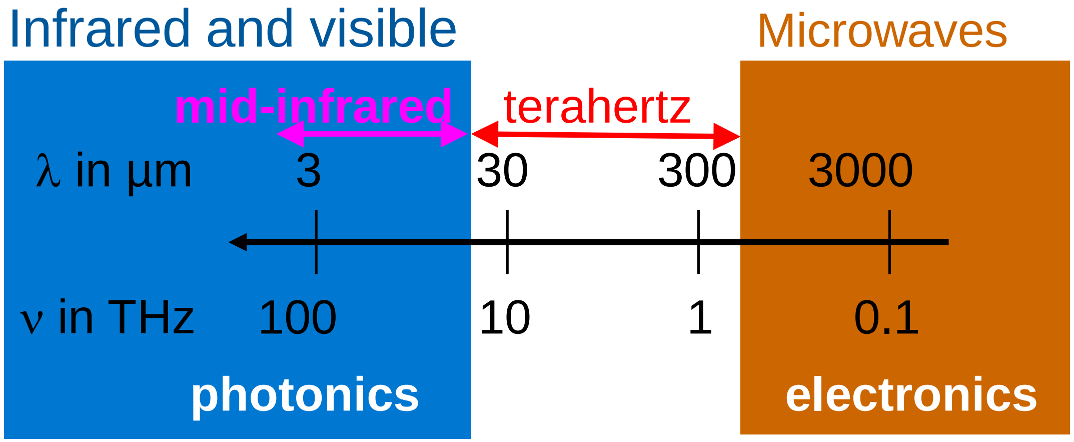
Tuning range	1 – 80 THz (5 – 330 meV)
Average power	1 – 20 W
Peak power	0.1 – 1 MW
Pulse energy	0.1 – 2 µJ
Pulse width	1 – 25 ps
Spectral width	0.5 – 2 %
Repetition rate	13 MHz



→ FELBE and NovoFEL = Only continuously pulsing FELs in Asia and Europe

Terahertz radiation

1 THz = 10^{12} Hz = 1 / (1 ps) → $\hbar\omega$ = 4.1 meV, λ = 300 µm



- Frequency **too high for electronics** and **too low for photonics**
- **Lack of efficient devices** for generation and detection

In the THz regime:

Vibrations and rotational transitions in molecules

Frequency of elementary and collective excitations in solids

- Phonons, polarons, plasmons, magnons, Cooper pairs
- Electronic excitations in semiconductors & nanostructures

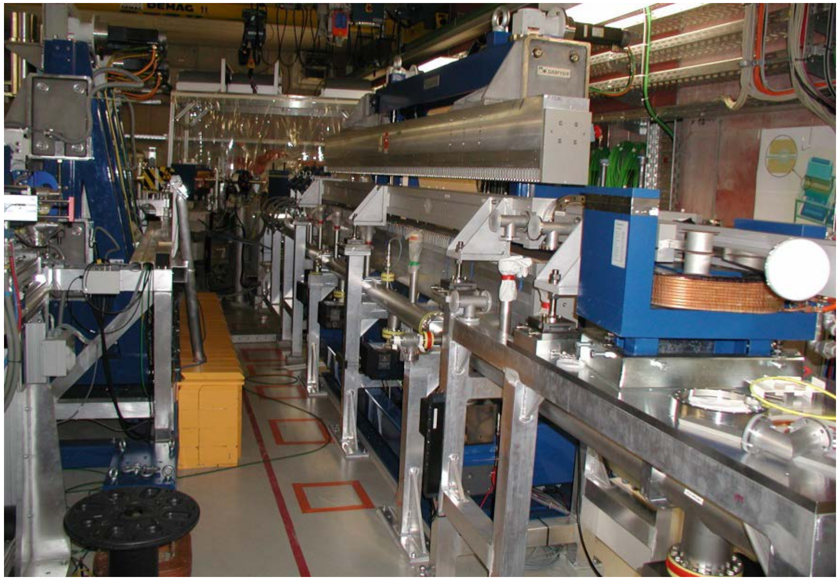
Intrinsic time constants in solids

- Electronic scattering times, tunneling time constants
- Coherence, decay of optical polarization into population
- Phase transitions in complex materials, metal-insulator transition

Research Infrastructure

Mid-IR/THz free-electron laser

- 1 – 80 THz, $\Delta\lambda/\lambda$ ~ 1%
- 0.5 - 30 ps, 0.1 - 3 µJ
- tens of Watts average power
- 13 MHz continuous pulse train
- external user access



Tabletop laser systems

- fs/ps oscillators
- µJ and mJ amplifier systems
- conversion to mid-IR & THz
- streak camera system

Tabletop lasers synchronized to FEL

German-Russian collaborative project "InTerFEL"

10/2014 – 09/2017

GEFÖRDERT VOM

Bundesministerium für Bildung und Forschung

Time-resolved infrared and terahertz spectroscopy of carrier dynamics in semiconductors with free electron lasers

- Institute for Physics of Microstructures, Nizhny Novgorod (IPM RAS)
- **Budker Institute of Nuclear Physics, Novosibirsk (NovoFEL)**
- Karpov Institute of Physical Chemistry, Obninsk (Karpov IPC)

- Humboldt Universität Berlin (HUB)
- Helmholtz-Zentrum Dresden-Rossendorf (HZDR)
- Leibniz Institut für Kristallzüchtung, Berlin (IKZ)

IPM RAS

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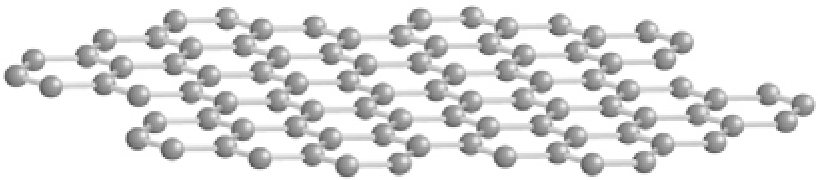
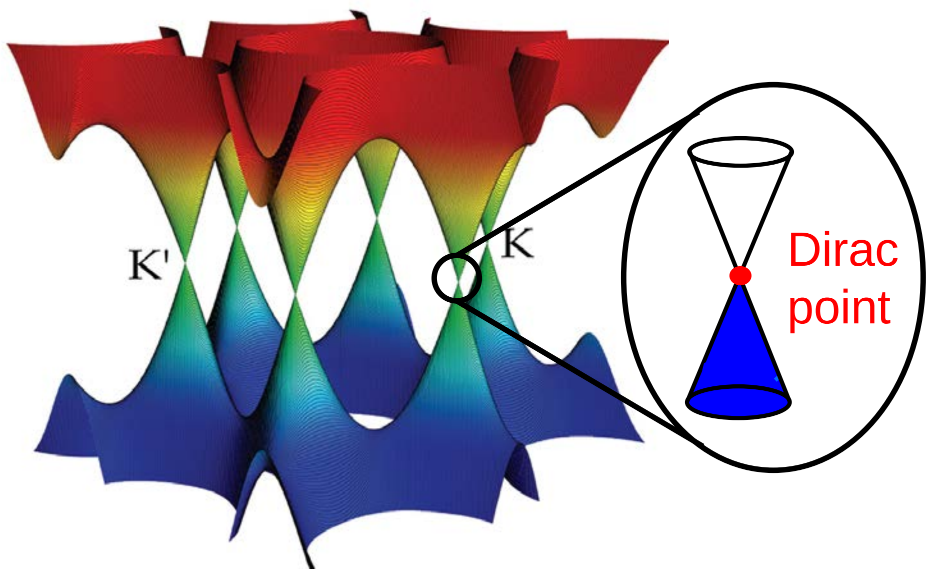
Graphene

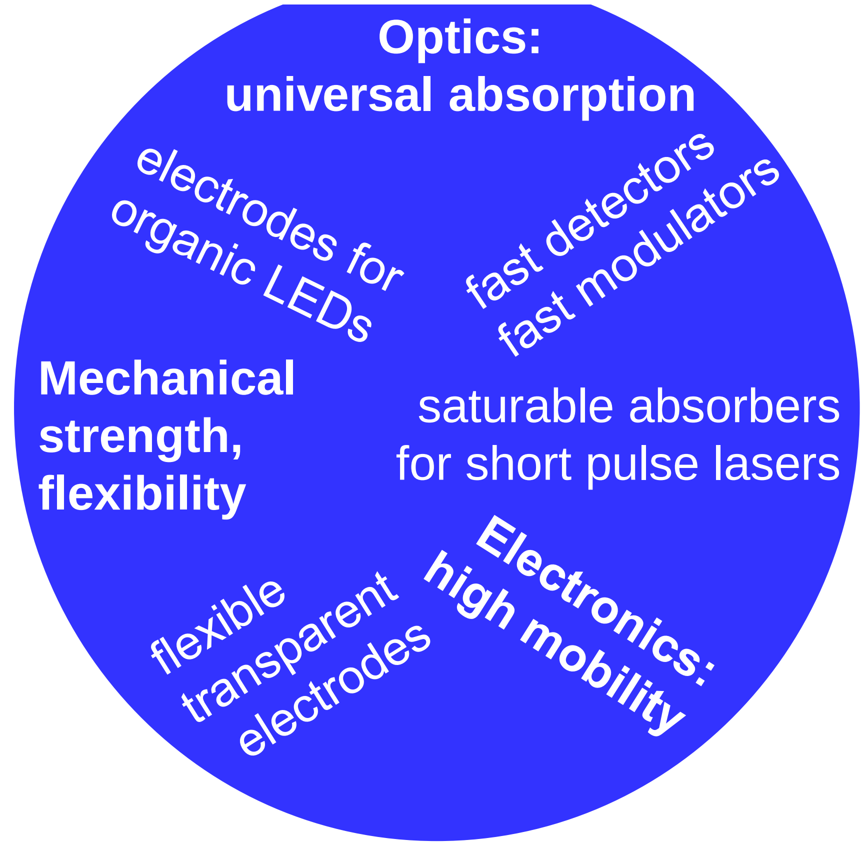
Graphene = mono-atomic layer of
sp^2 bonded carbon atoms
hexagonal lattice





Band structure with Dirac points

Many potential applications



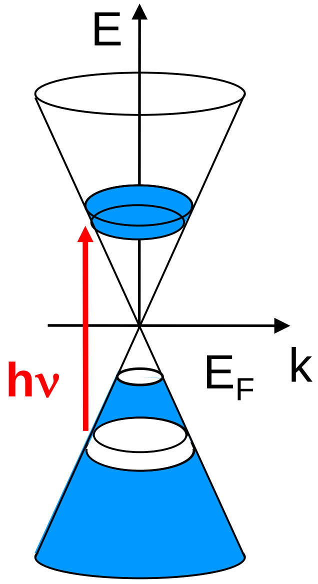
Images from M.I. Katsnelson, Materials Today **10**, 20 (2007).

Carrier dynamics in graphene

Short THz pulses

- Excitation close to the Dirac point

  i.e., low photon energies, 5 - 300 meV
- Photon energy comparable with
  optical phonon energy, Fermi energy



Our motivation

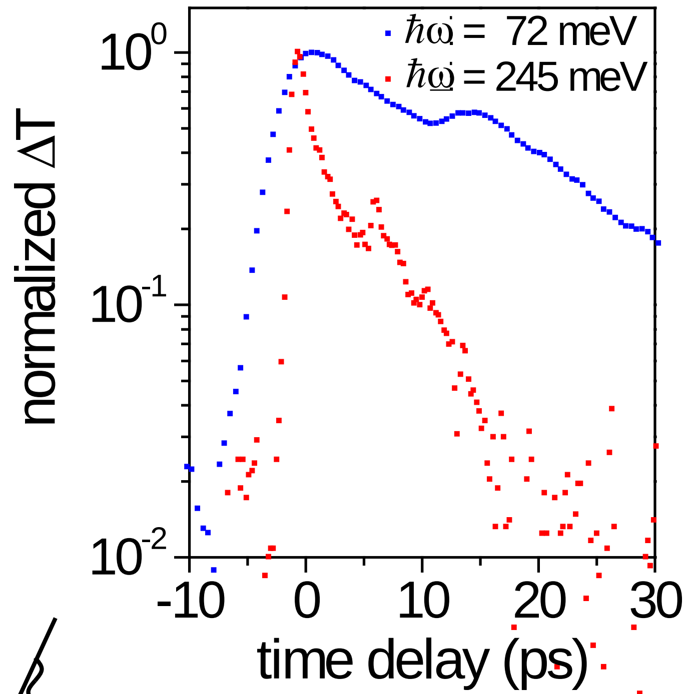
- Basic interest

  - Understanding of carrier dynamics, electron-phonon interaction,…
- Applications in optoelectronics

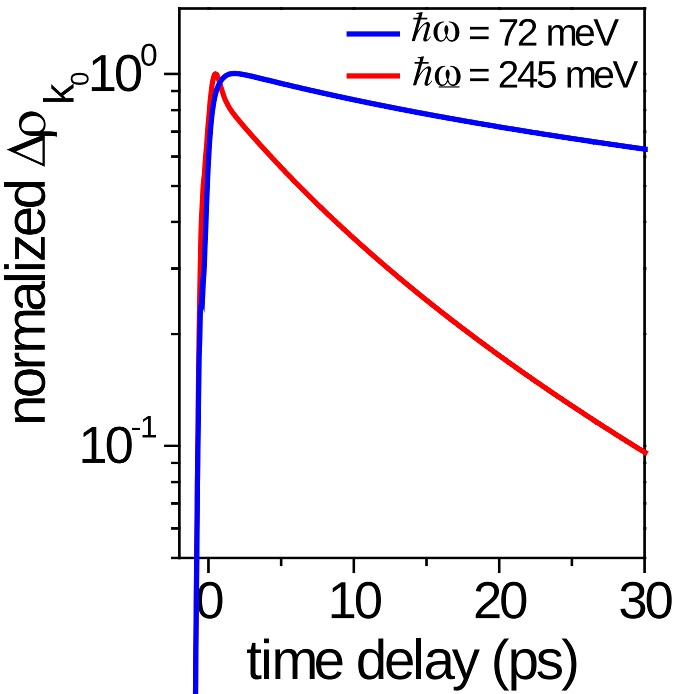
  - detectors, modulators, frequency mixers, saturable absorbers,…
  - from VIS/NIR to THz

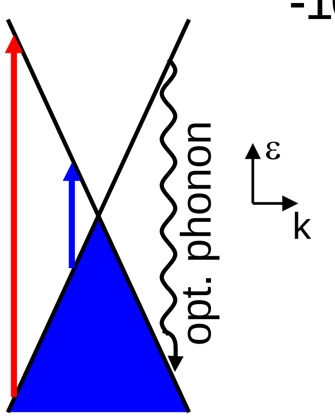
Pumping above and below the optical phonon energy

Experiment at FELBE



Microscopic theory (TU Berlin)



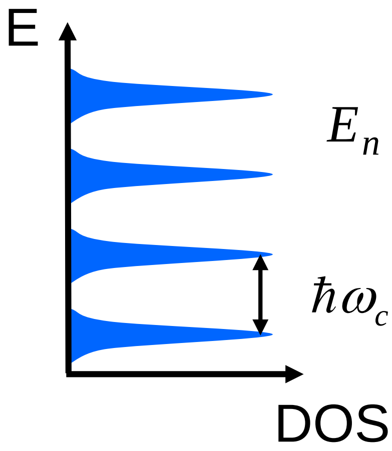


Slower relaxation for photon energies below the optical phonon energy (~200 meV)

S. Winnerl et al., Phys. Rev. Lett., **107**, 237401 (2011)

Landau quantization basics: from 2D to 0D

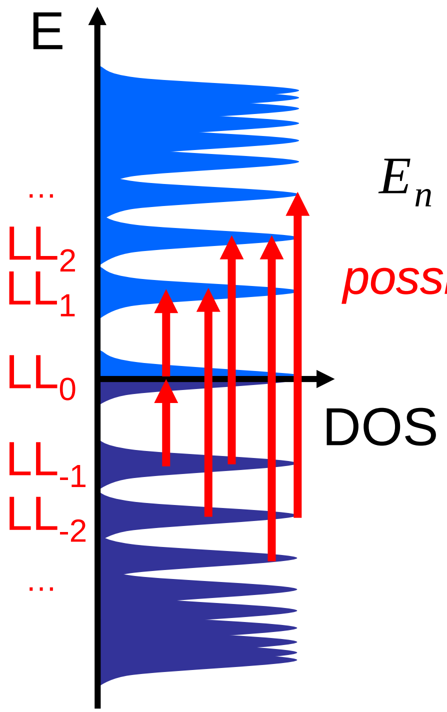
2DEG with parabolic dispersion



$$E_n = \left(n + \tfrac{1}{2}\right)\hbar\omega_c$$

$$\hbar\omega_c = \frac{\hbar e B}{m_{eff}}$$

Graphene



$$E_n = \tilde{c}\sqrt{2e\hbar B n}$$

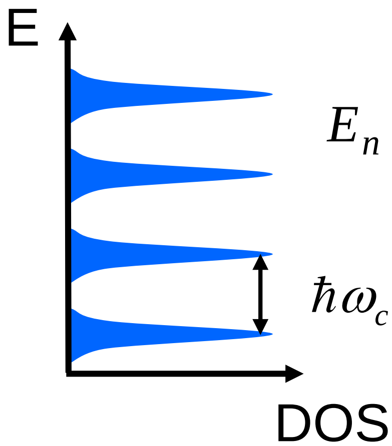
possible optical transitions

- Graphene has non-equidistant Landau levels

Mitglied der Helmholtz-Gemeinschaft

Landau quantization basics: from 2D to 0D

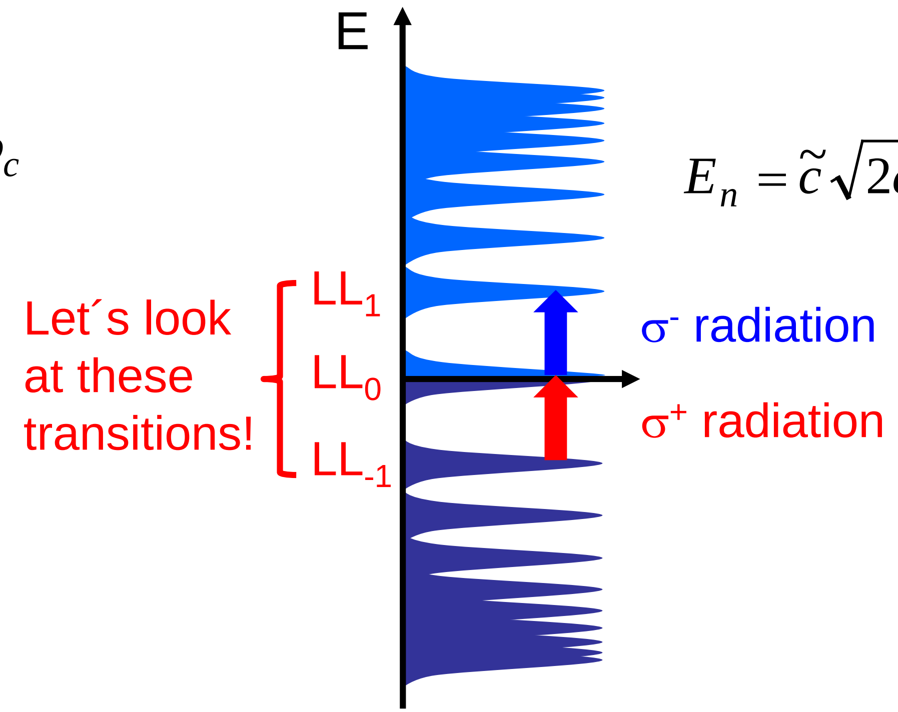
2DEG with parabolic dispersion



$$E_n = \left(n + \tfrac{1}{2}\right)\hbar\omega_c$$

$$\hbar\omega_c = \frac{\hbar eB}{m_{eff}}$$

Graphene



$$E_n = \tilde{c}\sqrt{2e\hbar Bn}$$

- Graphene has non-equidistant Landau levels
- Seperate excitation of $LL_{-1} \rightarrow LL_0$ and $LL_0 \rightarrow LL_1$ transitions possible by circularly polarized radiation

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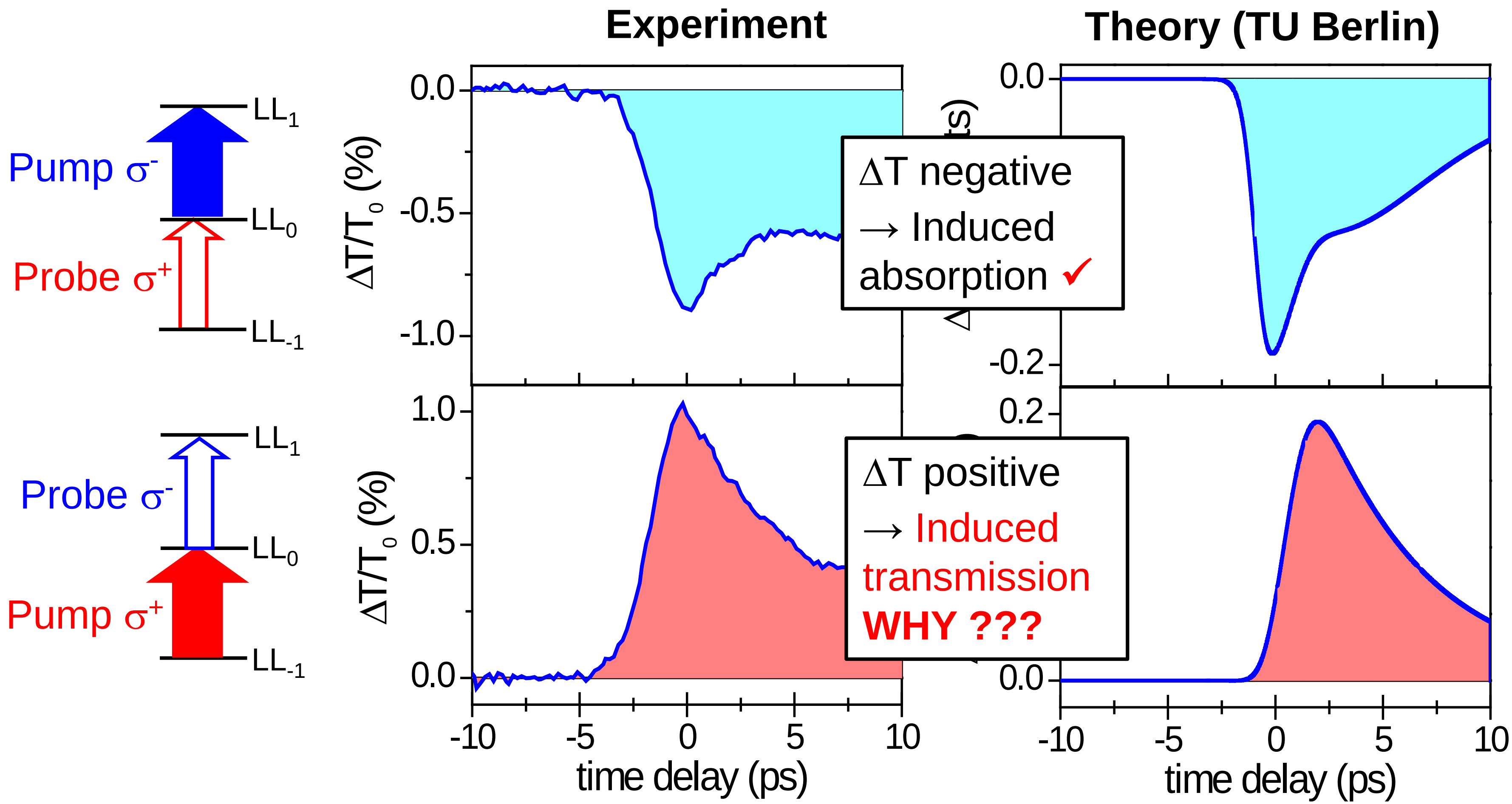
Pumping and probing with opposite polarization



Experimental conditions:

$\hbar\omega$ = 75 meV, B = 4.2 T

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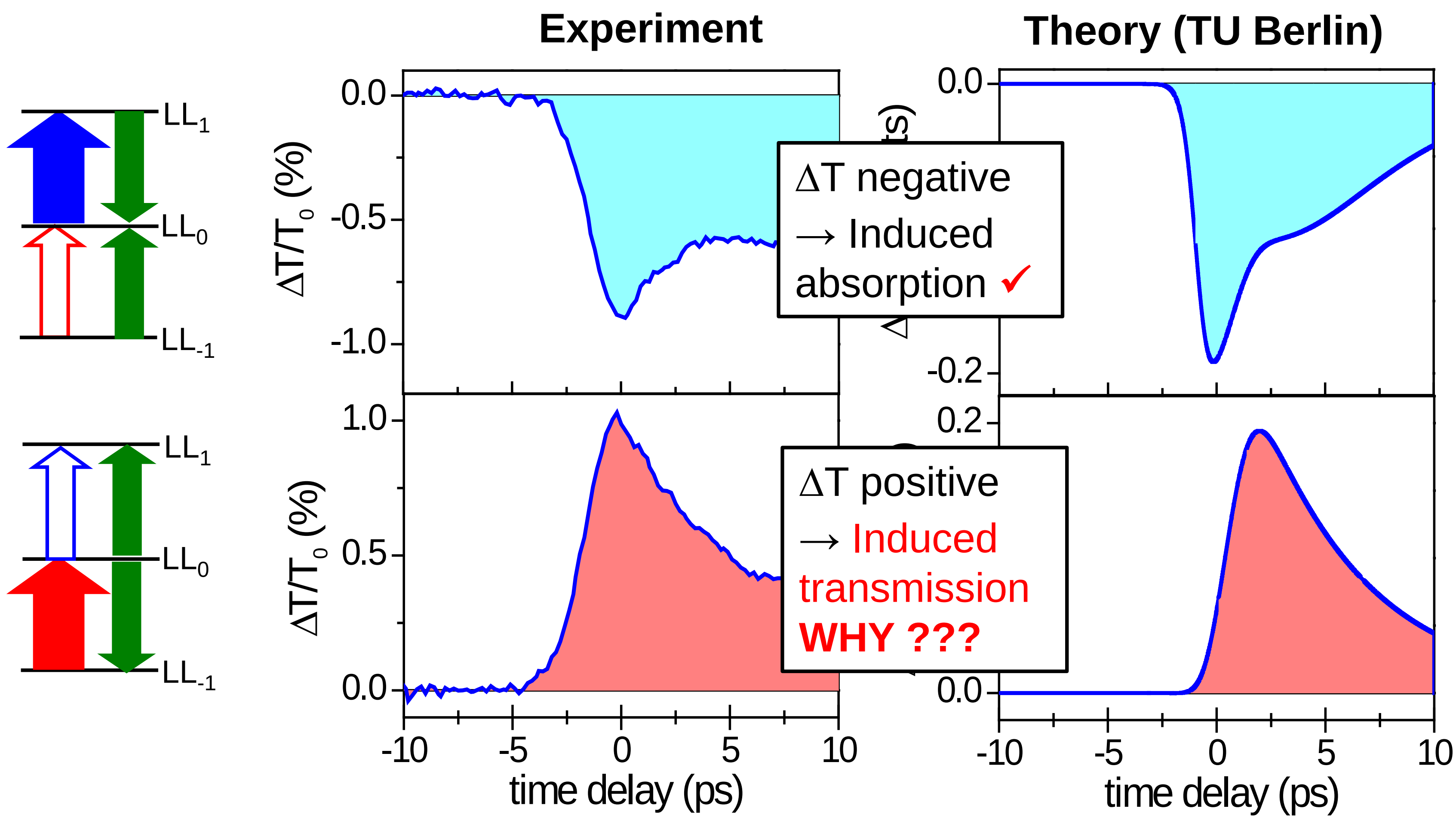
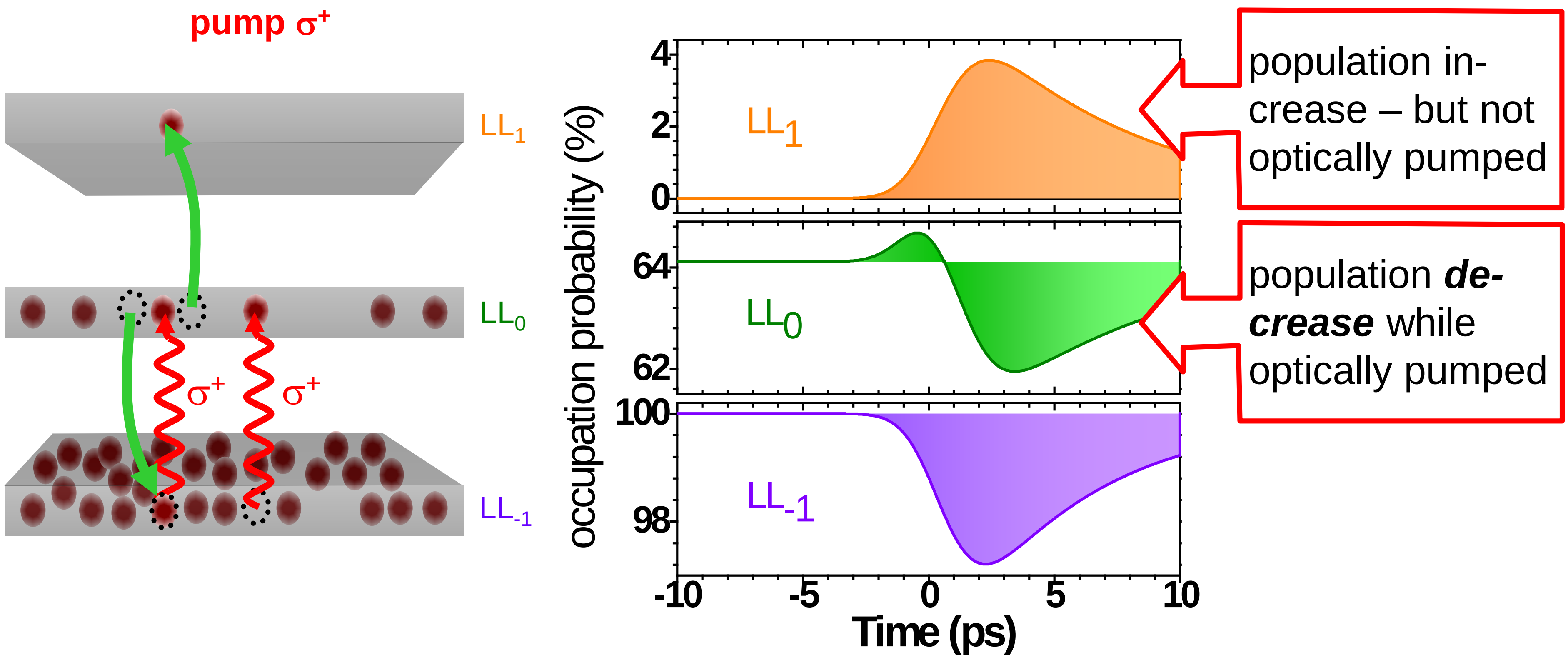
Pumping and probing with opposite polarization



- **Carrier redistribution by Auger scattering**
- **Small doping breaks symmetry between σ^+ and σ^- excitation**

Landau level occupation



- **Carrier redistribution by Auger scattering**

M. Mittendorff et al., Nature Phys. ***11****, 75 (2015).*

Mitglied der Helmholtz-Gemeinschaft

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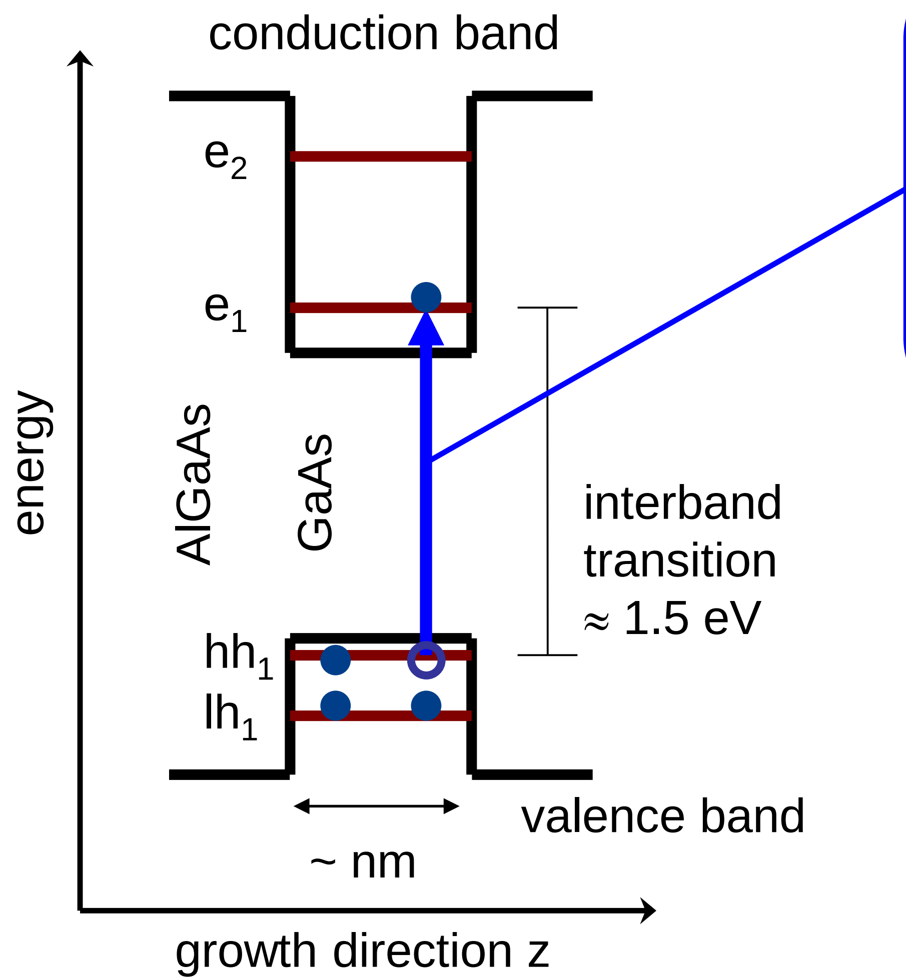
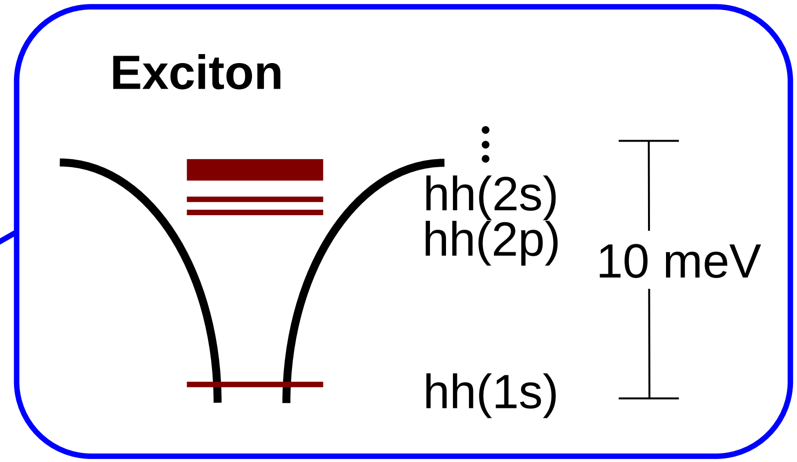
Excitons in semiconductor quantum wells

Quantum Well





- Coulomb attraction between electron and hole
- Bound hydrogen-atom-like state $E_{Rydberg} = \frac{m_e q^4}{8\varepsilon_r^2 \varepsilon_0^2 h^2}$
- Exciton binding energy in the THz range, 1 THz = 4.1 meV

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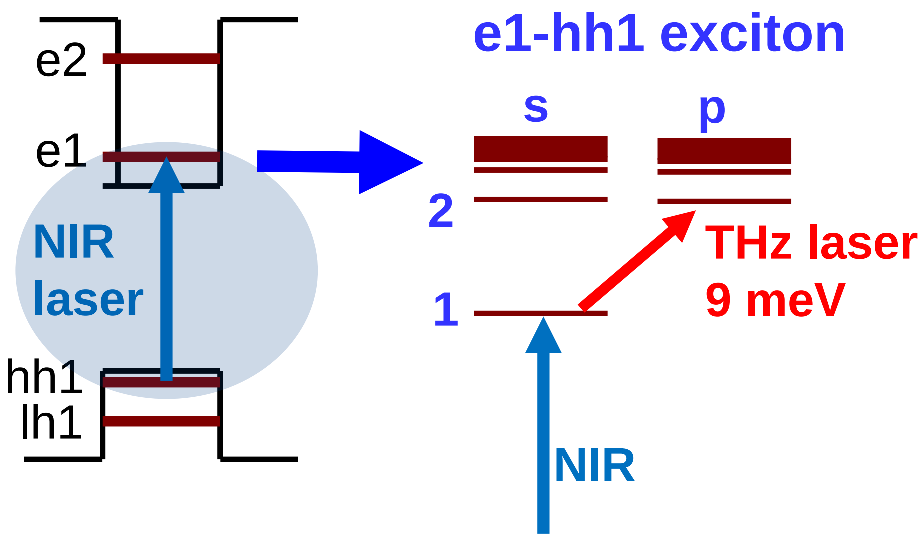
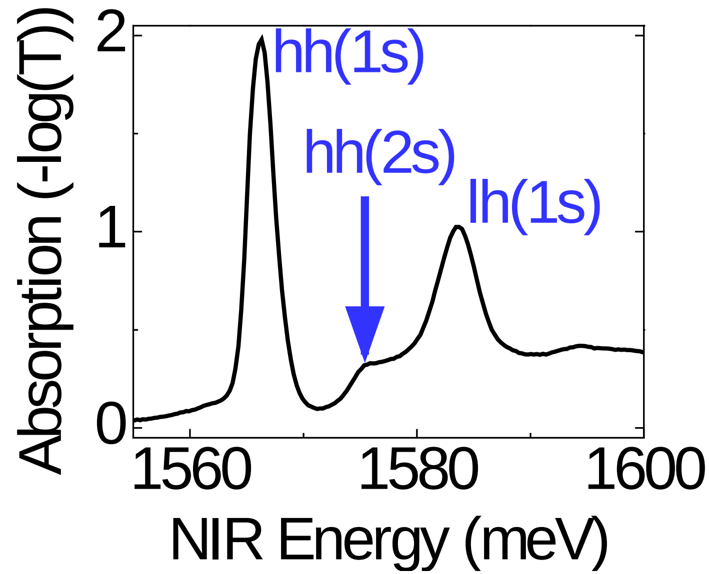
Harald Schneider | Institute of Ion-Beam Physics and Materials Research | www.hzdr.de

Exciting excitons …



8.2 nm GaAs QWs
$Al_{0.34}Ga_{0.66}As$ barriers

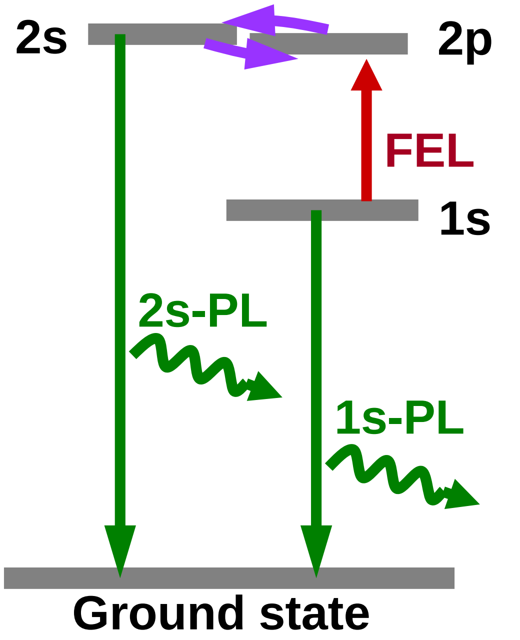


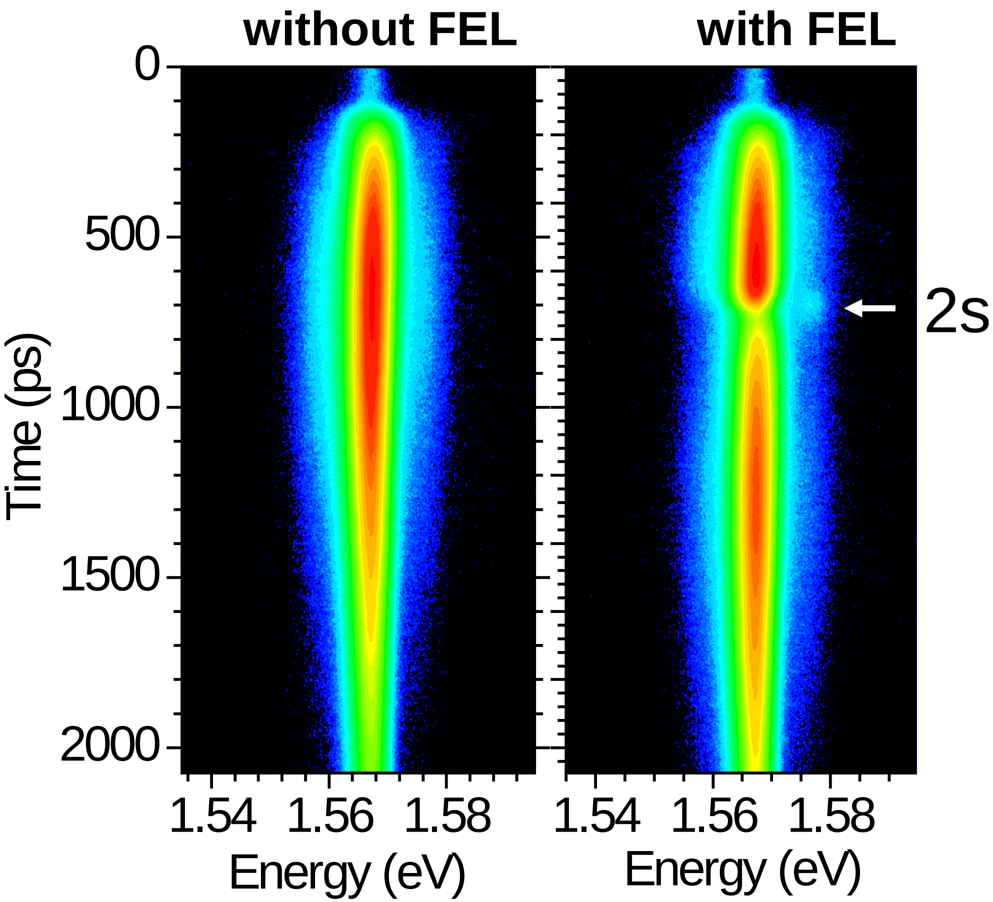
- Generate excitons with **NIR laser**
- Drive **intra-excitonic resonance** with **THz free-electron laser**

  → 1s-2p transition

M. Wagner et al., Phys.Stat.Sol.(b) ***248****, 859 (2011)*

THz-induced photoluminescence of 2s-exciton

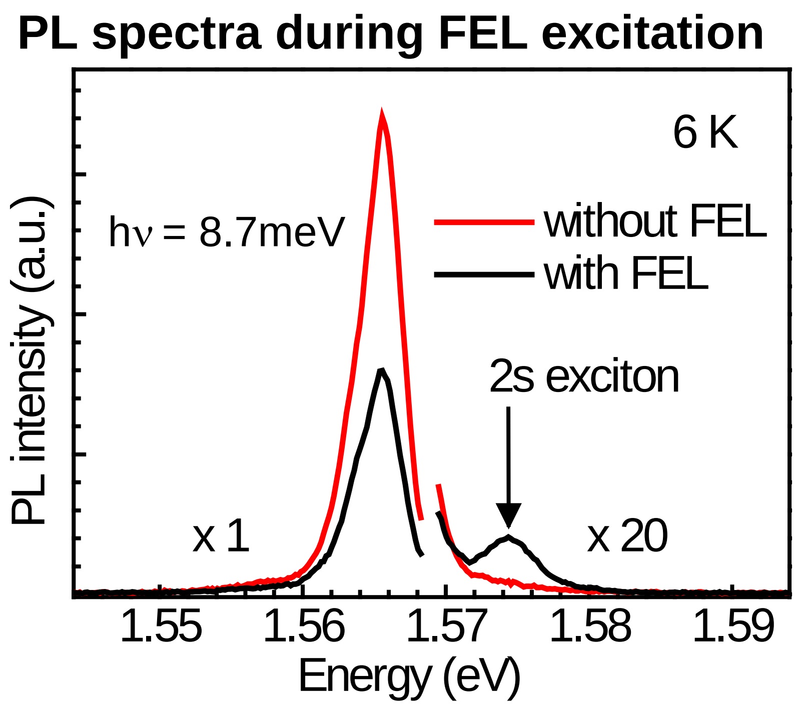


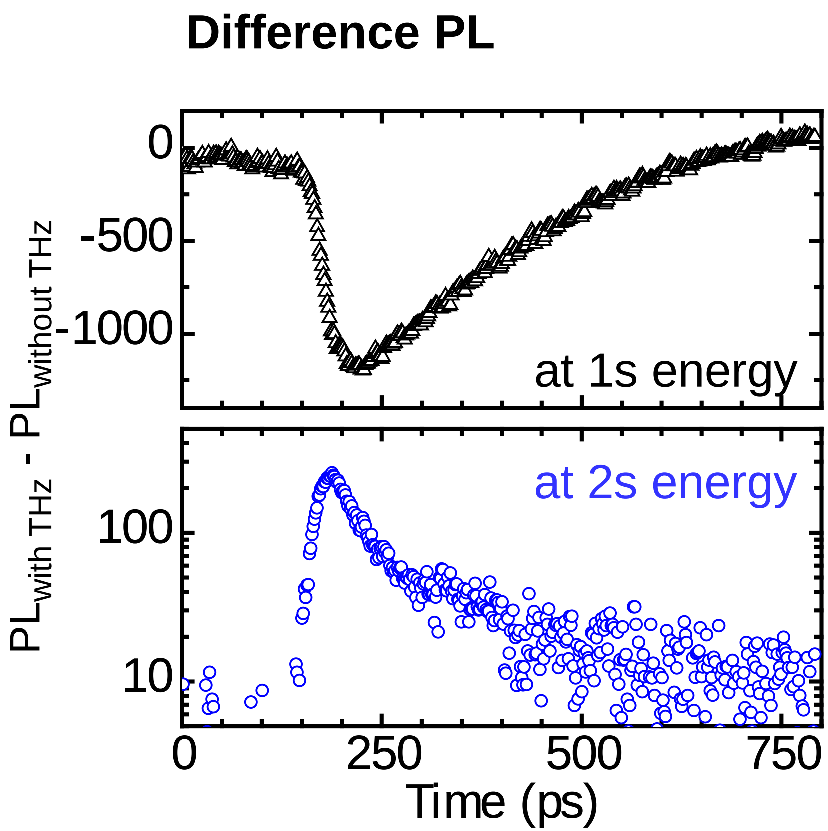


- Excitation from 1s into 2p exciton state by FEL
- Photoluminescence of 2s state → evidence of fast 2p-2s population transfer

W. D. Rice et al., Phys. Rev. Lett. ***110****, 137404 (2013).*
S. Zybell et al., Appl. Phys. Lett. ***105****, 201109 (2014)*

Dynamics





Dynamics of 2s exciton

Rapid formation, ~ 10 ps

→ evidence of extremely fast 2p-2s transfer

Fast initial decay, ~ 50 ps

→ scattering into lower-energy states

W. D. Rice et al., Phys. Rev. Lett. ***110****, 137404 (2013).*
S. Zybell et al., Appl. Phys. Lett. ***105****, 201109 (2014)*

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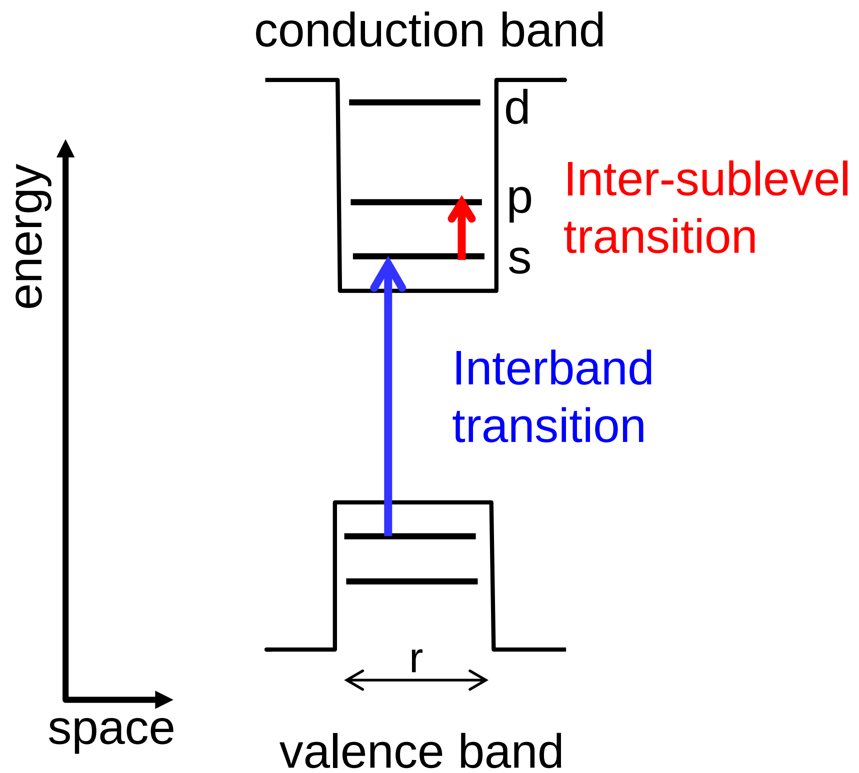
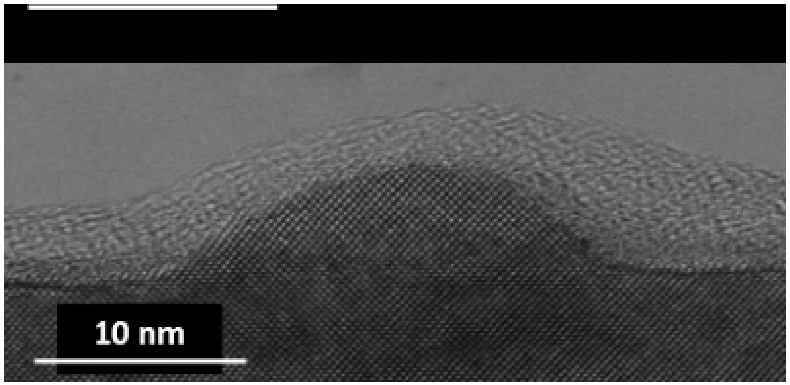
Quantum dot (QD)

- **3-dimensional confinement → 0-dimensional system**
- **discrete energy spectrum → "artificial atom"**



TEM of InGaAs/GaAs QD



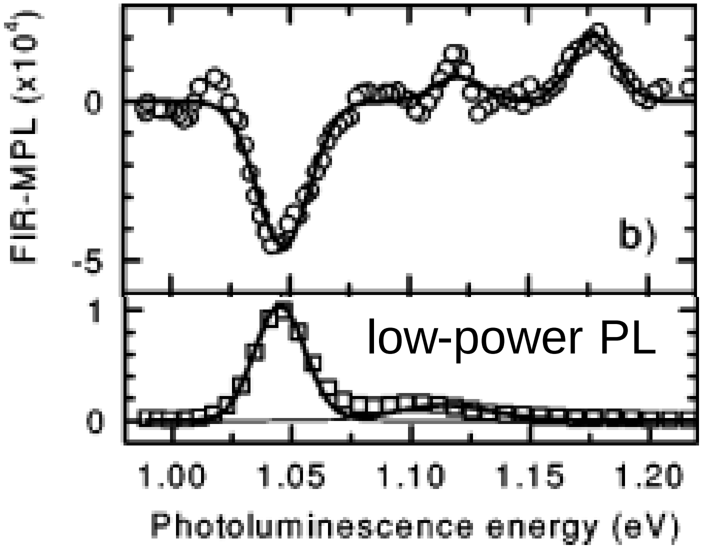
Applications

- QD laser:
  interband (NIR)
- single-photon emitter (NIR)
- QD infrared/THz photodetector:
  inter-sublevel (THz)

Optical studies of inter-sublevel dynamics in QDs

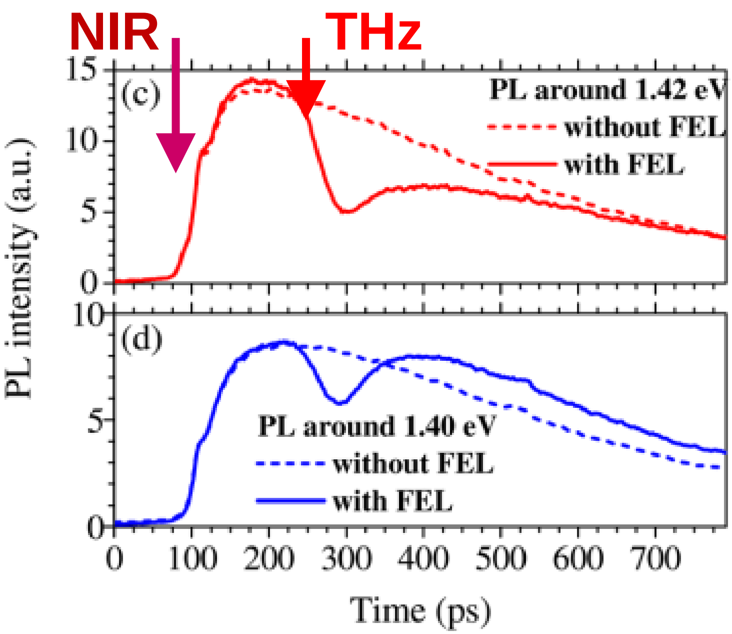
PL of quantum dot (QD) ensembles under intense THz excitation

THz-induced PL difference



B. Murdin et al., PRB **62**, R7755 (2000)

Time-resolved PL



J. Bhattacharyya et al., APL **100**, 152101 (2012)

Problem: Fluctuations of QD size and composition

→ pronounced inhomogeneous broadening

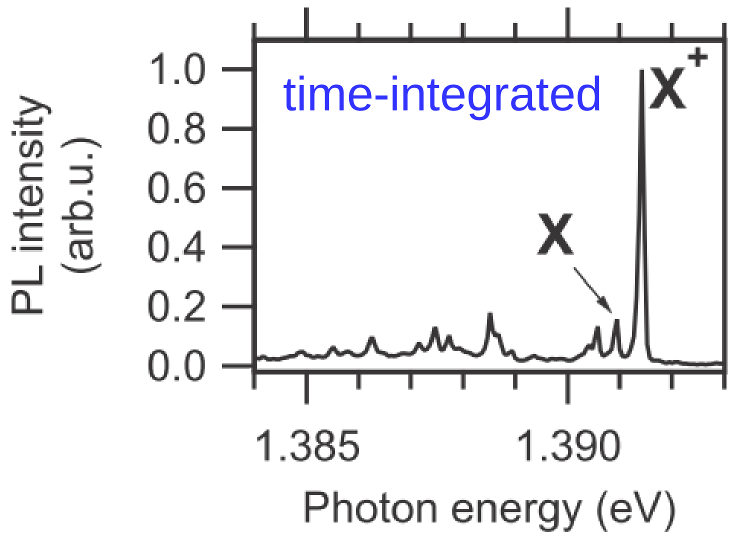
Solution: Investigate individual QD!!

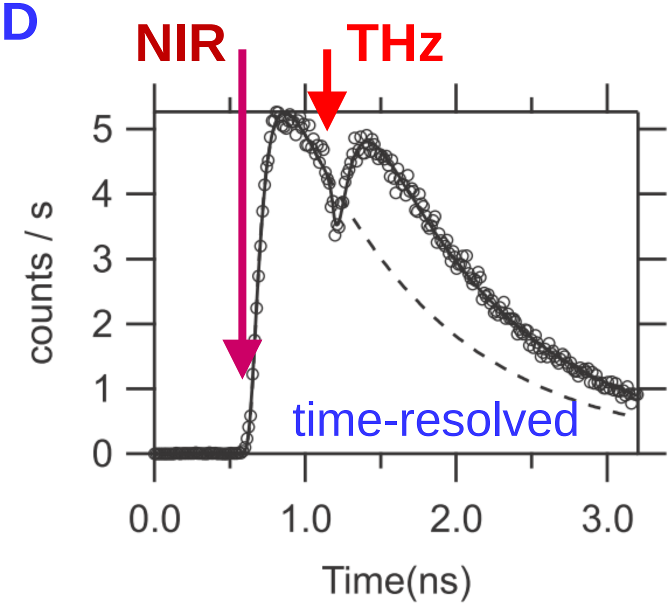
Investigation of a single QD by micro-PL

Micro-PL setup

- optical cryostat, microscope objective
- spectrometer with Si CCD → time-integrated PL
- time-correlated single-photon counting → time-resolved PL

PL of single InGaAs/GaAs QD





X exciton
X^+ pos. trion

NIR/THz two-pulse excitation with ps Ti:Sapphire laser and FEL

- FEL photon energy ≡ s-p transition

D. Stephan et al., Appl. Phys. Lett. ***108****, 082107 (2016)*

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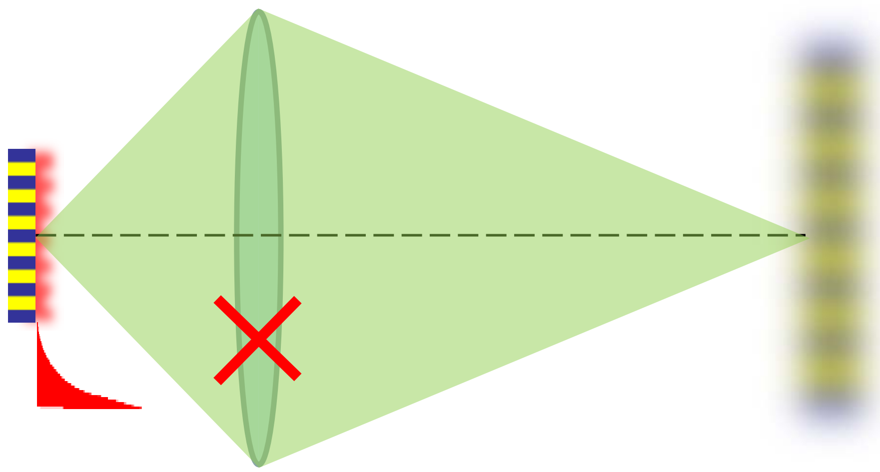
Near field and diffraction limit

Fundamental resolution of conventional optical microscopy



Far field, propagating waves $\mathbf{E} = \mathbf{E}_0\, e^{\,ik_\perp z}$ $\Delta_{\min} \approx \lambda/2$

Near field, evanescent waves $\mathbf{E} = \mathbf{E}_0\, e^{-k_\perp z}$ $\Delta \ll \lambda/2$

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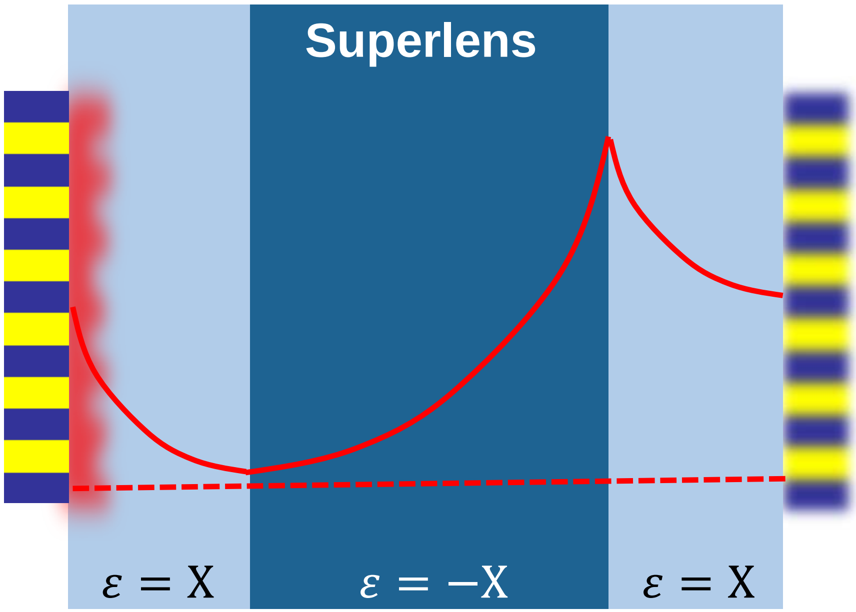
Harald Schneider | Institute of Ion-Beam Physics and Materials Research | www.hzdr.de

The superlens

How to improve resolution?

→ By reconstructing evanescent fields via negative permittivity!



Near-field recovery

$\varepsilon < 0$, dimension << λ

J. Pendry, *Phys. Rev. Lett.* **85**, 2000

First experimental demonstration

$SiO_2/SiC/SiO_2$

superlensing due to phonons → **fixed resonance** @ 10.86 µm

T. Taubner et al., Science ***313****, 2006*

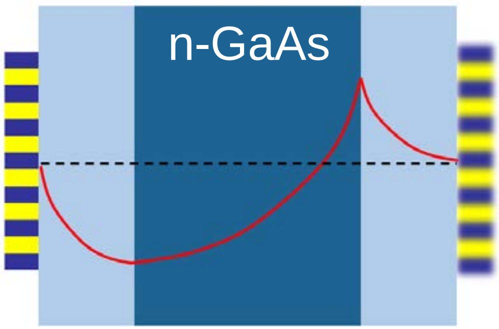
Spectrally tunable n-GaAs superlens

Dielectric function of n-GaAs: *Drude-Lorentz model*

$\varepsilon = \varepsilon(\lambda)$

$$\varepsilon(\omega) = \varepsilon_{\text{optic}} - \underbrace{\frac{\omega_p{}^2 \varepsilon_{\text{optic}}}{\omega^2 - i\omega\gamma_{\text{el}}}}_{\text{Plasmons}} - \underbrace{\frac{\omega_{\text{TO}}{}^2 (\varepsilon_{\text{static}} - \varepsilon_{\text{optic}})}{\omega^2 - \omega_{\text{TO}}{}^2 + i\omega\gamma_p}}_{\text{Phonons}}$$

$$\omega = \frac{2\pi c}{\lambda}$$



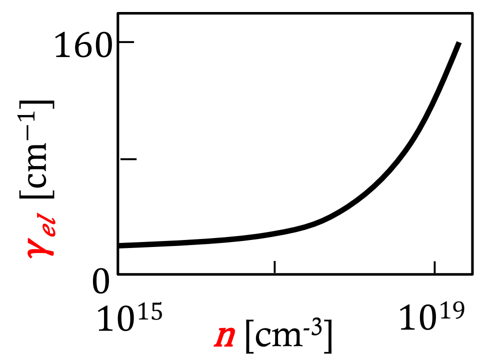
Plasmonic superlens

↓

Adjustable resonance

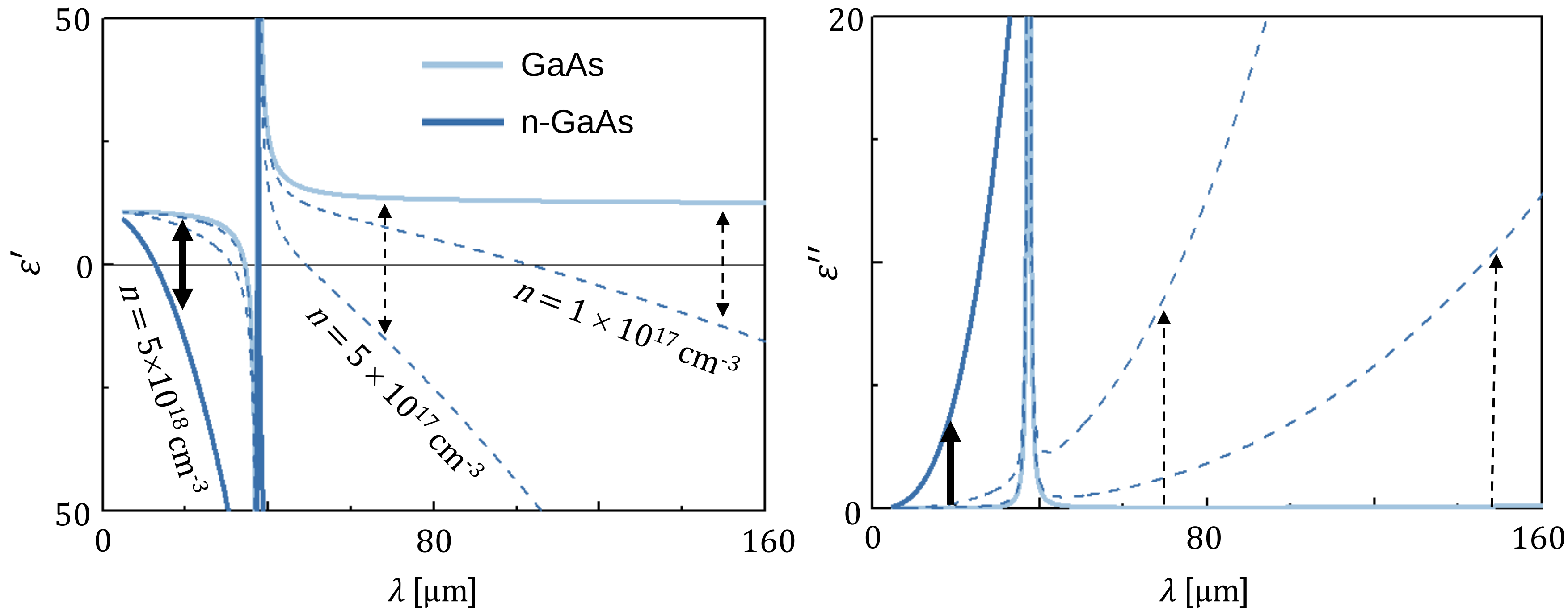
ω_p plasma frequency
n carrier density

$$\omega_p = \sqrt{\frac{ne^2}{m^* \varepsilon_0 \varepsilon_{\text{optic}}}}$$



γ_{el} electronic damping (from literature)

Spectral tunability of n-GaAs superlens



Resonance condition

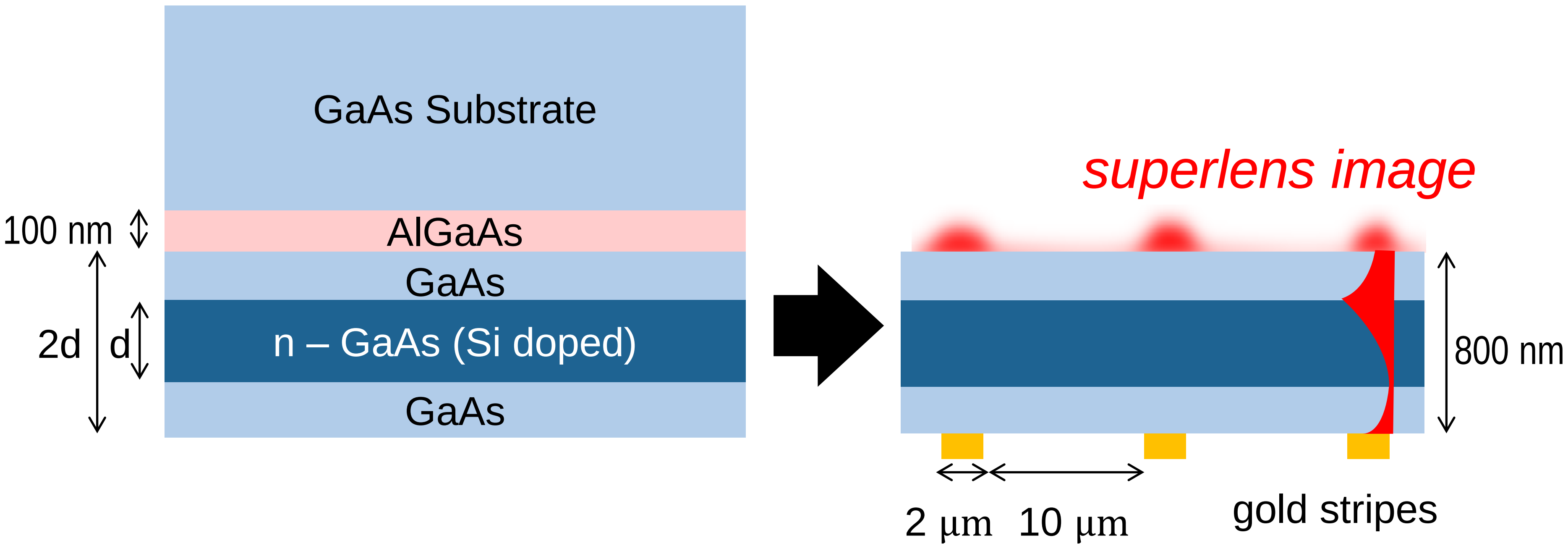
$$\varepsilon'_{\text{n-GaAs}}(\lambda_{SL}) = -\varepsilon'_{\text{GaAs}}(\lambda_{SL})$$

Quality

$$\varepsilon''_{\text{n-GaAs}} \ @ \ \lambda_{SL}$$

→ major part of the mid/far-infrared(THz) spectral range can be covered!

Samples



Sample parameters d = 200 nm and 400 nm

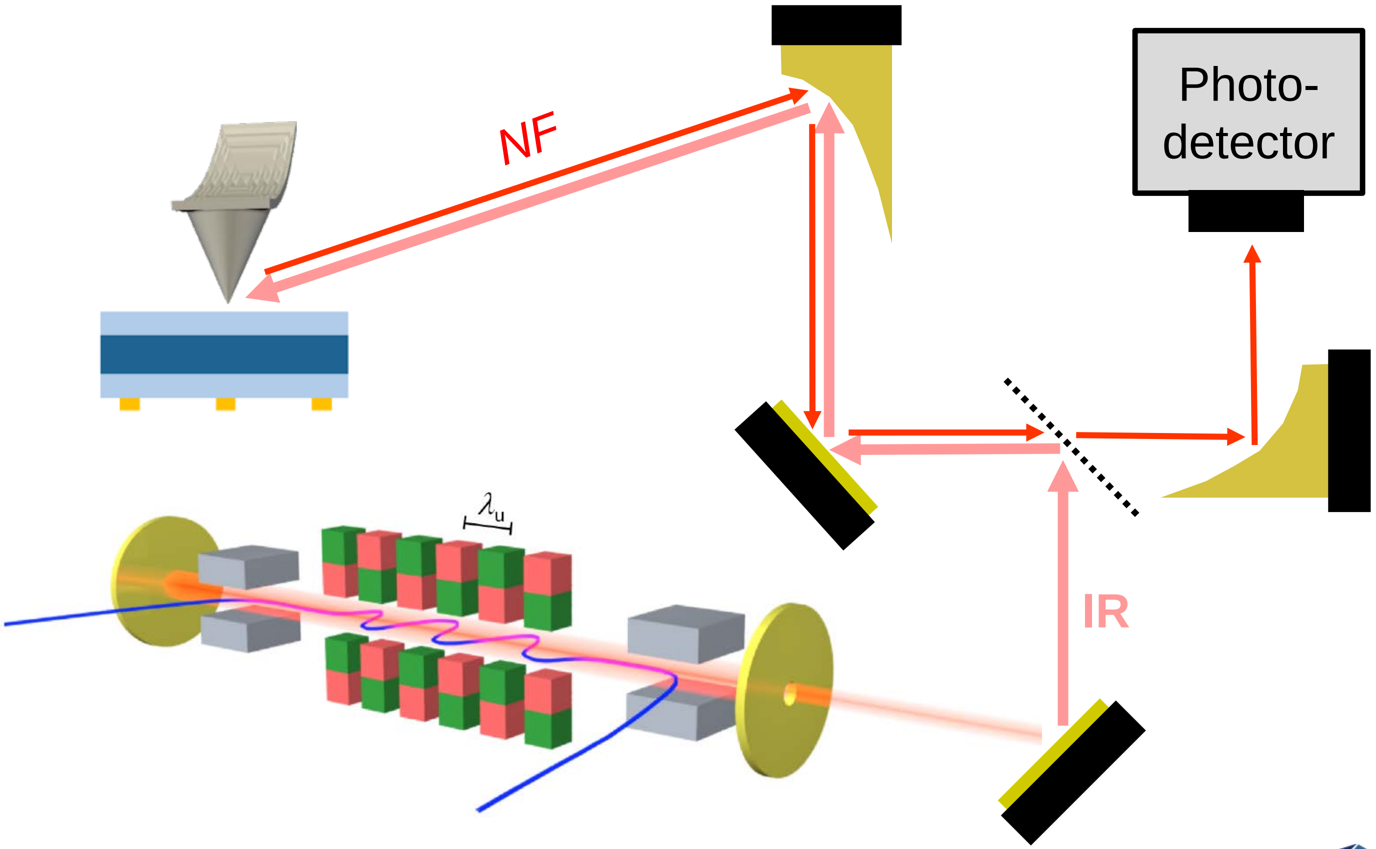
n = 3…4 x 10^{18} cm^{-3}

Growth by molecular beam epitaxy

Setup

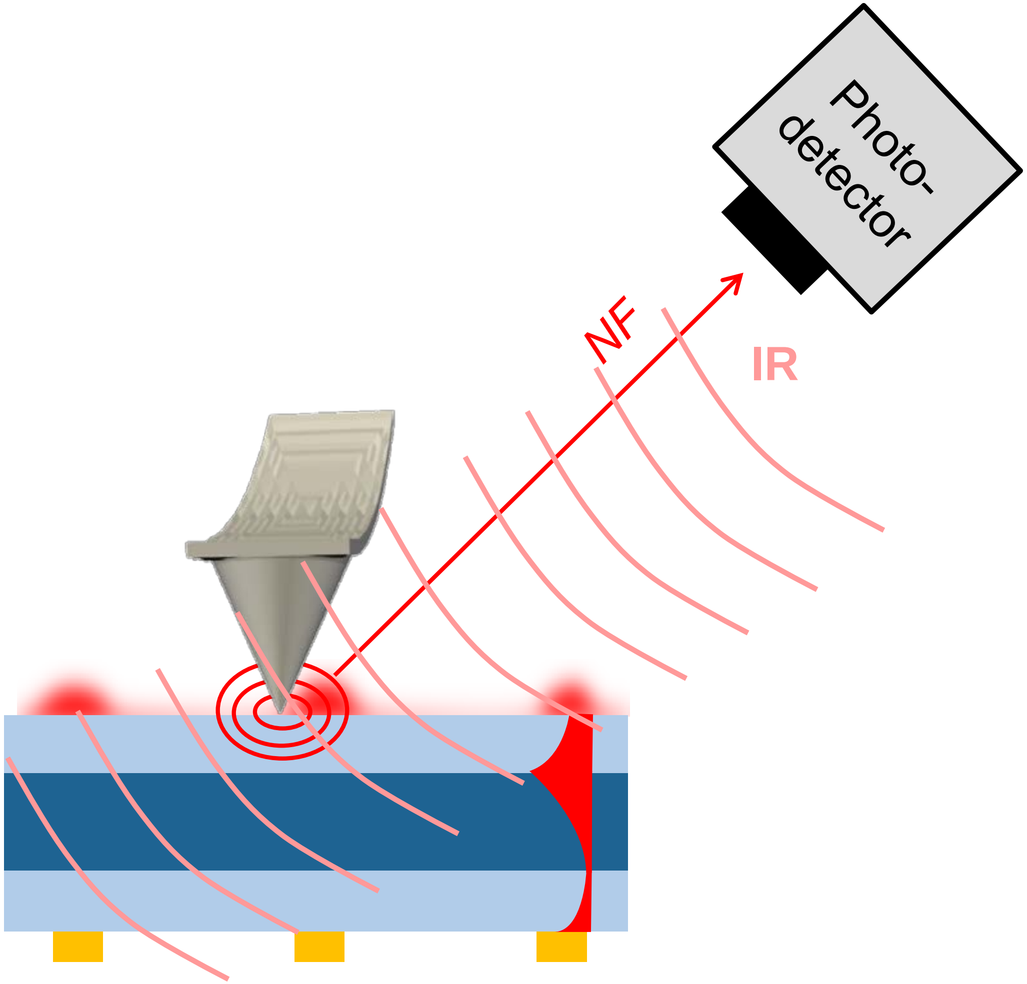
Near-field microscopy with a free-electron laser

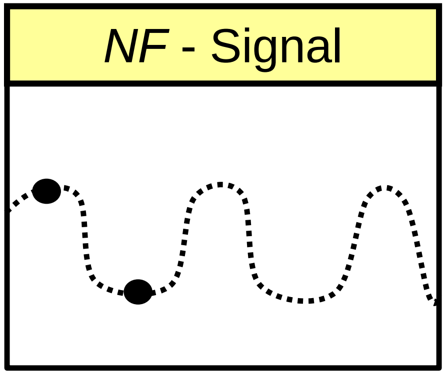
FELBE: $\lambda = 4 - 250\ \mu\text{m}$

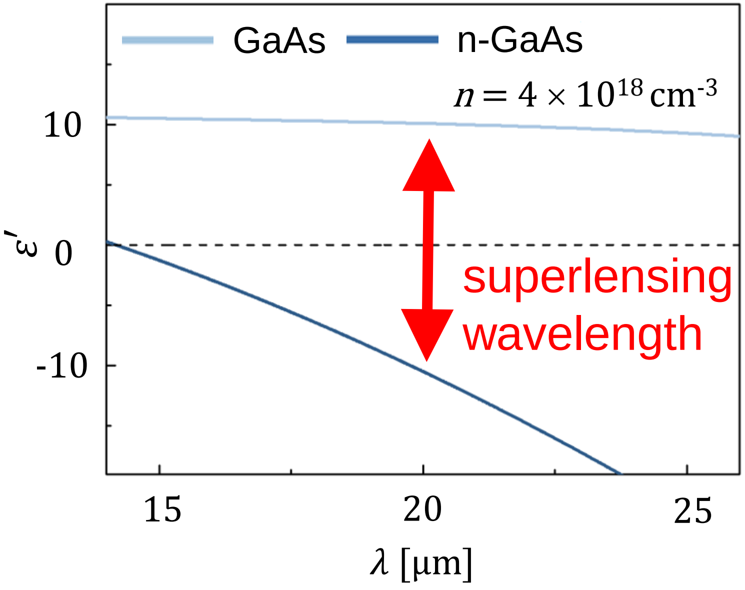


Near-field imaging across a superlens

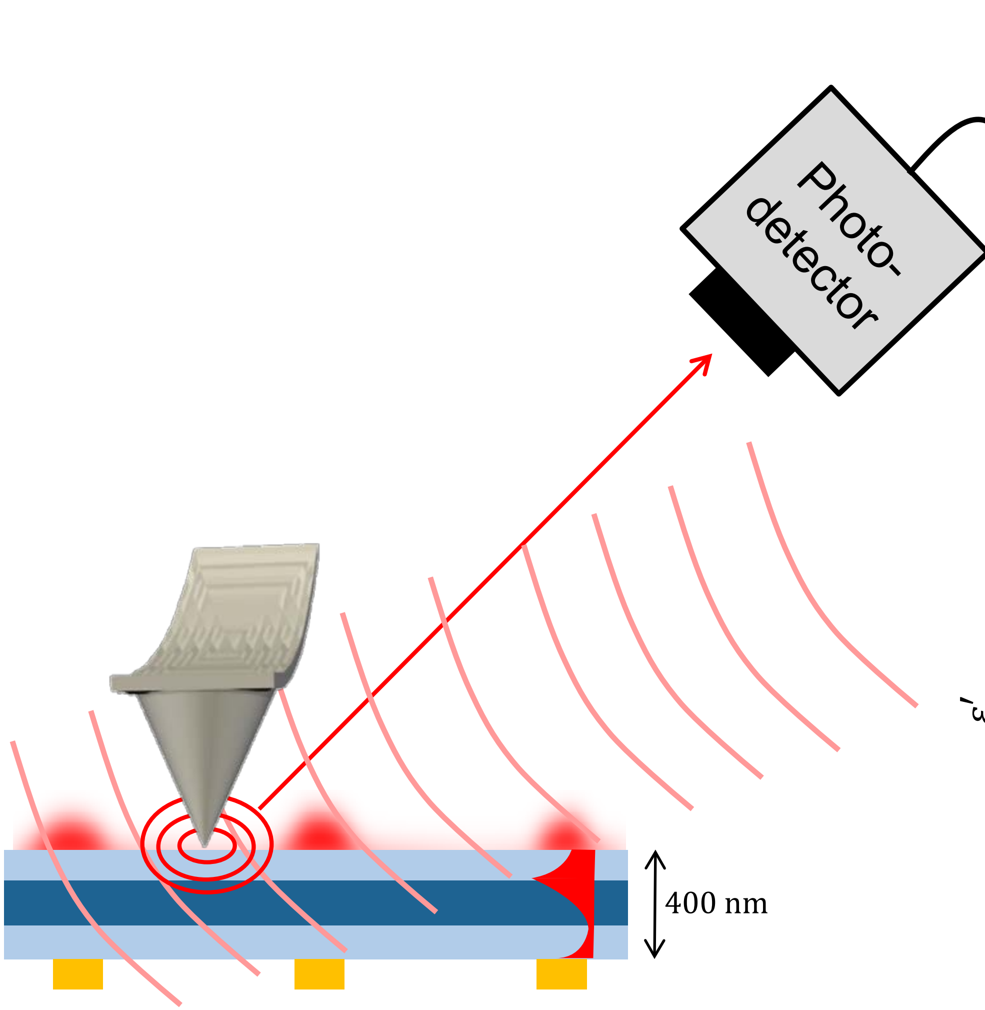
Resolution enhancement at the resonant wavelength

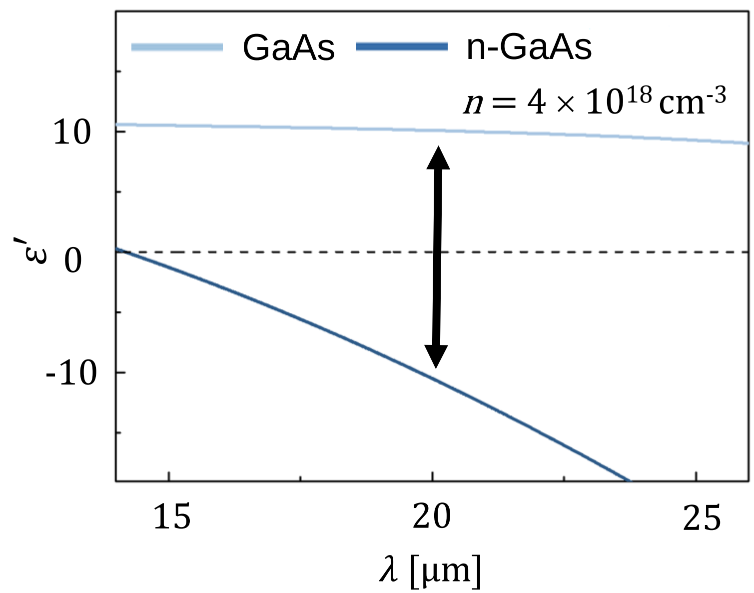




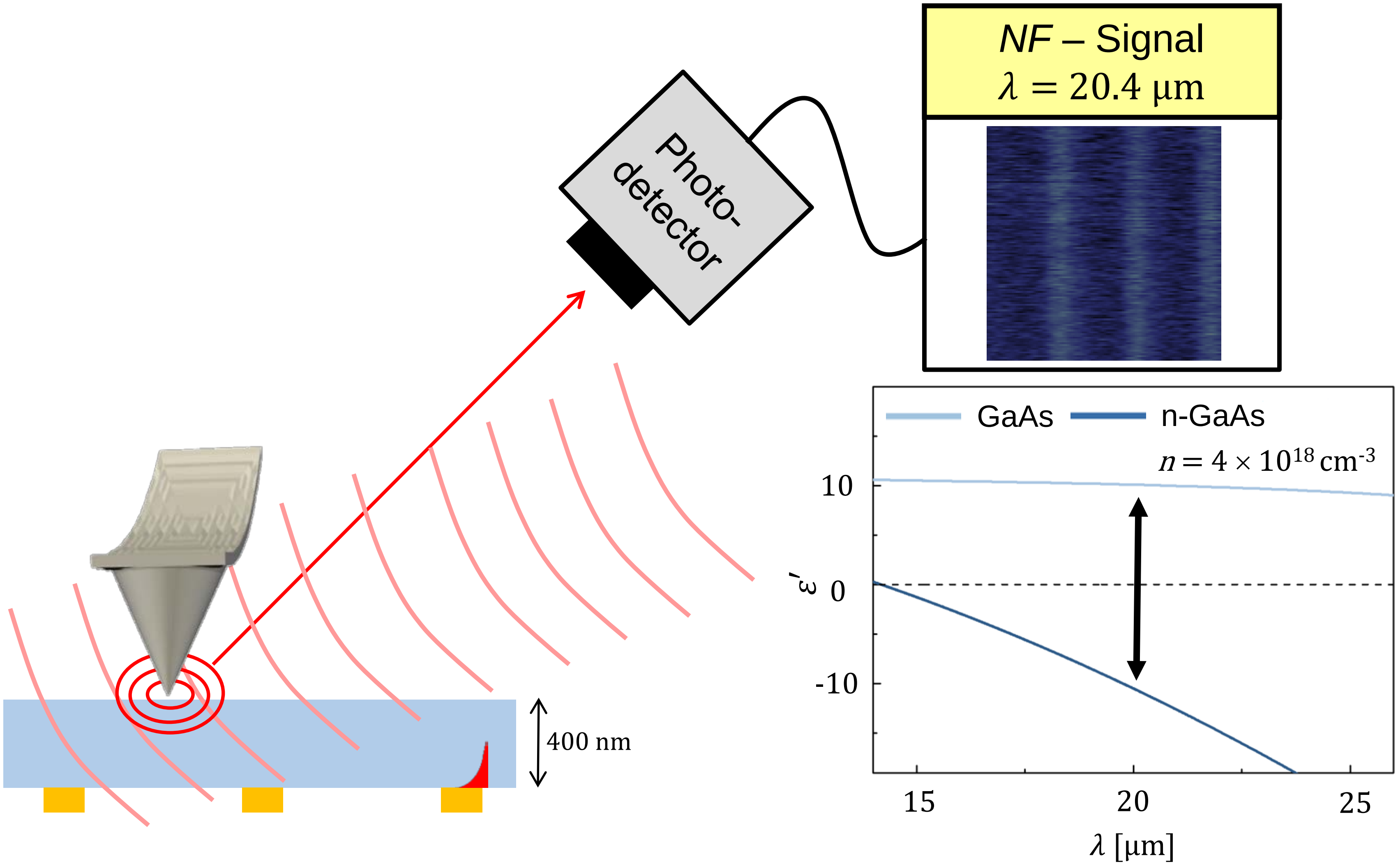


Results: Image with superlens

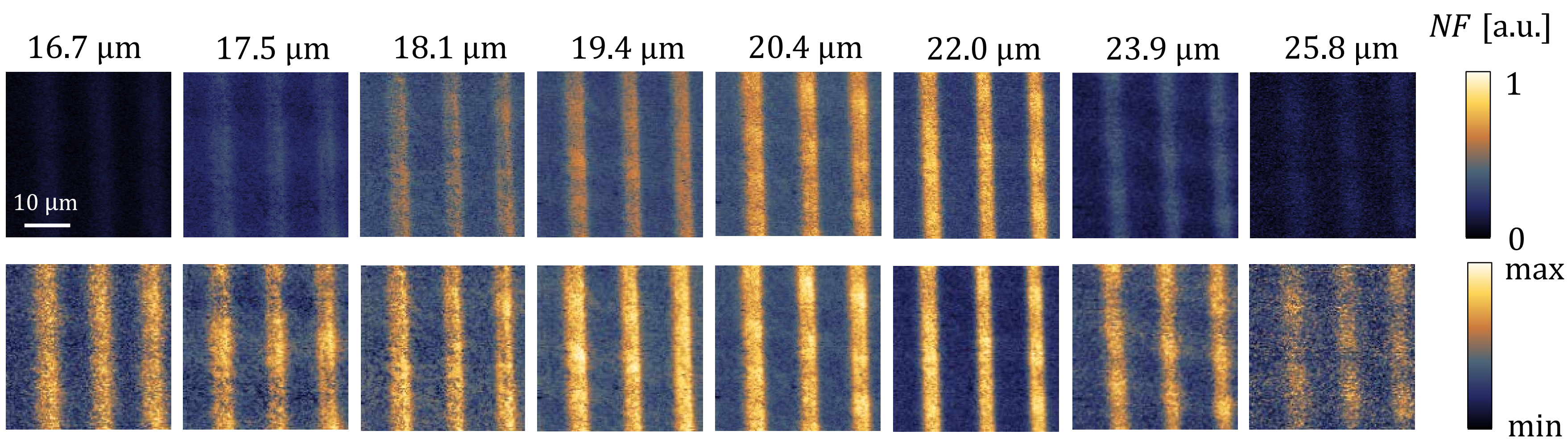


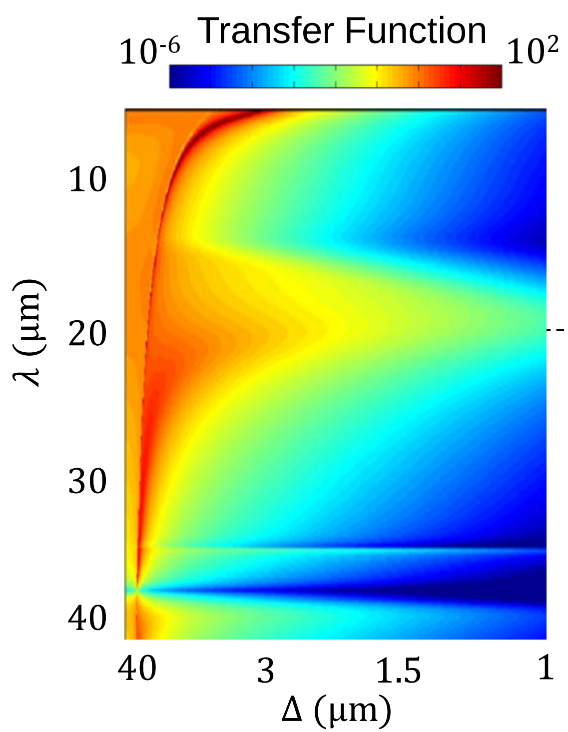


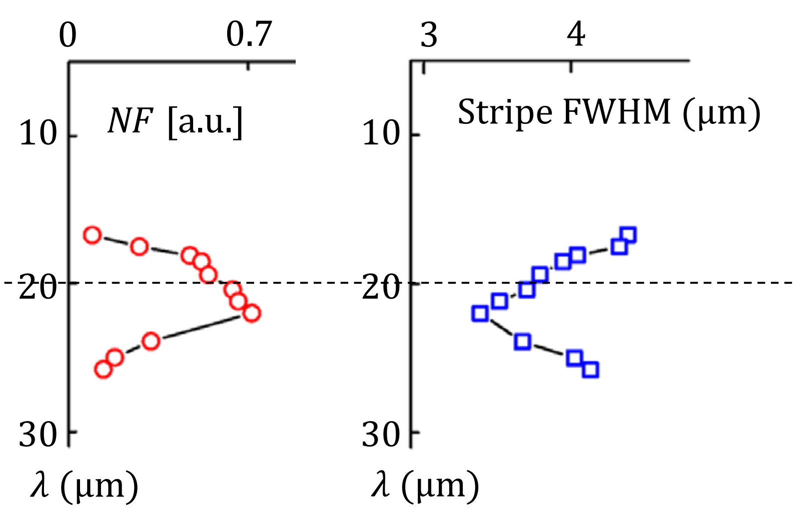
Results: Comparison with reference sample



Results: Spectral dependence





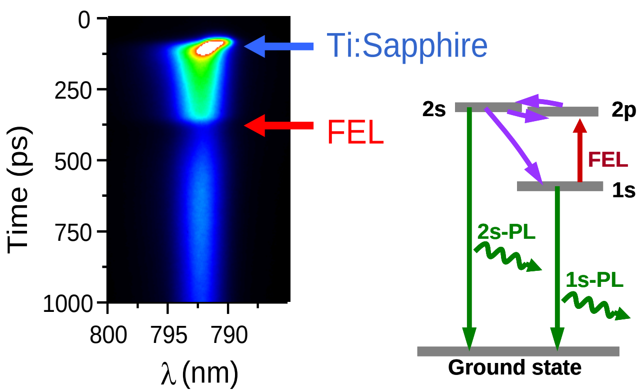


M. Fehrenbacher et al., Nano Lett. **15**, 1057 (2015)

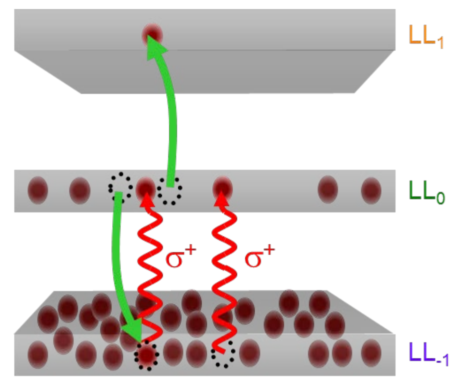
Conclusion

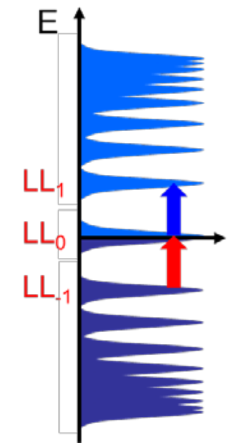
- **Free-electron laser: a unique high-power mid-IR/THz source**
- **Plenty of opportunities for nonlinear & high-field spectroscopy**

Excitons in QWs

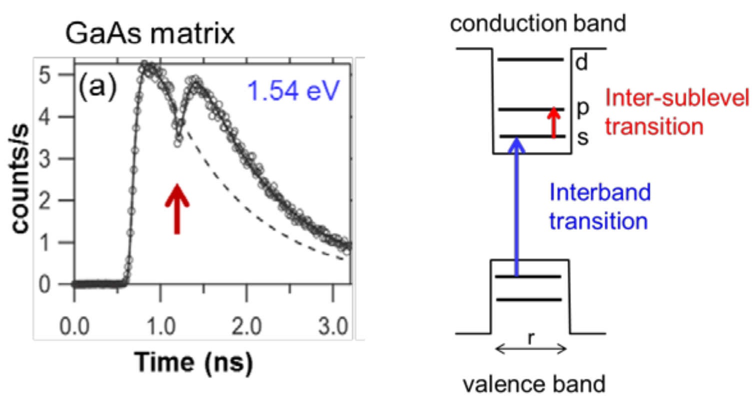


Graphene

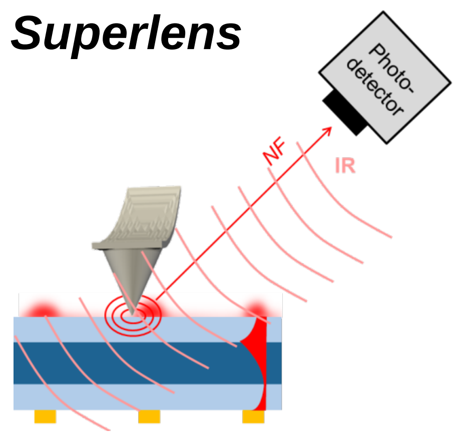


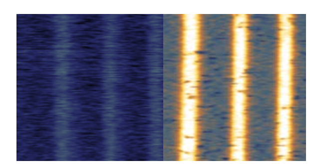


Carrier dynamics in QDs



Superlens





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graphene

QWs, QDs

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SEVENTH FRAMEWORK PROGRAMME

elisa

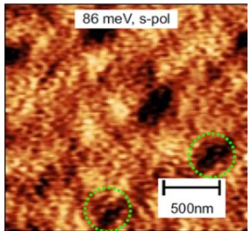
DRESDEN concept

HZDR

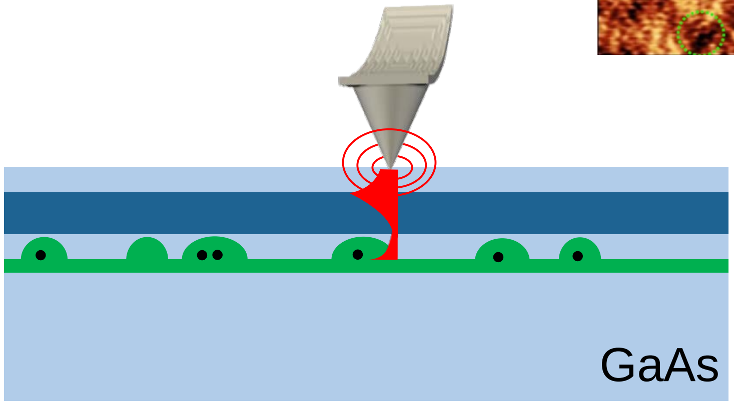
Superlens applications

- Near-field imaging of objects which have to be buried under a cover layer

→ InAs quantum dots



R. Jacob et al.,
Nano Letters **12**, 4336 (2012)



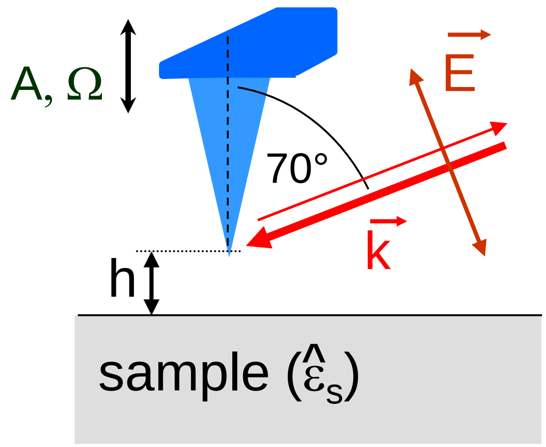
→ Doping profiles (ion implantation)

→ Carrier distribution in transistors

→ …

… also possible with other semiconductor materials

s-SNOM in the infrared/THz regime

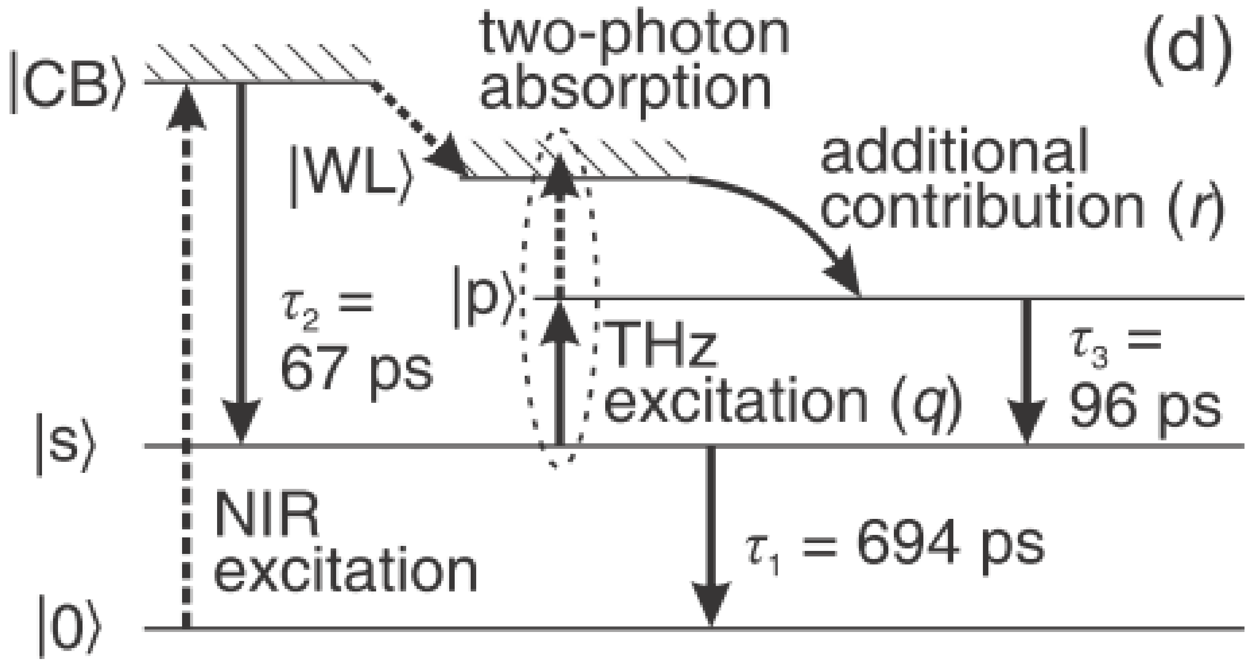


Free-electron laser (FEL) as tunable light source in the IR/THz regime

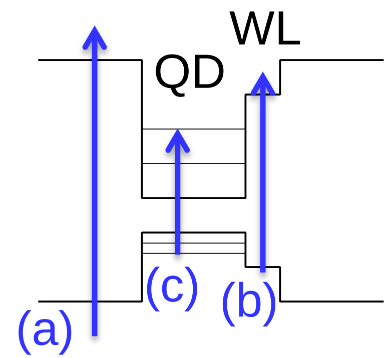
Non-contact atomic-force microscope

- cantilever frequency ν = 170 kHz at constant amplitude of ~40 nm (pp)
- detection at 2nd or 3rd harmonics of ν
  → near-field enhancement
  → spatial resolution ~100 nm for ~50 nm tip radius

Rate equation analysis



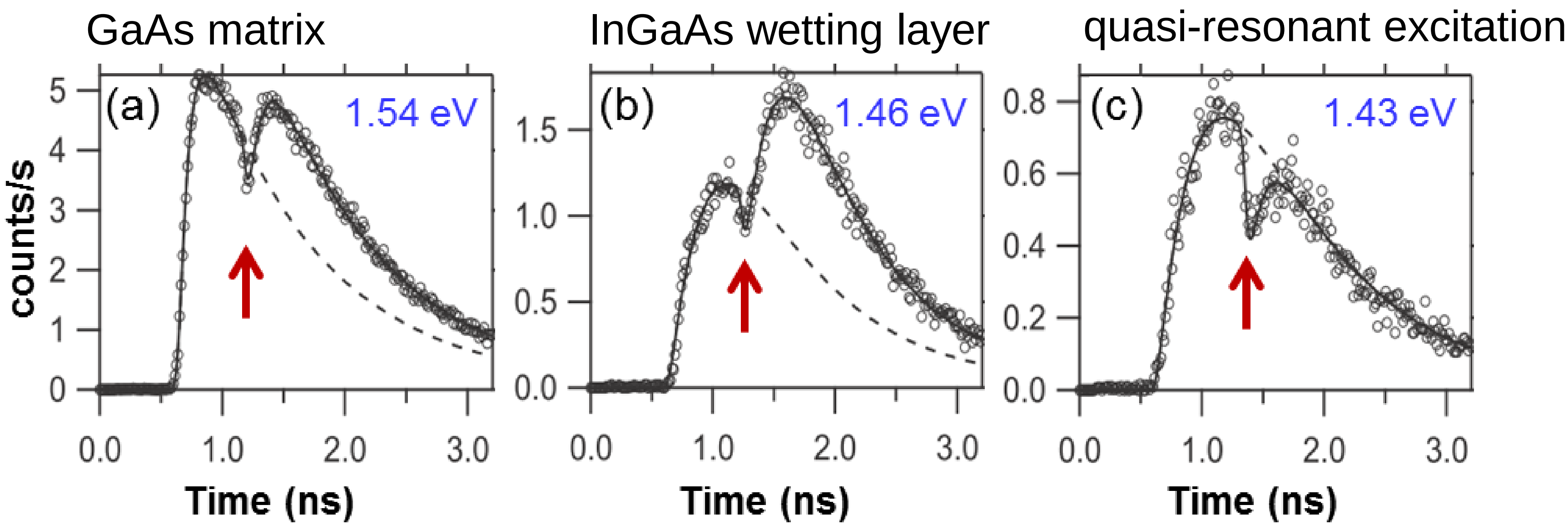
Dependence on near-IR excitation energy



PL transients of X^+ trion

FEL@13.3 meV

Interband excitation at different photon energies



- THz pulse induces a release or trapping of charge carriers
  → qualitatively different PL transients
- Carrier surplus originates from InGaAs wetting layer

D. Stephan et al., Appl. Phys. Lett. ***108****, 082107 (2016)*