Semiconductors in high THz fields

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Heraeus Seminar
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Bad Honnef

Partner von DRESDEN concept exzellenz-dresden.de

HZDR
HELMHOLTZ ZENTRUM DRESDEN ROSSENDORF

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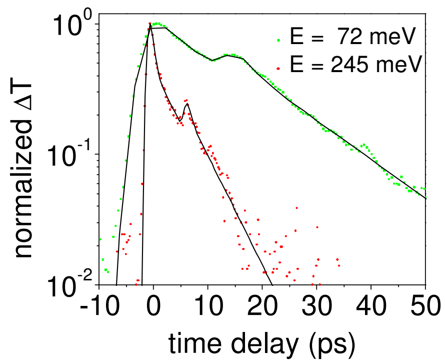
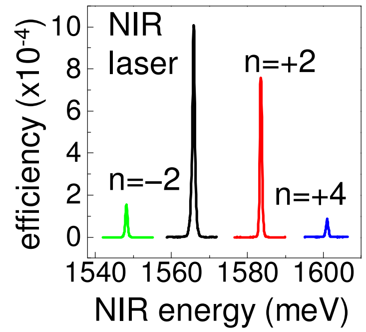
Outline

1. **Motivation**

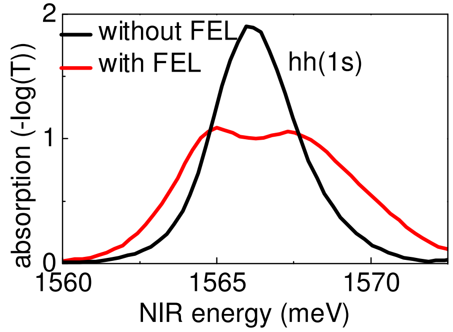
2. **Population dynamics:**
   pump-probe spectroscopy on graphene



3. **Perturbative nonlinear interaction:**
   sideband generation,
   excitons in semiconductor quantum wells



4. **Nonperturbative regime:**
   intraexcitonic AC Stark effect
   (Autler-Townes splitting)



5. **Summary**

Motivation

different regimes of nonlinear optics:

- **perturbative regime:** expansion in powers of electric field E
  $\rightarrow$ nonlinear susceptibilities $\chi^{(1)}$, $\chi^{(2)}$, $\chi^{(3)}$ etc.

- **non-perturbative regime:** finite population in excited states generated;
  Rabi energy $\hbar\Omega = \mu E$ larger than broadening γ
  $\rightarrow$ dressed states

- **non-resonant high-field regime:** electric field so strong to modify the „atomic“ potential:
  - electric-field induced ionization
  - high-harmonic generation …

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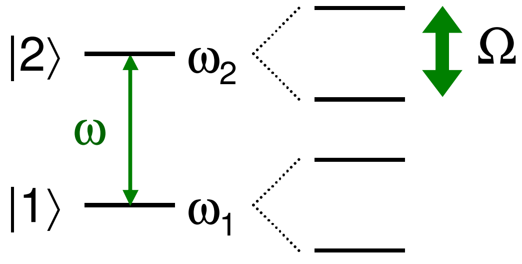
Motivation

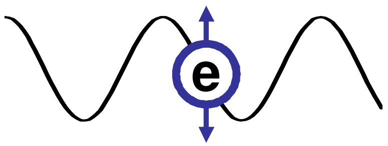
energies/frequencies in light-matter coupling:

- photon energy & transition energy ω
  also binding energy



- coupling (Rabi) energy $\hbar\Omega = \mu E$

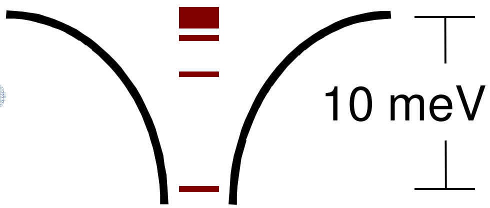
- ponderomotive energy $U_p = \frac{1}{T}\int_0^T \left(\frac{1}{2}mv^2\right)dt = \frac{e^2 E_0^2}{4m\omega^2}$



In the **THz range** (1 THz = 4.1 meV) all of them can easily become of the order of 10 meV!

+

excitons represent our **artifical atoms** with $E_{binding} \approx 10\,\mathrm{meV}$



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Unexplored regime!

DRESDEN concept HZDR

Free-electron laser FELBE

Tuning range

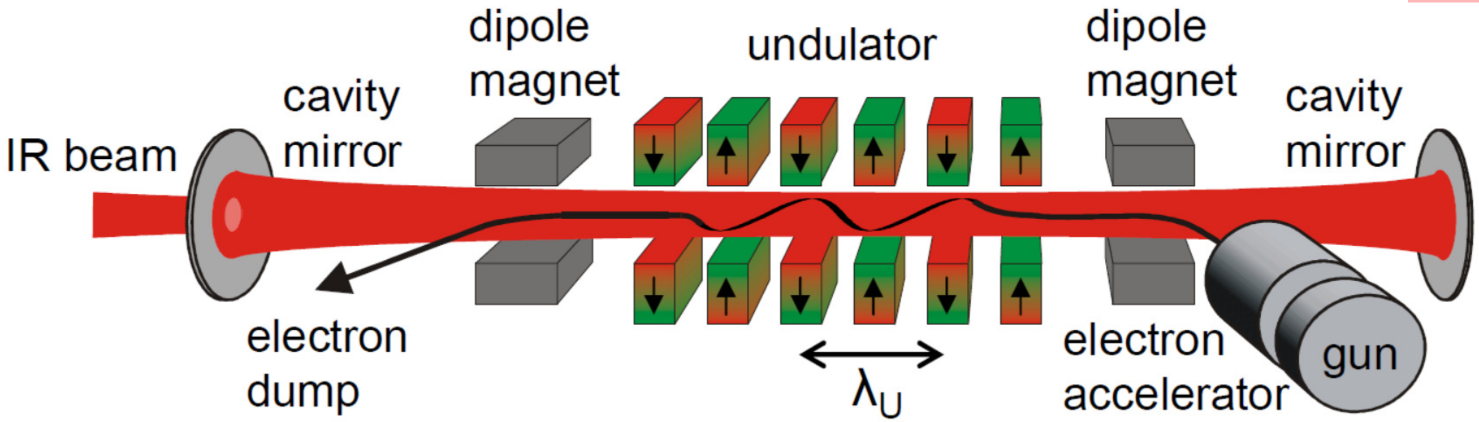
FEL I λ = 4 – 22 µm
FEL II λ = 18 – 250 µm

⇒ 1 – 75 THz (5 – 310 meV)

Average power: 0.1 – 20 W
Spectral width: 0.5 – 2 %
Pulse energy: 0.1 – 2 µJ
Pulse width: 1 – 25 ps
Repetition rate: 13 MHz

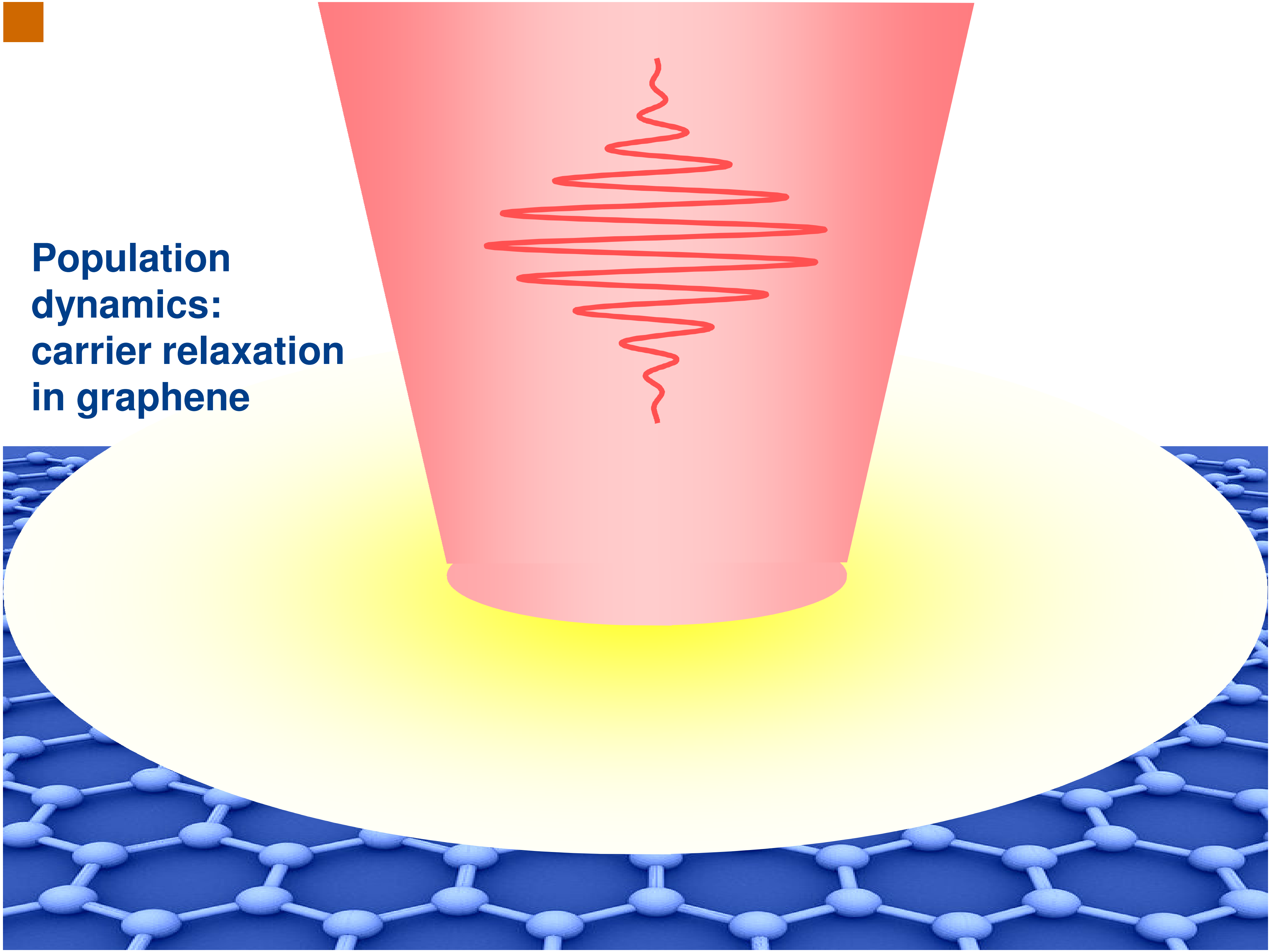
Only continuously pulsing FEL in Europe

See Poster FELBE facility



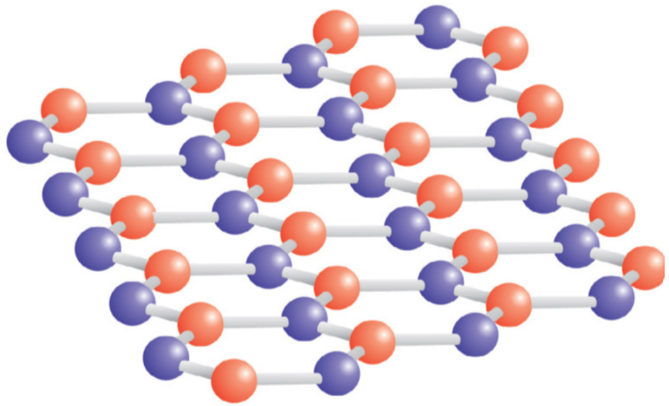
HZDR

Population dynamics: carrier relaxation in graphene

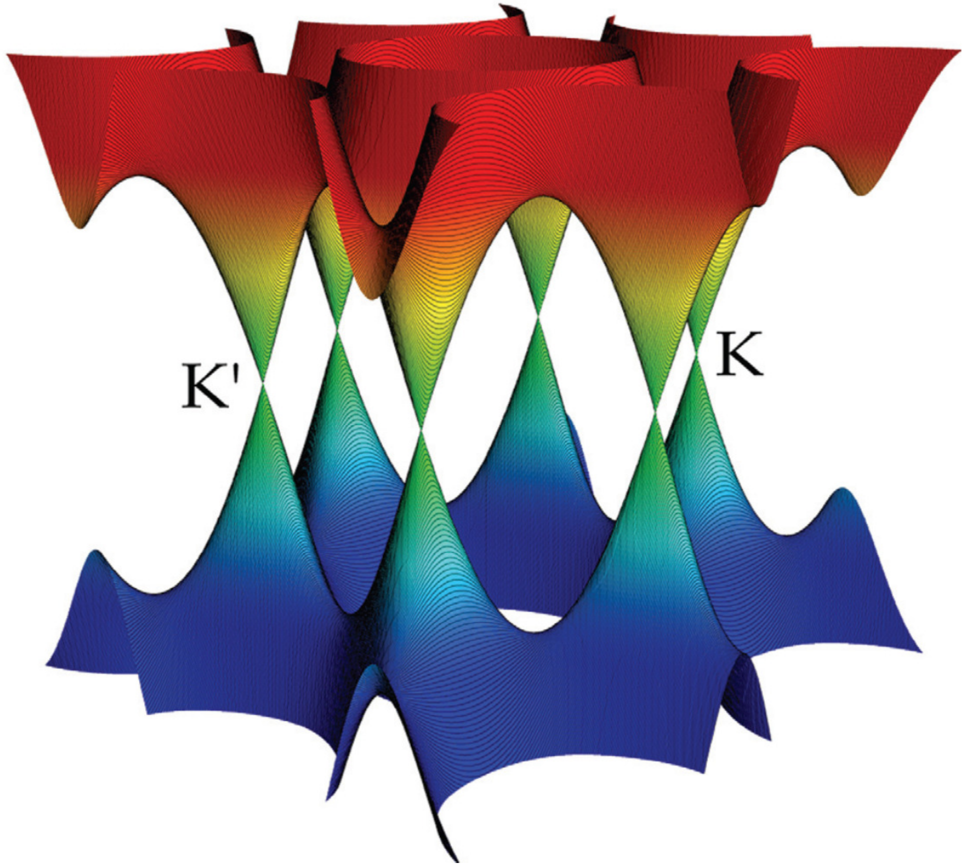


Introduction graphene

Graphene: mono-atomic layer of *sp*^{2} bonded carbon atoms



Hexagonal structure,
two atoms per unit cell

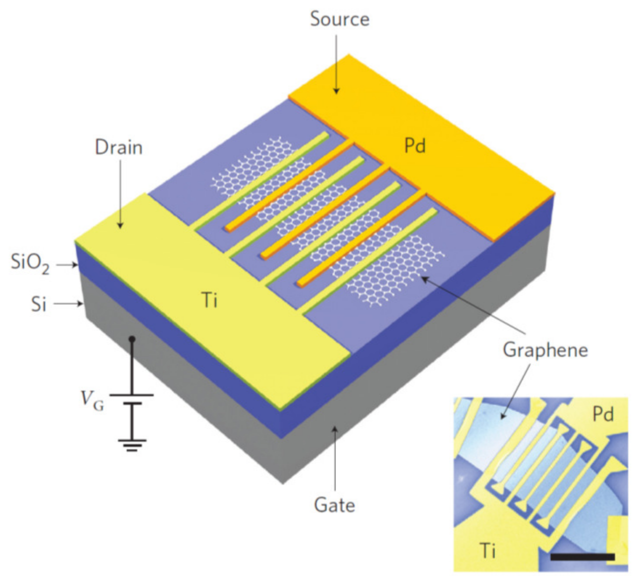


Band structure with Dirac points

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

Why study the carrier relaxation dynamics in graphene?

- Basic interest
  - Understanding of carrier dynamics, electron-phonon interaction,…
- Applications in optoelectronics
  - Conductive transparent coatings
  - Detectors, saturable absorbers
  - Novel THz devices



T. Mueller et al., Nature Photonics 2010

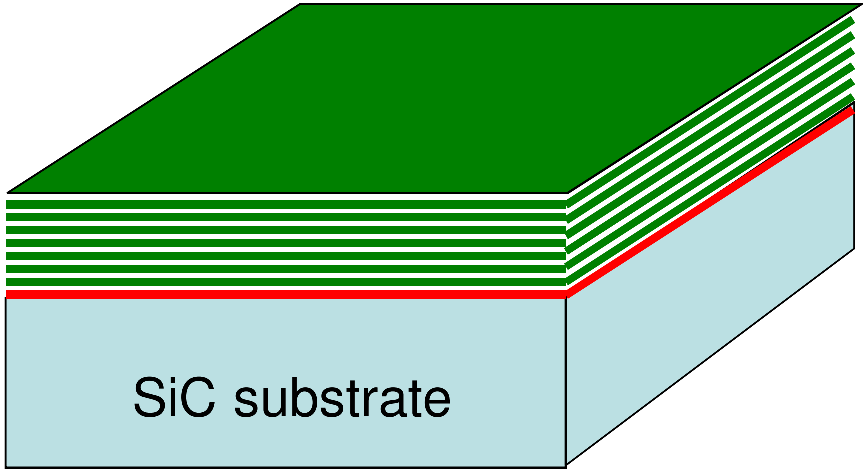
- **Previous studies: excitation at E ≈ 1.5 eV**

*J.M. Dawlaty et al., APL **92**, 042116 (2008),*
*H. Wang et al., APL **96**, 081917 (2010),*
*D. Sun et al., PRL **104**, 136802 (2010),*
*M. Breusing et al., PRB **83**, 153410 (2011),…*

- **Focus of our study: low energy excitation, E = 10 – 300 meV**

Optical phonons (~200 meV), Fermi energy (~10 meV)

Our graphene samples

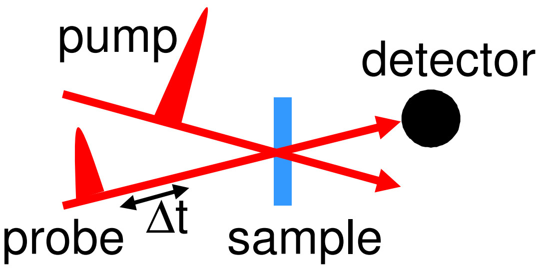


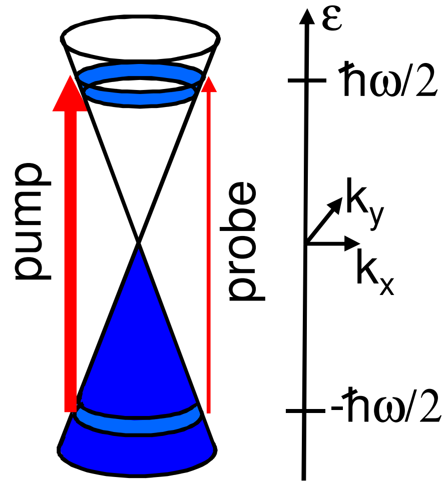
Quasi-intrinisic electronically uncoupled graphene layers (~70)

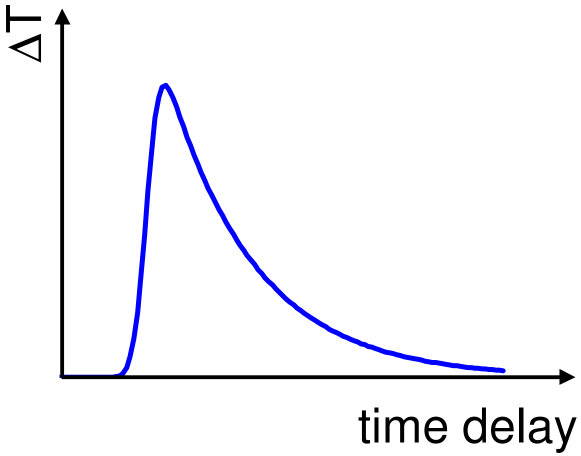
Epitaxial growth: thermal decomposition on the carbon terminated surface of 4*H*-SiC.

Samples well characterized by Raman spectroscopy and static magneto-spectroscopy.

Degenerate pump-probe spectroscopy on graphene







$$\frac{\Delta T}{T_0} \approx -\Delta\alpha(\hbar\omega) = -\alpha_0\left(\Delta f_e\left(\frac{\hbar\omega}{2}\right) + \Delta f_h\left(-\frac{\hbar\omega}{2}\right)\right)$$

population dynamics

$$\alpha_0 = e^2/(4\varepsilon_0\hbar c) \approx 2.3\ \%$$

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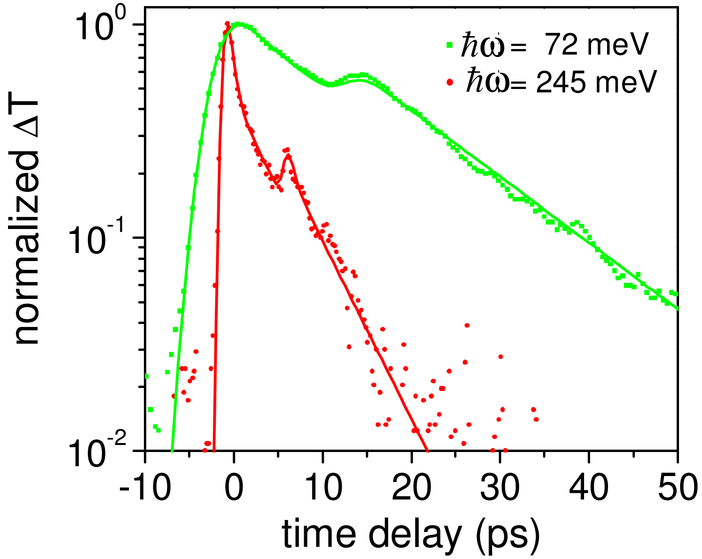
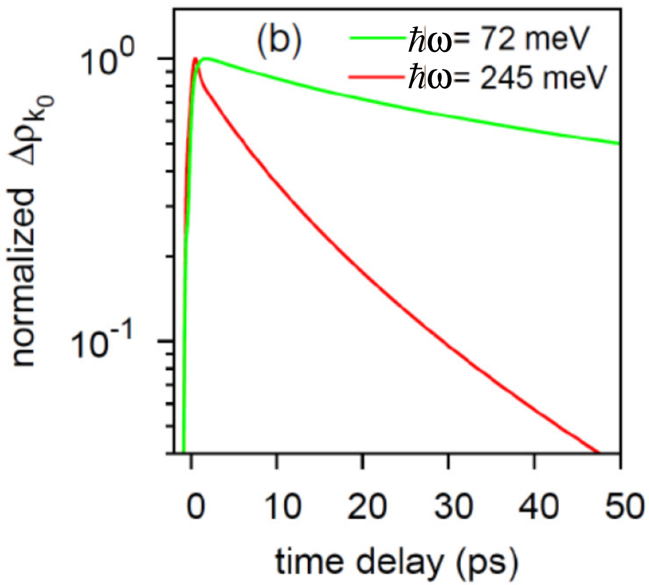
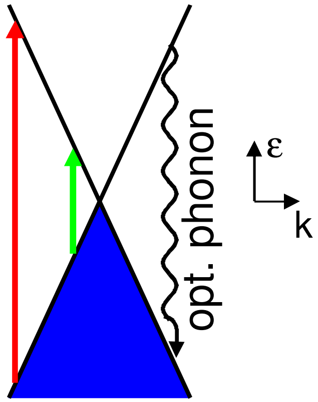
Pumping above and below the optical phonon energy

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)

Pumping above and below the optical phonon energy

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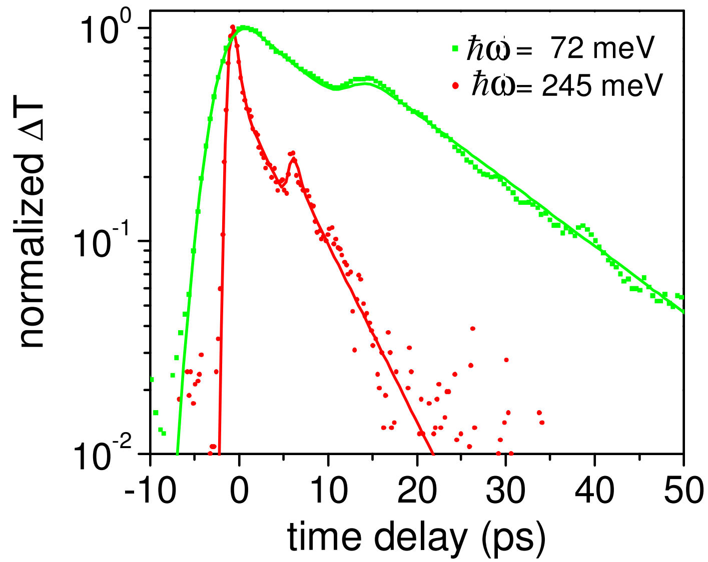

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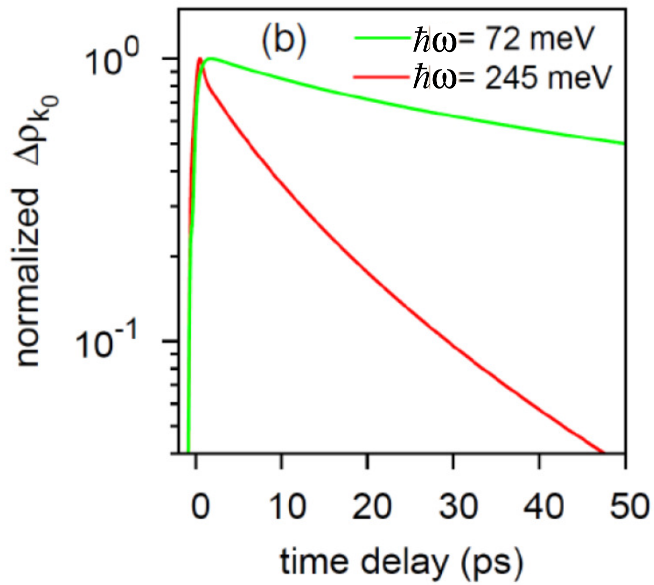
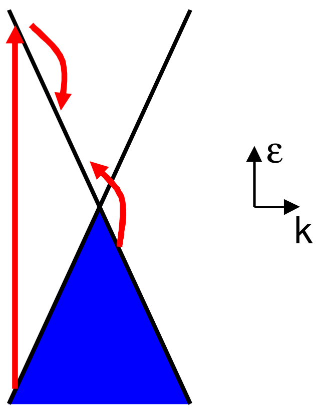




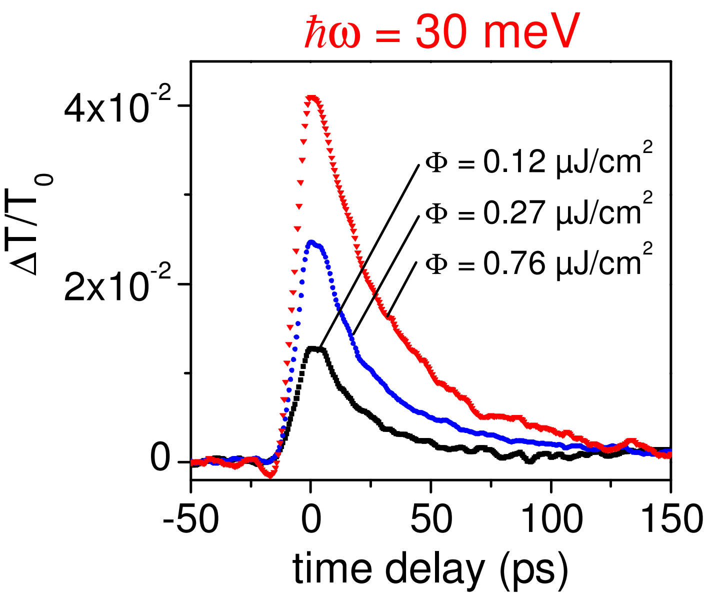
Slower relaxation for photon energies below the optical phonon energy (~200 meV)

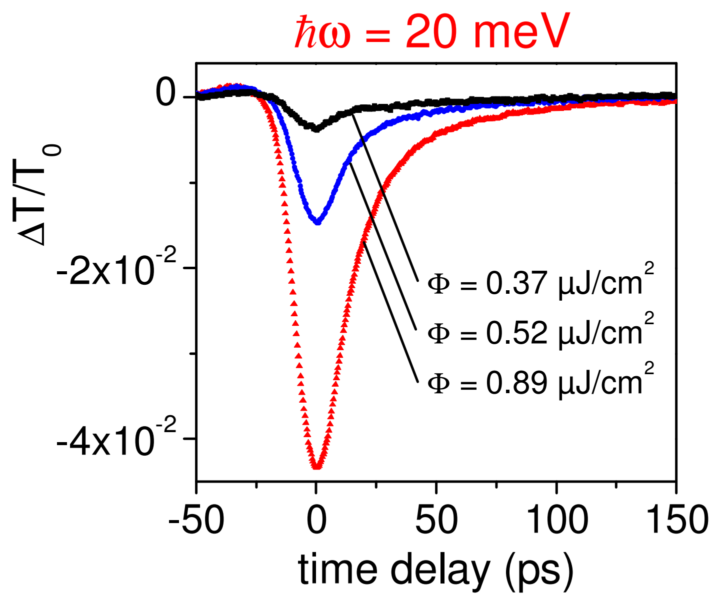
Furthermore:

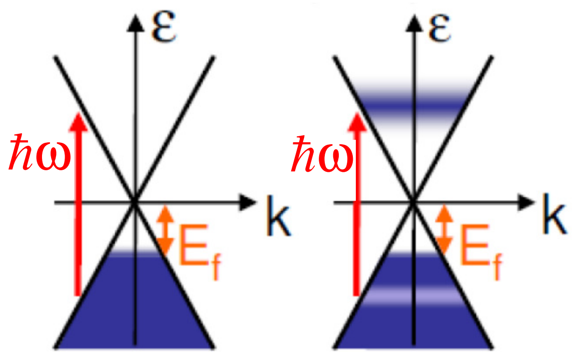
- Role of Auger-type processes (~ 1ps for hw = 245 meV)
- Acoustic phonons: τ ≈ 300 ps

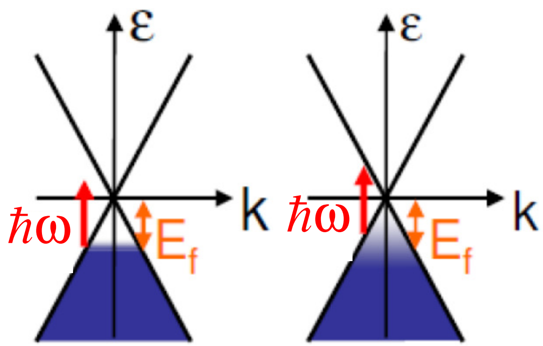
S. Winnerl et al., Phys. Rev. Lett., **107**, 237401 (2011)

Sign reversal of pump-probe signal: Interplay of pump-induced transmission and absorption





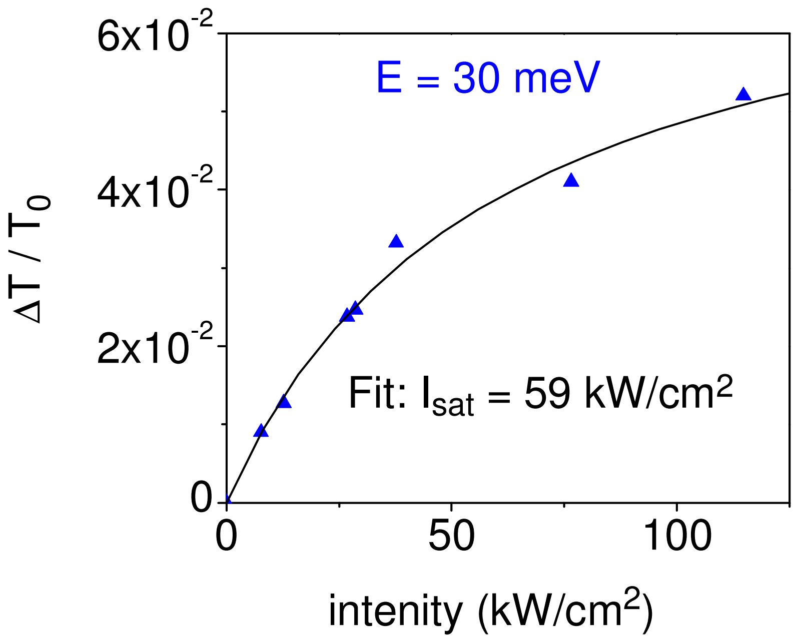


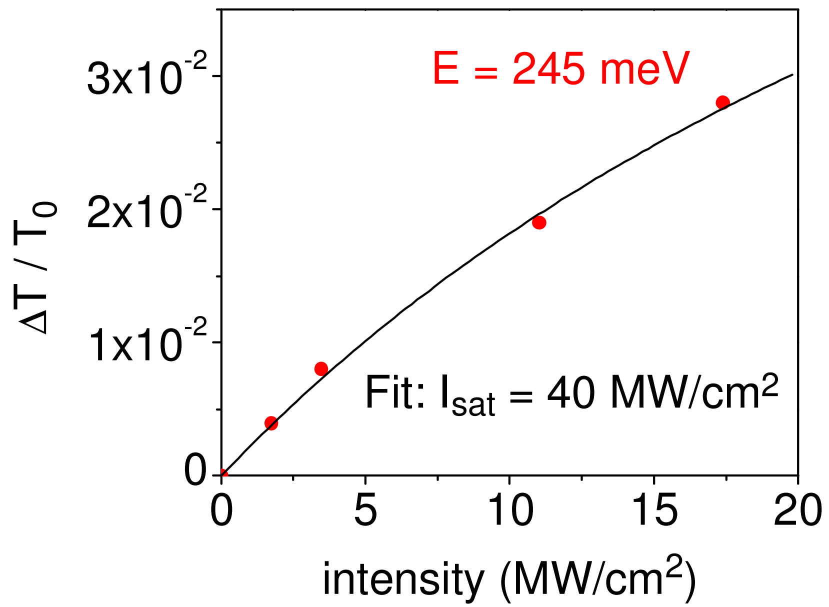


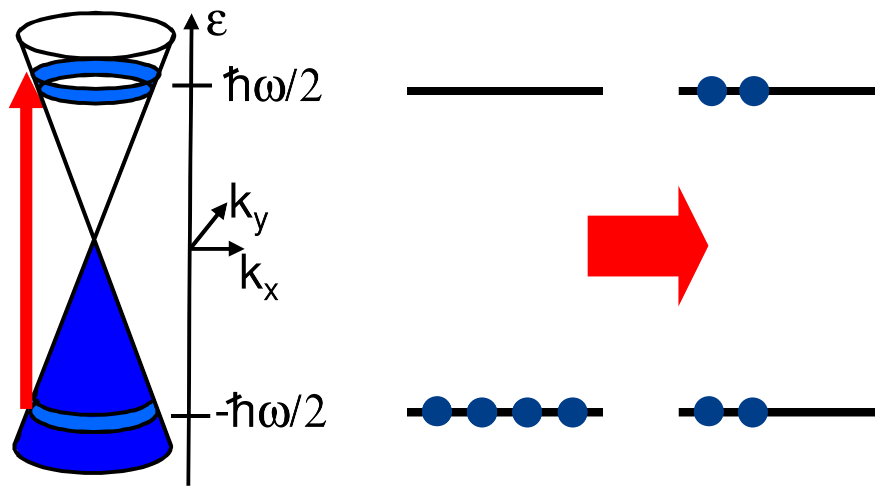
Induced *transmission* for $\hbar\omega > 2|E_F|$ due to Pauli blocking

Induced *absorption* for $\hbar\omega < 2|E_F|$ due to heating of free carriers

Saturation of the pump-probe signal



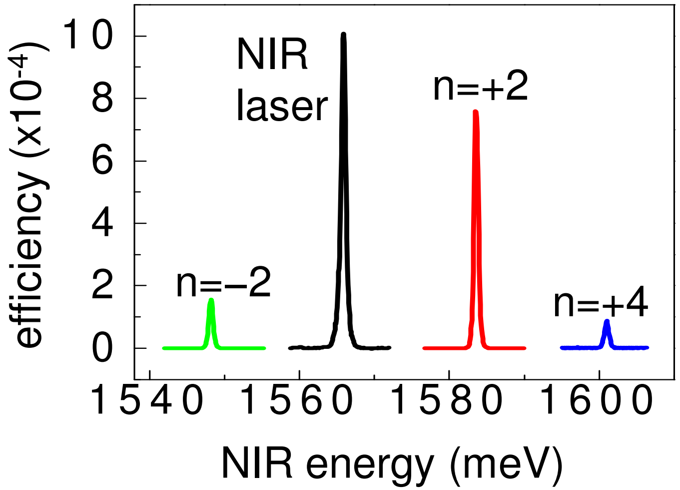




$$\Delta T / T_0 \propto \frac{I / I_{sat}}{1 + I / I_{sat}}$$

Example for
perturbative nonlinear interaction:
sideband generation in semiconductor quantum wells

Resonant excitation of excitonic levels

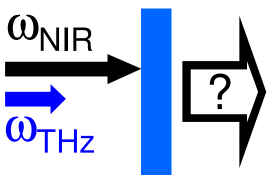


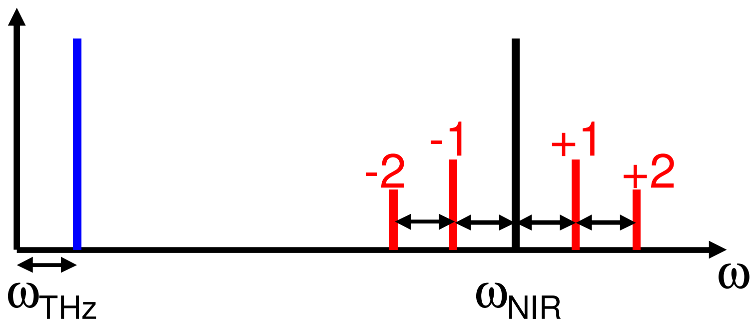
Nonlinear optics in the perturbative regime

$$P_i = \epsilon_0 \sum_j \chi^{(1)}_{ij} E_j + \epsilon_0 \sum_{jk} \chi^{(2)}_{ijk} E_j E_k + \epsilon_0 \sum_{jkl} \chi^{(3)}_{ijkl} E_j E_k E_l + \ldots$$

$$= P_i^{(1)} + P_i^{(2)} + P_i^{(3)} + \ldots$$

Mixing of NIR and THz waves: THz sidebands around NIR line





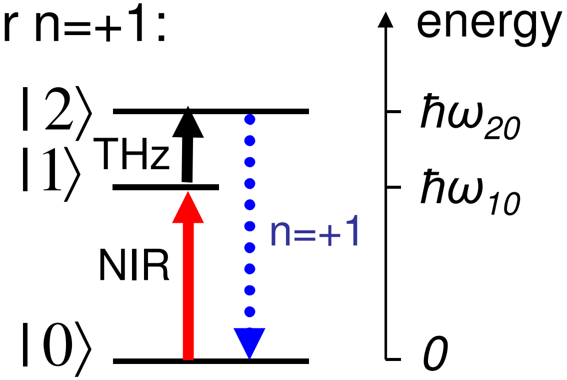
Efficient sidebands for resonant excitations, e.g. for n=+1:

$$P^{(2)} = \varepsilon_0 \chi^{(2)} E_{\mathrm{NIR}} E_{\mathrm{THz}}$$

matrix element

$$\chi^{(2)} \propto \sum_{m,\nu=1,2} \frac{\mu_{0m}^{NIR} \mu_{m\nu}^{THz} \mu_{\nu 0}^{n=+1}}{(\omega_{m0} - \omega_{NIR} - \omega_{THz} - i\gamma_{m0})(\omega_{\nu 0} - \omega_{NIR} - i\gamma_{\nu 0})}$$

line width

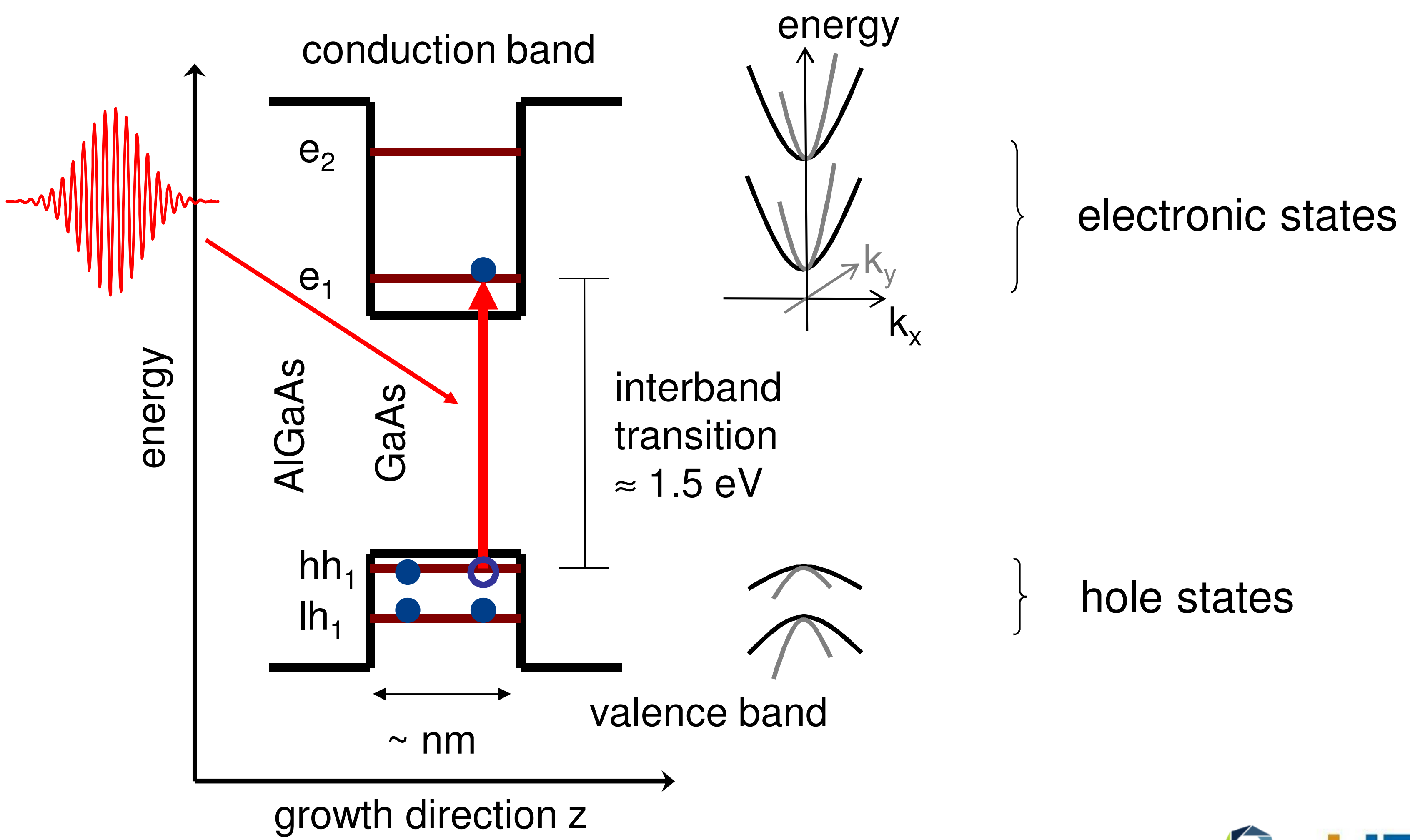


Folie 16

FZD-MA1 FZD Mitarbeiter; 30.03.2012

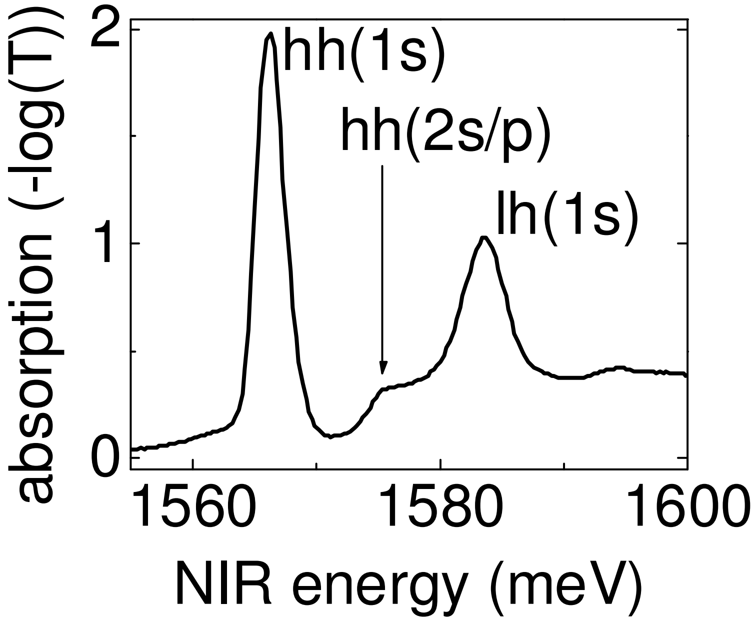
Electrons in quantum wells, interband excitation

- optical interband excitation



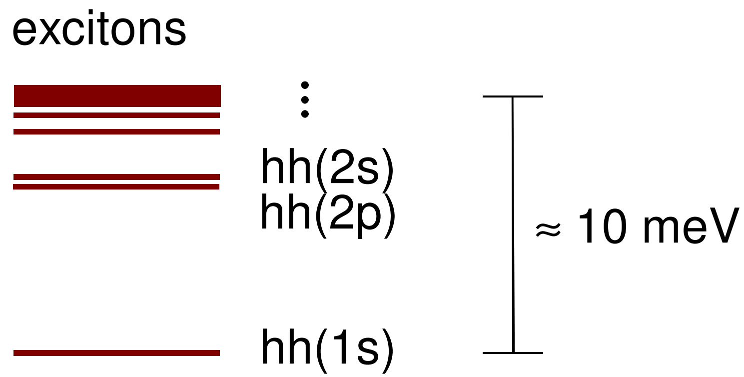
Excitonic energy spectrum

- example: near-infrared absorption



- hydrogen atom like energy structure with binding energies in the THz range

  (1 THz = 4.1 meV)



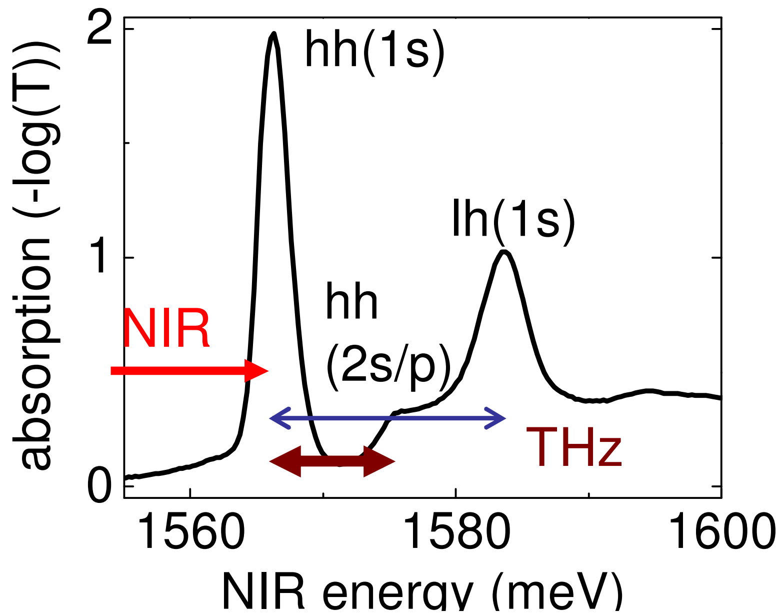
- selection rules:

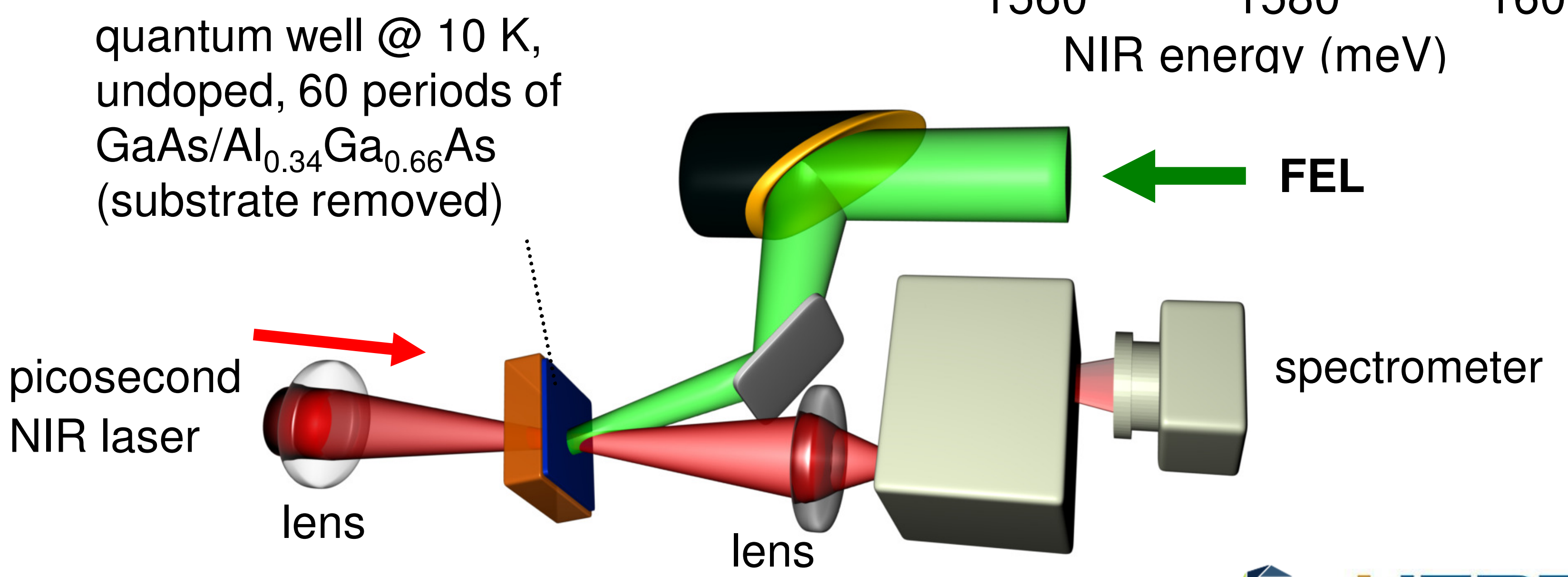
  - 2p state optically „dark“

  - intraexciton 1s-2p couples to THz

Sideband generation - Experiment

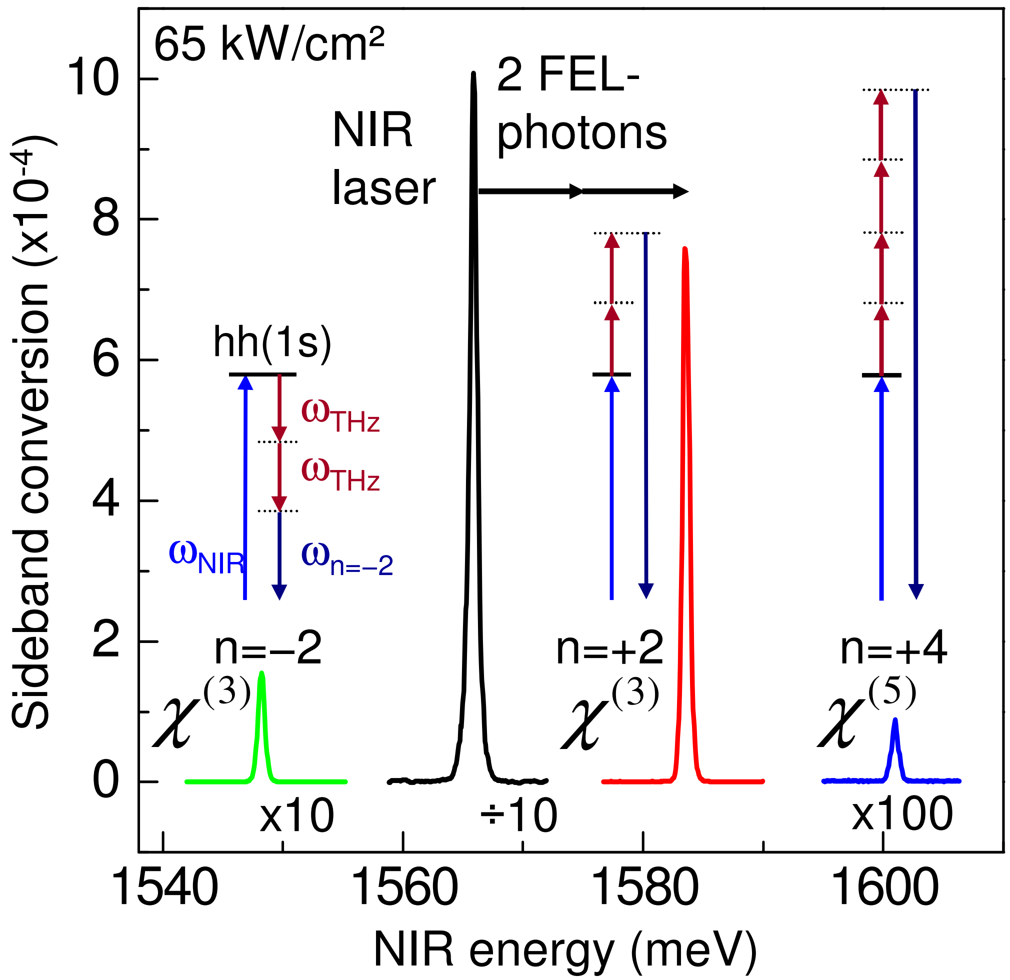
- *here*:
  intraexcitonic sideband generation using the 1s-2p transition



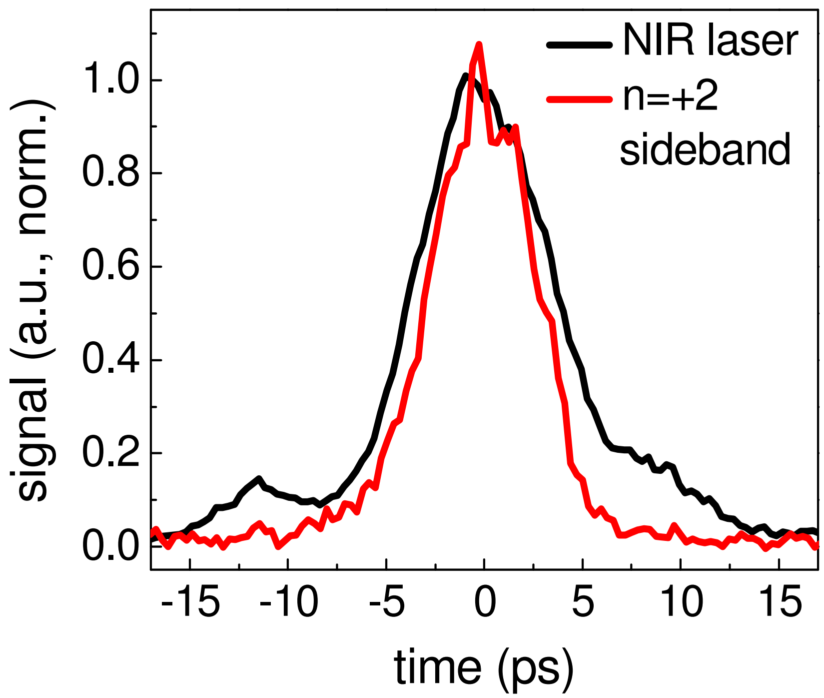


Results

- sidebands for NIR laser resonant with hh(1s) exciton and FEL resonant with 1s-2p transition:



- sidebands only at temporal overlap of NIR and THz pulses

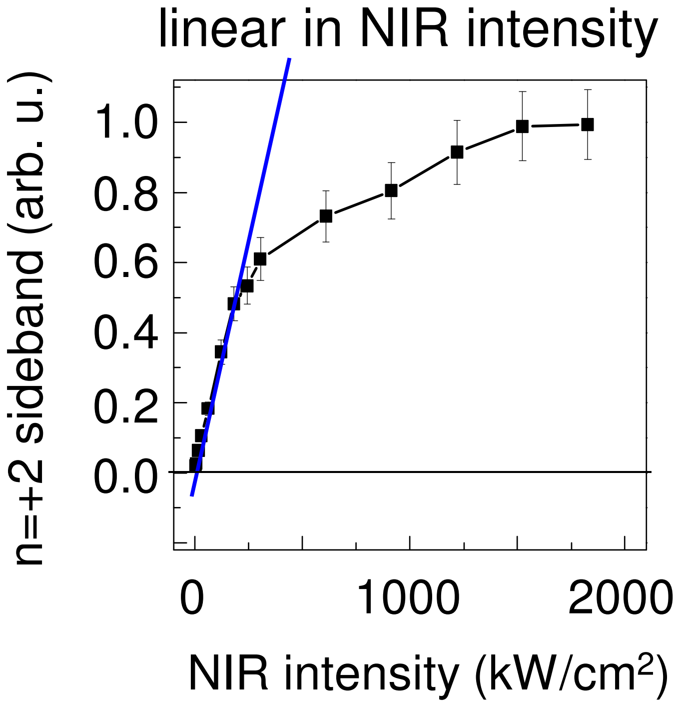


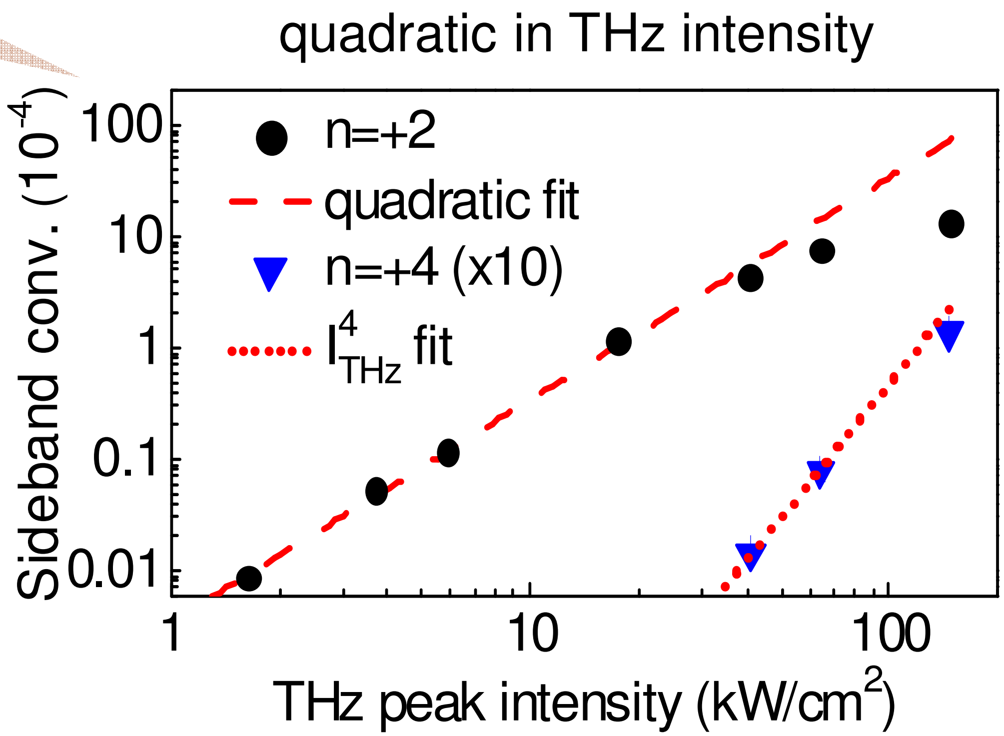
M. Wagner et al., APL **94**, 241105 (2009)

Results

- n=+2 sideband signal dependency on THz intensity as expected from

$$P^{(3)} = \varepsilon_0 \chi^{(3)} E_{\mathrm{NIR}} E_{\mathrm{FEL}}^2$$

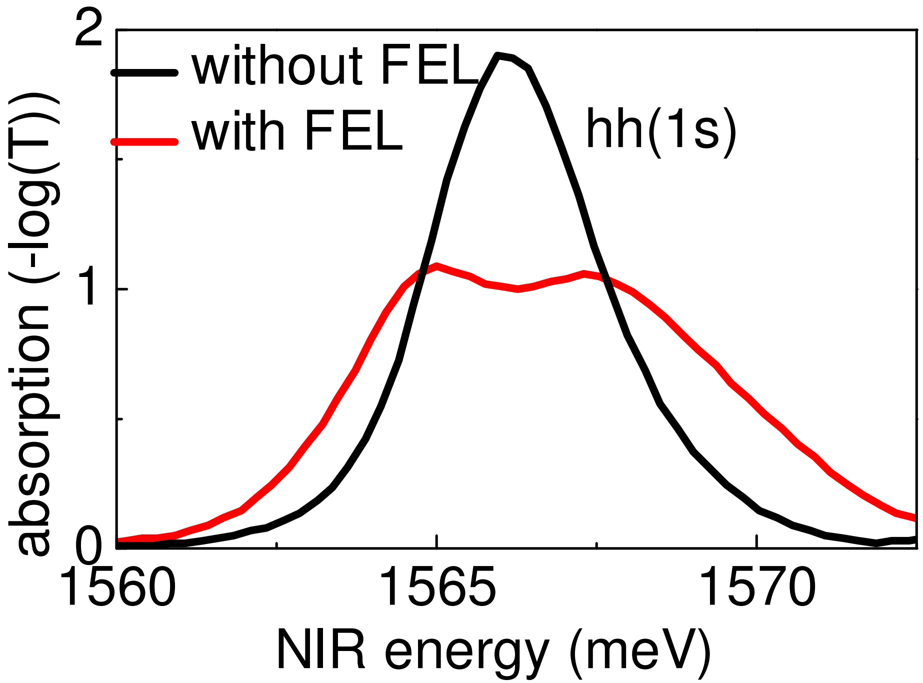




- **high conversion efficiency**
  > 0.1 % in 1.7 µm thin film

Possible application in transfer of THz signals, optical switches, wavelength division multiplexing...

Example for
nonperturbative nonlinear interaction:
Intraexcitonic Autler-Townes splitting

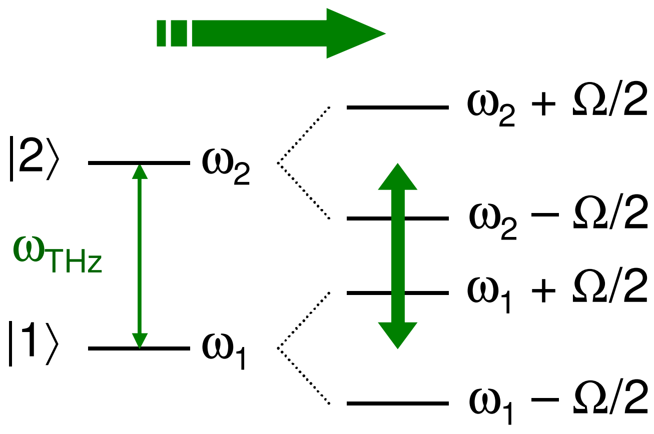


Nonlinear optics experiments - AC Stark effect

- simplest system to study light-matter interaction:
  two levels driven by intense light

Nonlinear optics experiments – Autler-Townes effect

- simplest system to study light-matter interaction: two levels driven by intense light



splitting given by Rabi frequency

$$\Omega = \left|\mu_{21} E_{THz}\right| / \hbar$$

$$\propto E_{THz}$$

→ mixed light-matter states or “dressed“ states
→ Autler-Townes* or AC Stark effect

*S. H. Autler & C. H. Townes, *Phys. Rev.* **100**, 703 (1955)

Nonlinear optics experiments - AC Stark effect

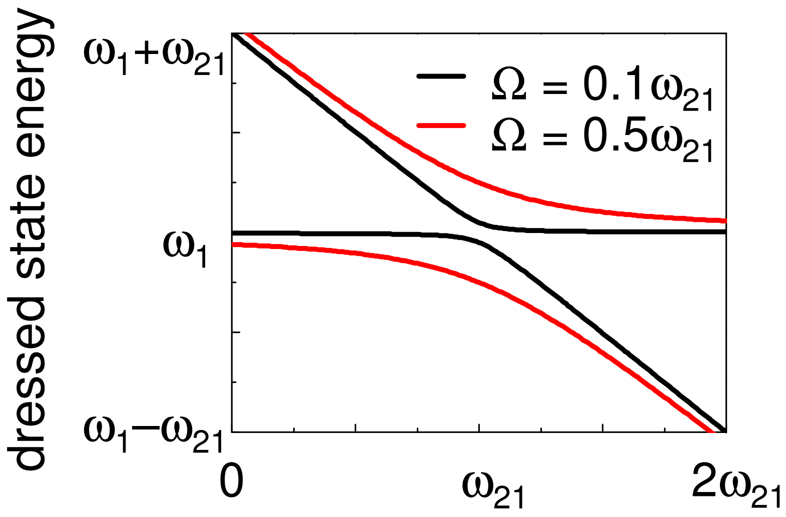
- simplest system to study light-matter interaction:
  two levels driven by intense light

- solution for two-levels:

$$\hbar\omega_1' = \hbar\left[\omega_1 - \frac{\Delta}{2} \pm \frac{1}{2}\sqrt{\Delta^2+\Omega^2}\right]$$
$$\hbar\omega_2' = \hbar\left[\omega_2 + \frac{\Delta}{2} \pm \frac{1}{2}\sqrt{\Delta^2+\Omega^2}\right]$$

with $\Delta = \omega\text{-}\omega_{21}$ and based on the **rotating-wave approximation**, i.e. $\Omega \ll \omega$

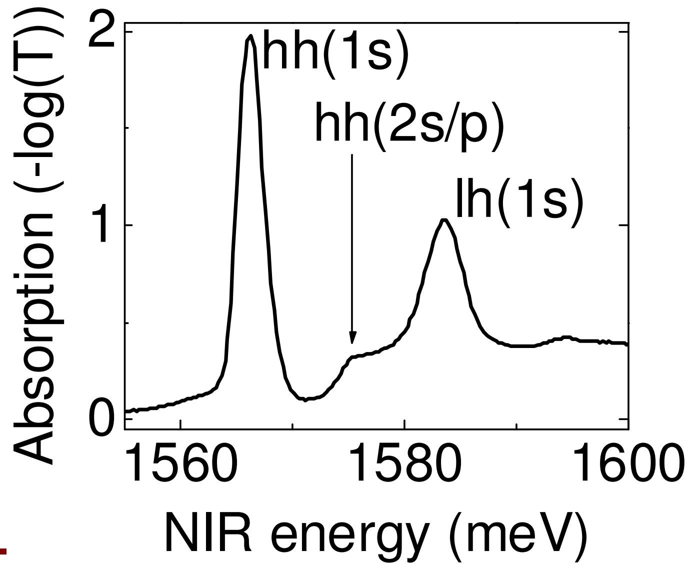
dressed states related to level $|1\rangle$

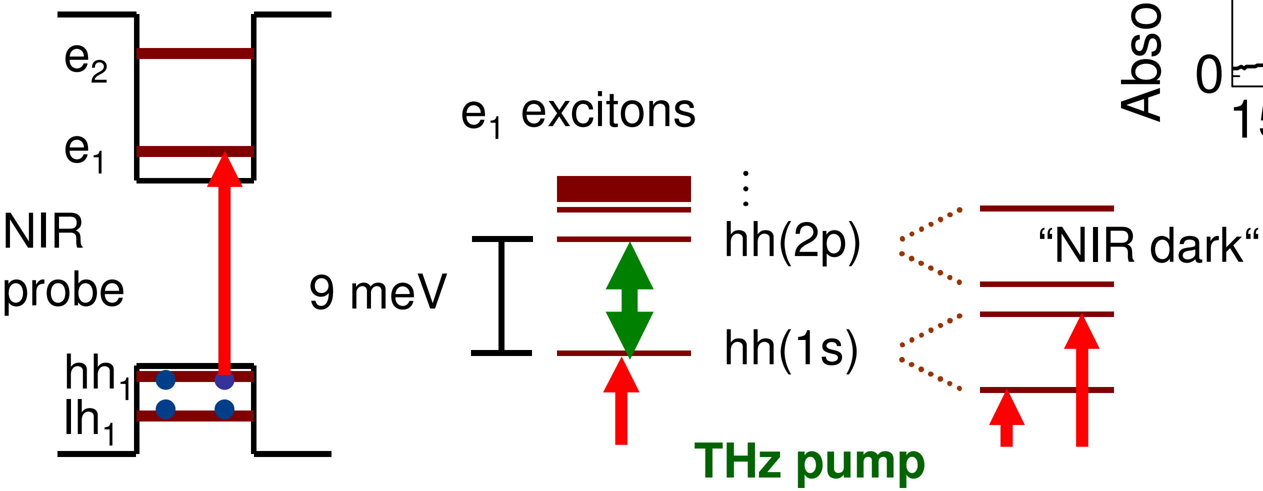


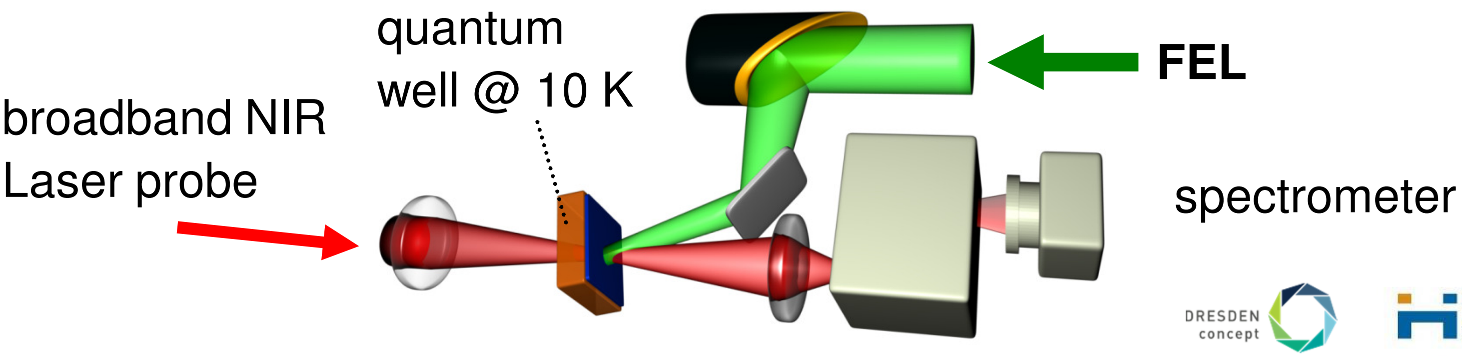
*S. H. Autler & C. H. Townes, *Phys. Rev.* **100**, 703 (1955)

AC Stark effect - Experiment

- *here*:
  intra-excitonic 1s-2p transition
  driven by a free-electron laser







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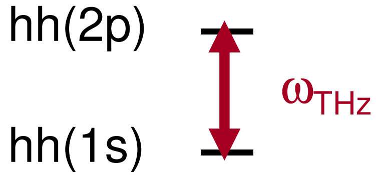
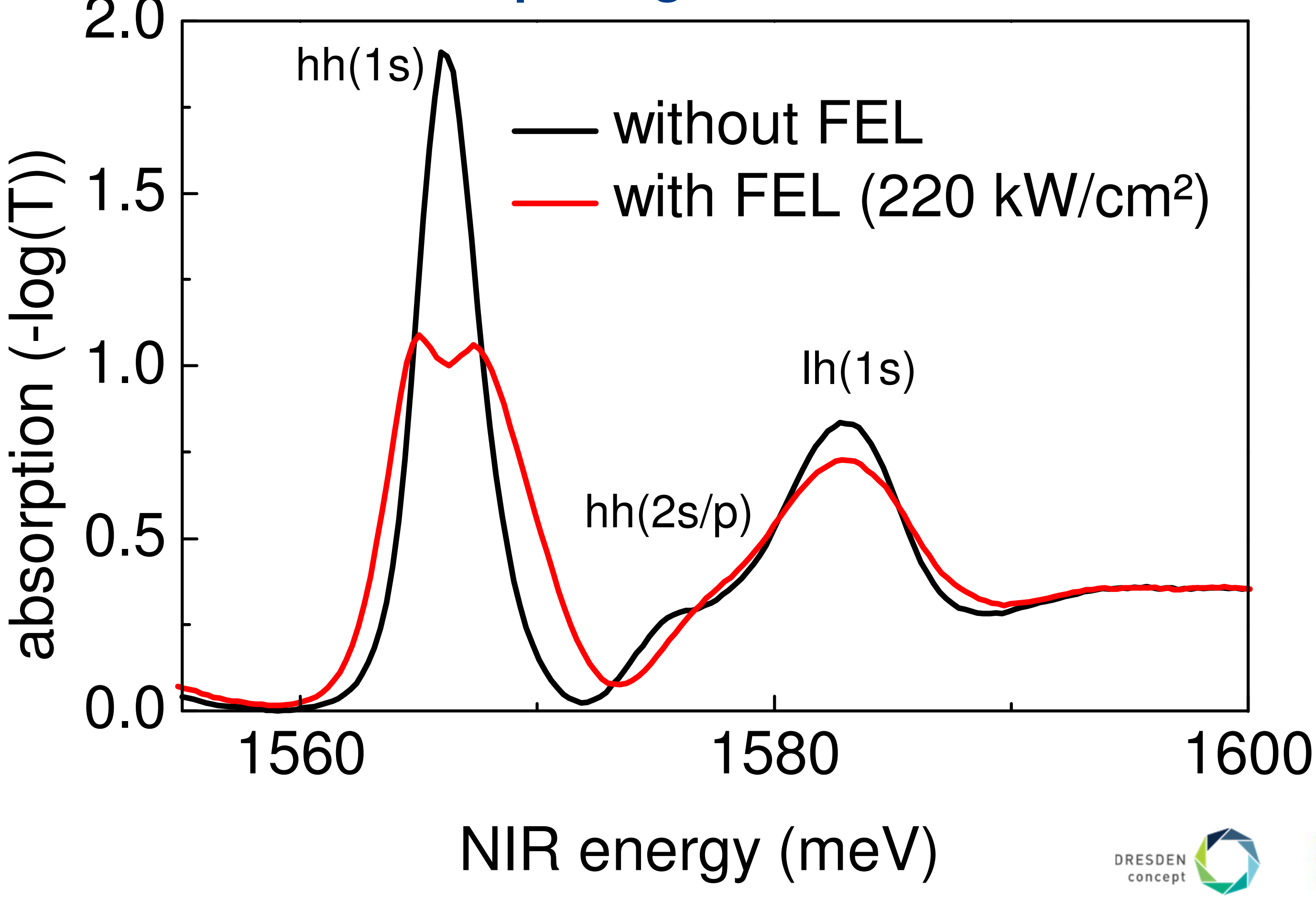
THz modifications of NIR absorption

- near resonance

  $\hbar\omega_{THz} = 10.5$ meV



clear splitting on resonance!



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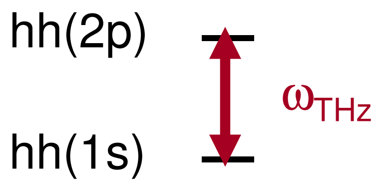

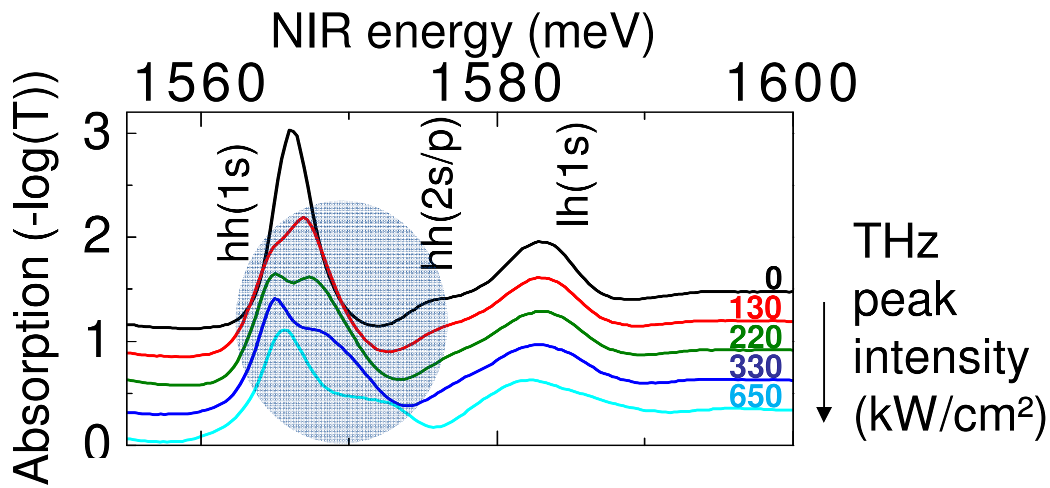

THz modifications of NIR absorption

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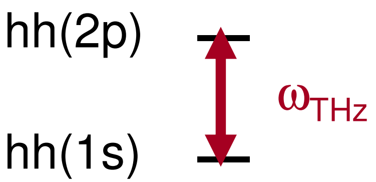
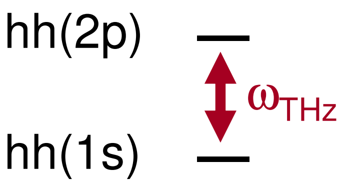
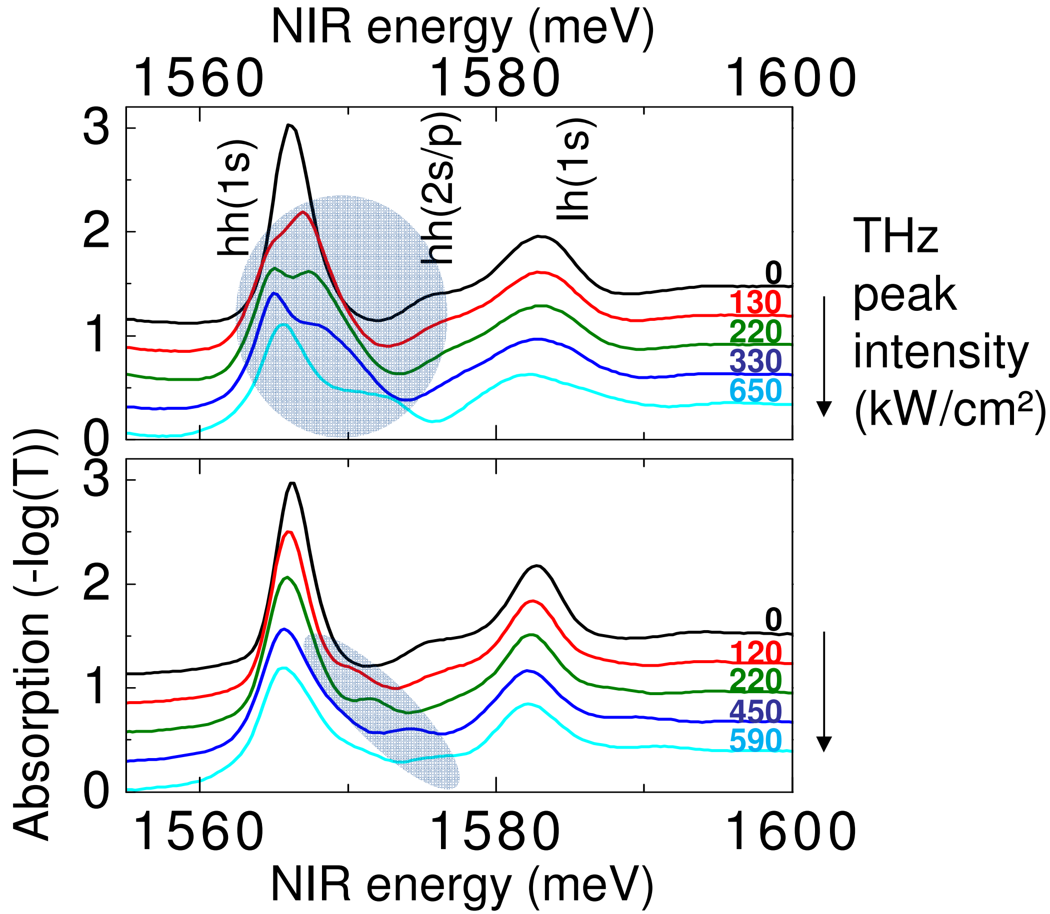


- below resonance

  $\hbar\omega_{THz} = 6.1$ meV

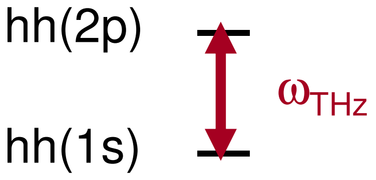




THz modifications of NIR absorption

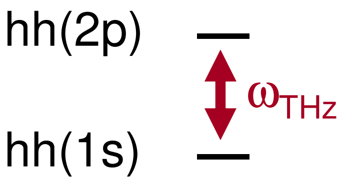
- near resonance

  $\hbar\omega_{THz}$= 10.5 meV



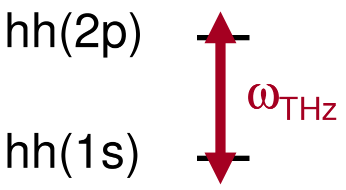
- below resonance

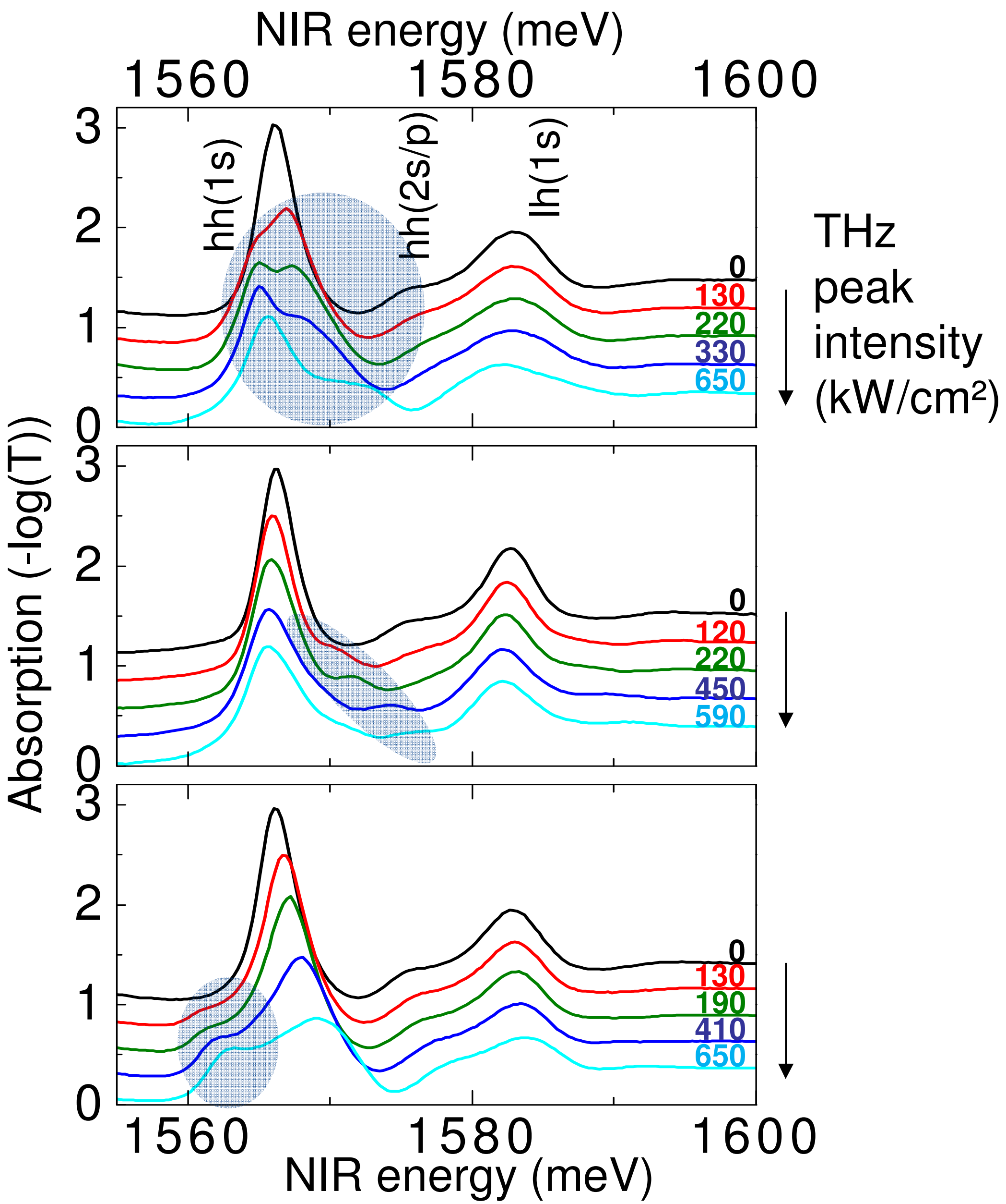
  $\hbar\omega_{THz}$= 6.1 meV



- above resonance

  $\hbar\omega_{THz}$= 14 meV

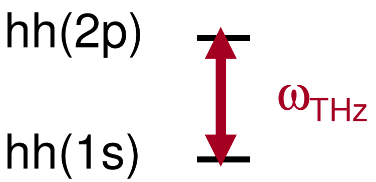




THz modifications of NIR absorption

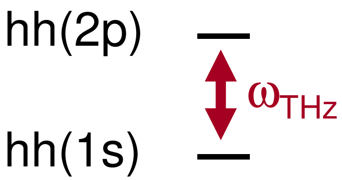
- near resonance

  $\hbar\omega_{THz}$= 10.5 meV



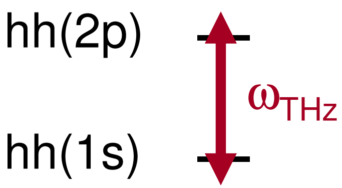
- below resonance

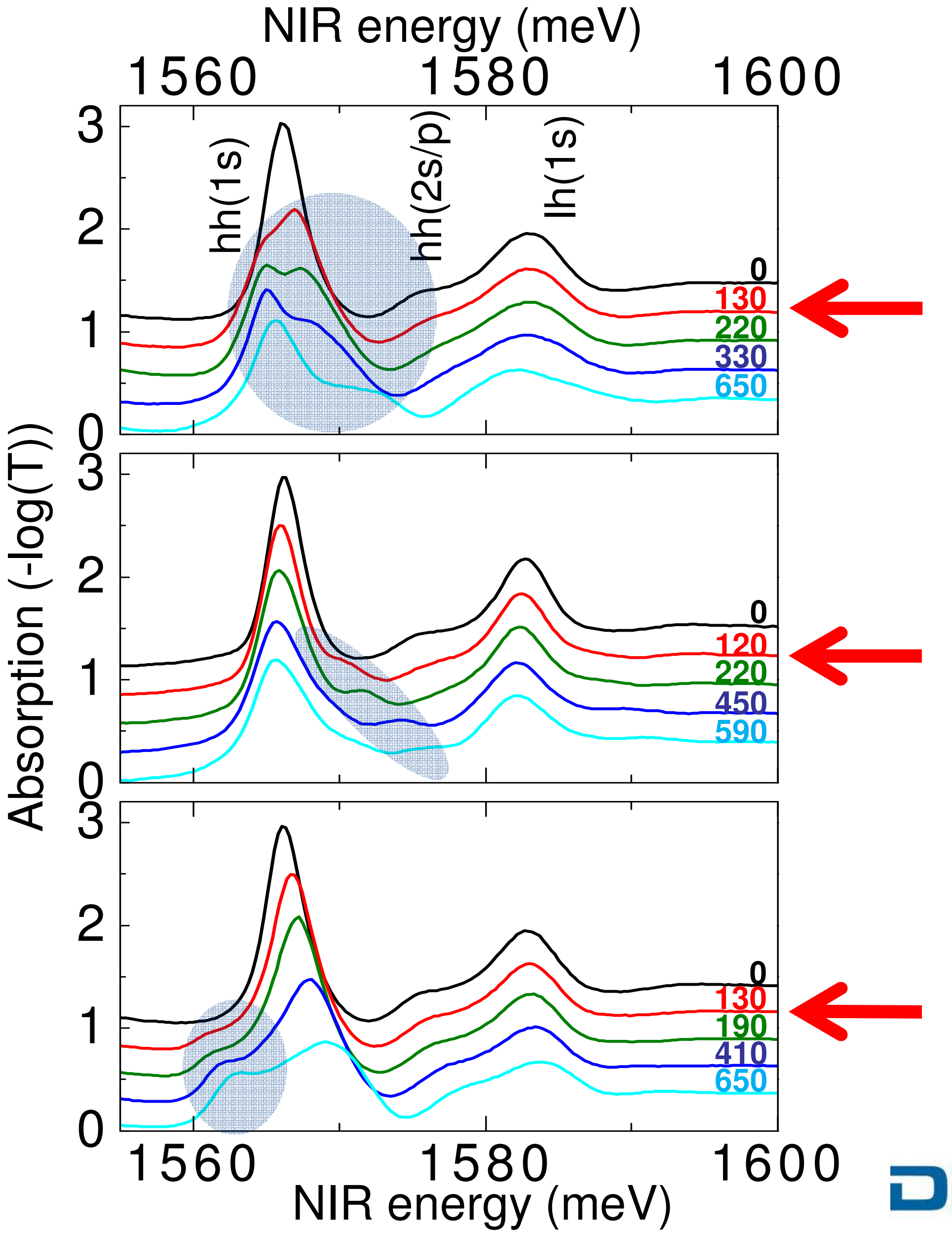
  $\hbar\omega_{THz}$= 6.1 meV



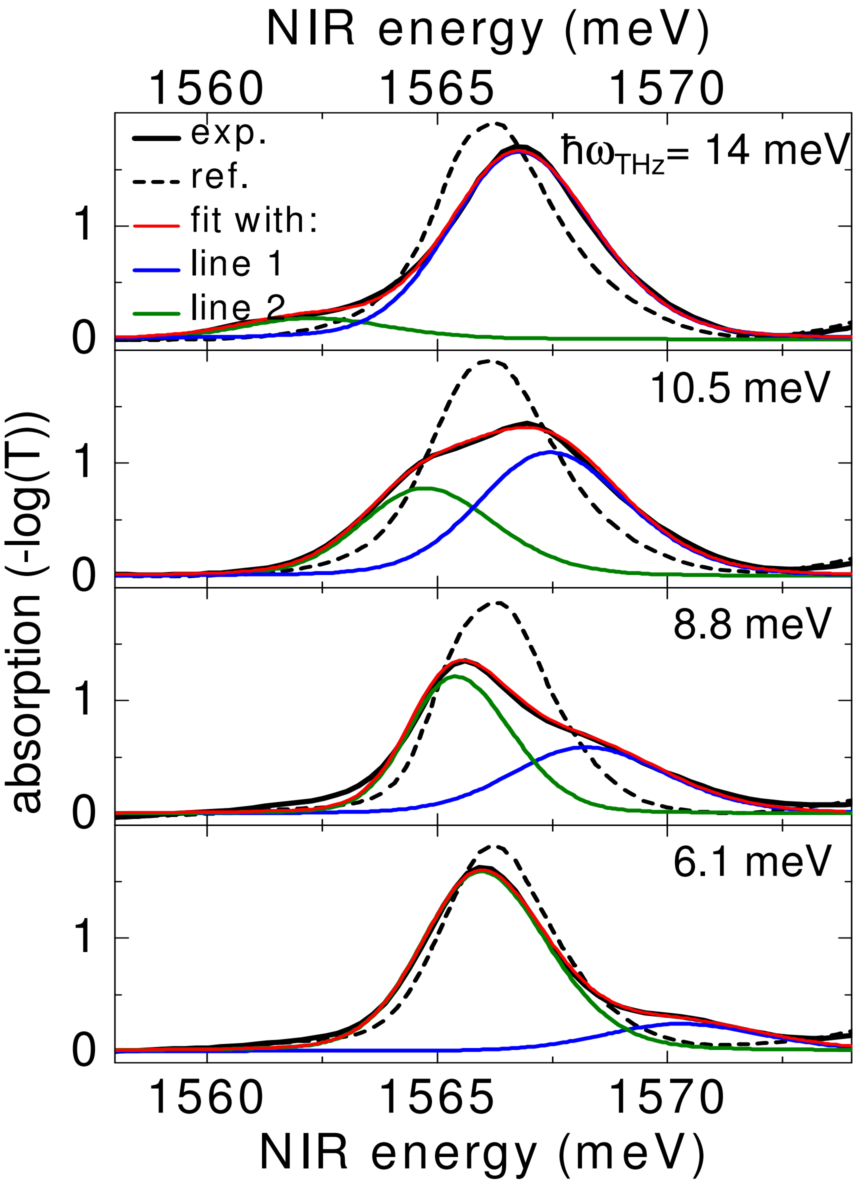
- above resonance

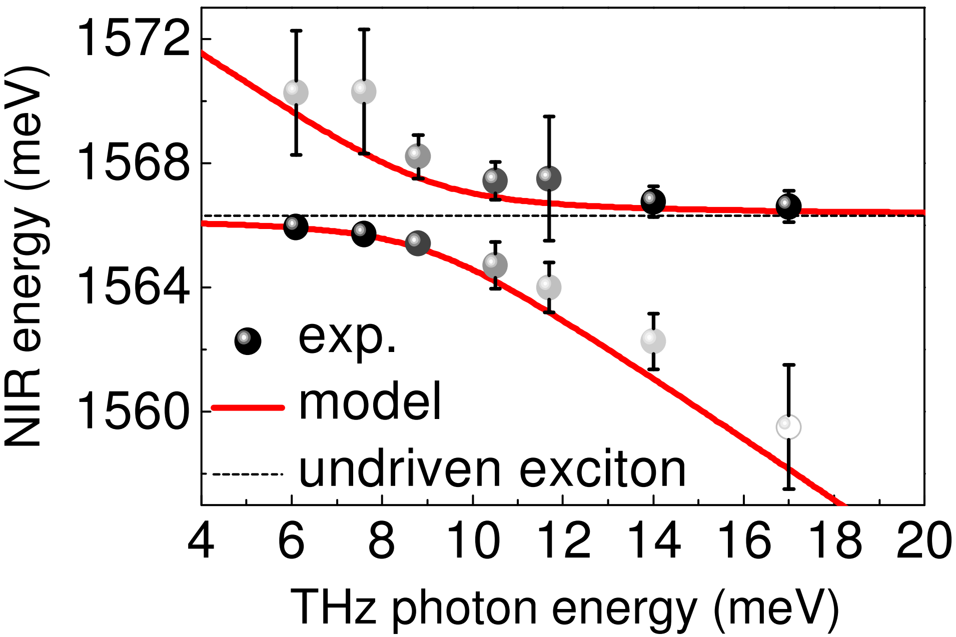
  $\hbar\omega_{THz}$= 14 meV





hh(1s) absorption for different THz photon energies





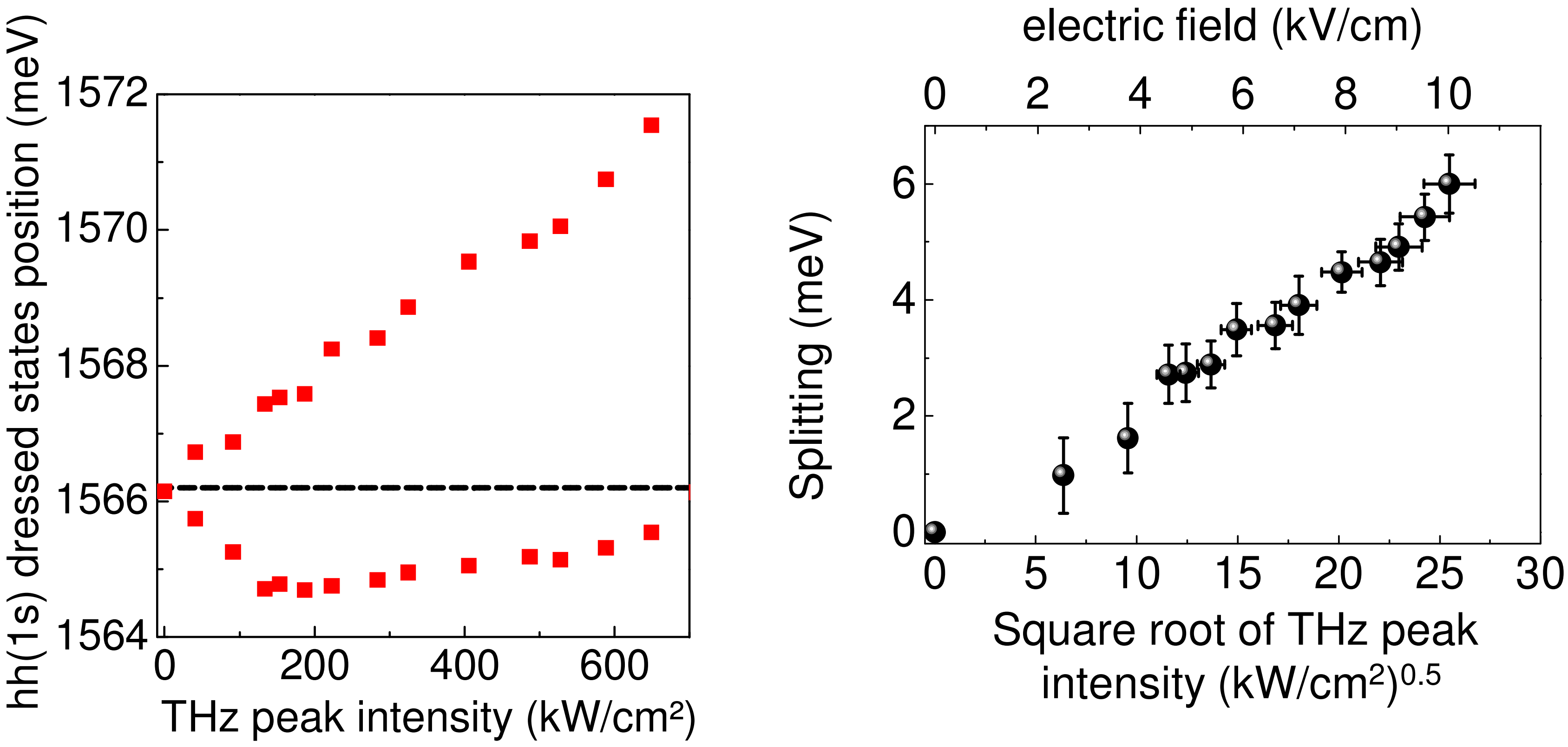
→ clear anticrossing

(130 kW/cm², 4.4 kV/cm)

at 1s-2p resonance (9 meV)

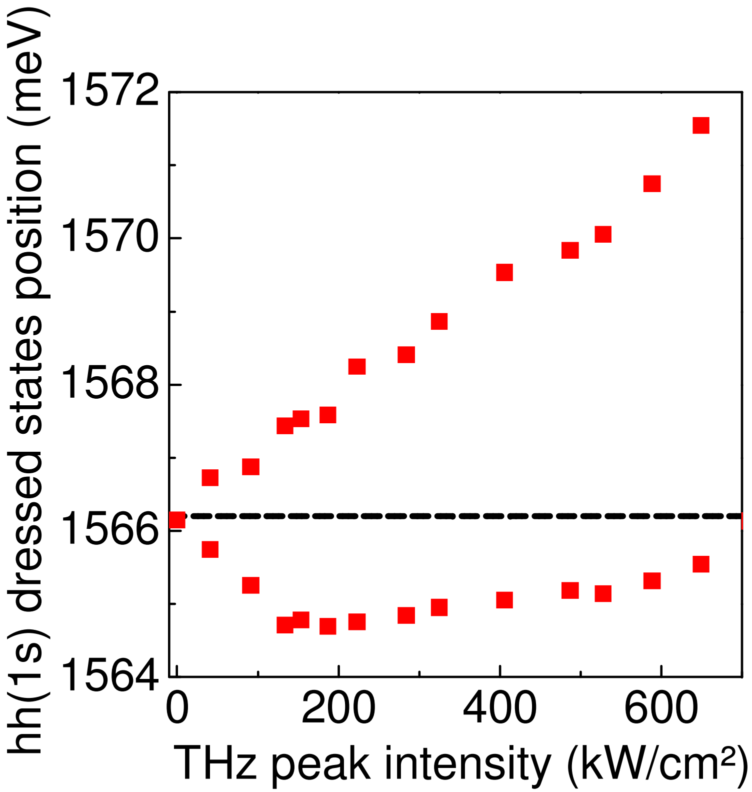
DRESDEN concept HZDR

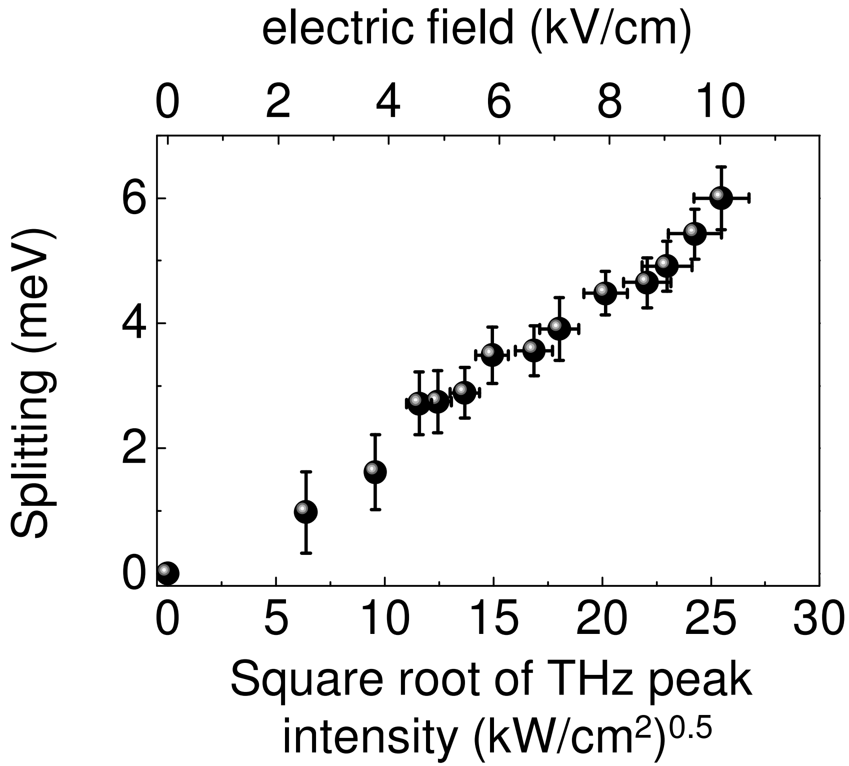
Splitting on resonance



- splitting increases linearly with field up to 650 kW/cm² (10 kV/cm)
- estimated splitting (from matrix element) @ 200 kW/cm², 5.4 kV/cm: 2.4 meV compared to 3 meV measured → good agreement

Splitting on resonance



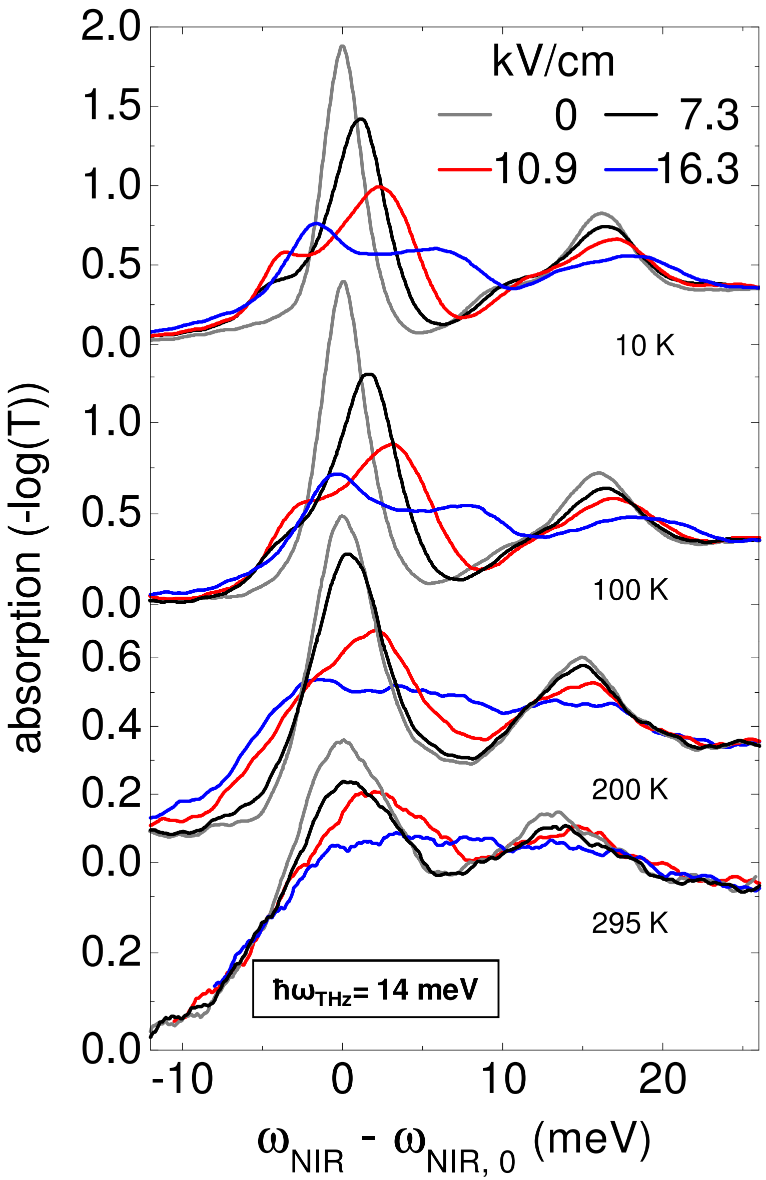


- important values @ 10 kV/cm:
  - 1s-2p transition energy 9 meV
  - THz photon energy 10.5 meV
  - splitting 6 meV
  - ponderomotive energy $U_p = 3$ meV

M. Wagner *et al.*, *PRL* **105**, 167401 (2010)

DRESDEN concept HZDR

Temperature dependence



Substantial NIR transmission change (at 200 K still threefold NIR transmission change)

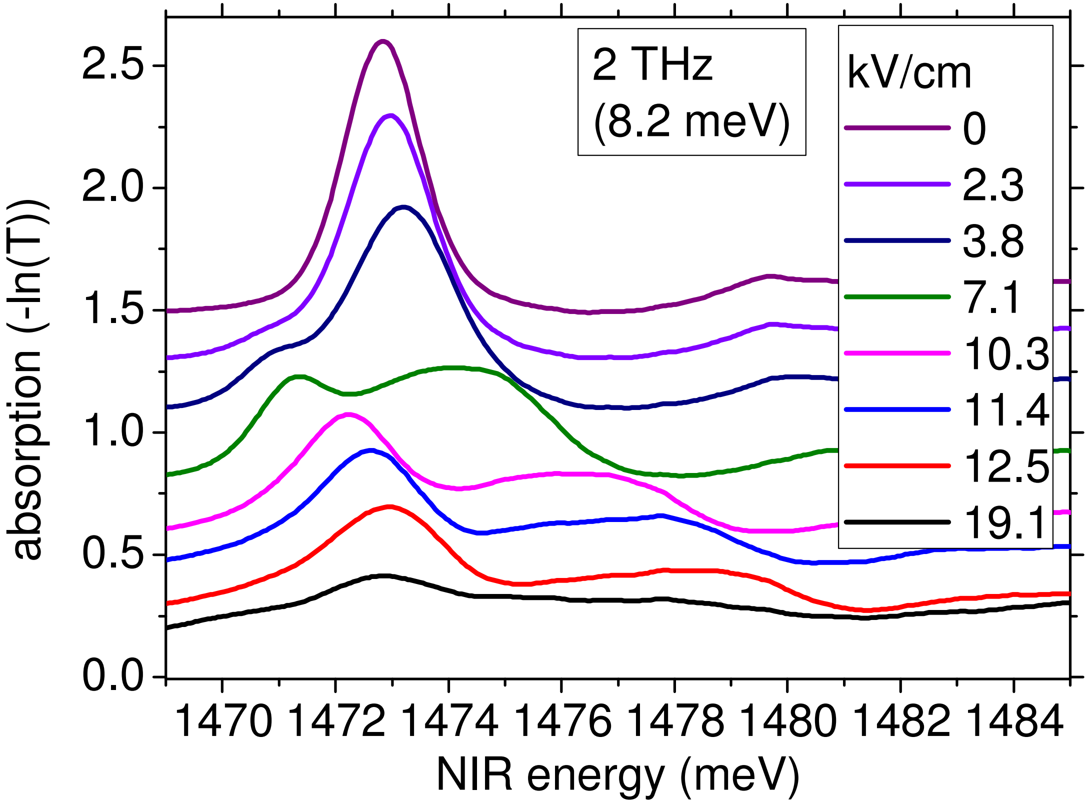
→ optical modulator application with Peltier-cooling

Poster:
Martin Teich

M. Wagner et al., APL **100**, 051109 (2012)

Stephan Winnerl | Institute of Ion Beam Physics and Materials Research | Spectroscopy Devision

What happens at even higher intensities (up to 1.75 MW/cm^2)



InGaAs quantum wells
With narrow linewidth

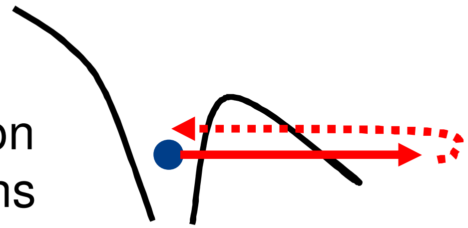
Features not explained by two-level model and RWA:
- relative peak intensities
- overall blueshift
- broadening of peaks

Here: extreme nonlinear optics accessible beyond rotating-wave regime, where

$$U_p \approx E_{binding} \approx h\omega_{THz} \approx \Omega$$

Outlook:

- high-harmonic generation
- field ionization of excitons



B. Zaks et al., Nature **483**, 580 (2012)
E. Ewers et al., Phys. Rev. B **85**, 075307 (2012)

Summary

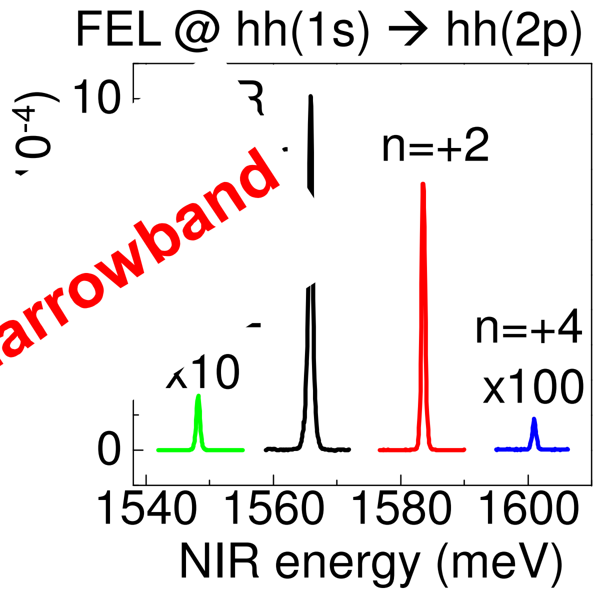
- Pump-probe spectroscopy on graphene
  - saturation of the signals
  - population dynamics

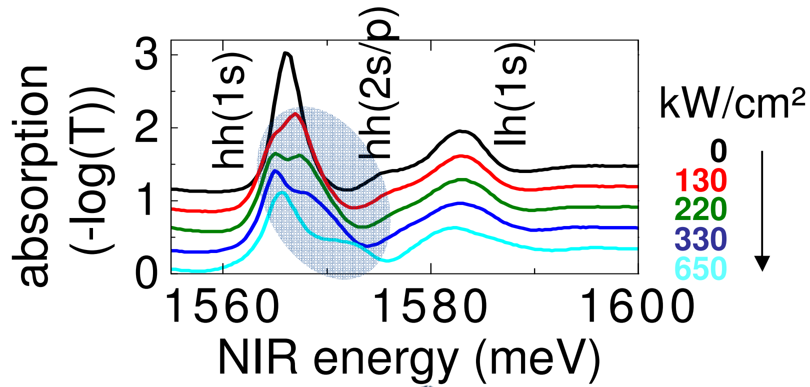
- Efficient sideband generation
  - perturbative nonlinear optics

- Autler-Townes splittinc
  even at elevated tc

- extreme nonlinear o… …cessible
  beyond rotating-wave …egime where
  $U_p \approx E_{binding} \approx h\omega_{THz} \approx \Omega$

FEL: tunable, intense and narrowband source





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HZDR

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G. Strasser

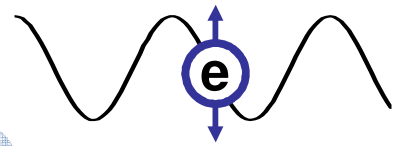
Georgia Tech

M. Sprinkle
C. Berger
W. A. de Heer

Deutsche Forschungsgemeinschaft

Motivation

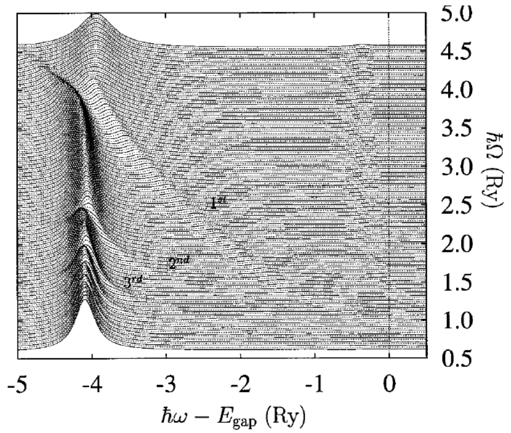
energy/frequency ranges in light-matter coupling



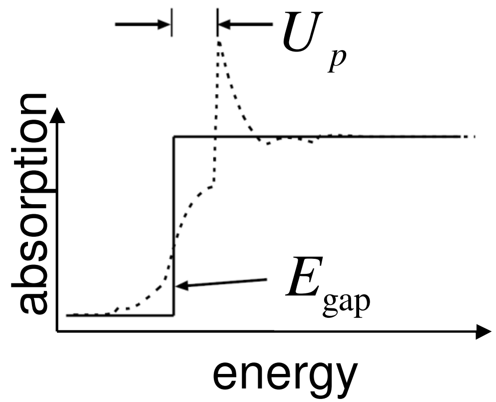
- ponderomotive energy ~ THz photon energy

$$\frac{1}{T}\int_0^T\left(\frac{1}{2}mv^2\right)dt = \frac{e^2E_0^2}{4m\omega^2} = U_p \sim \hbar\omega \quad \leftrightarrow \quad \frac{e^2E_0^2}{4m\hbar\omega^3} \sim 1$$

→ AC Stark effect, Rabi oscillations sidebands (require resonant driving) | dynamical Franz-Keldysh effect



K. Johnsen and A. P. Jauho, *PRL* **83**, 1207 (1999)



A. P. Jauho & K. Johnsen, *PRL* **76**, 4576 (1996)

K. Nordstrom *et al.*, *PRL* **81**, 457 (1998)

→ conditions easily met at **THz frequencies** (1 THz = 4.1 meV) for **excitons** as artifical atoms with $E_{binding} \approx 10\,\text{meV}$

- even extreme nonlinear optics possible:
  Rabi energy $\hbar\Omega = \mu E_{THz} \sim \hbar\omega_{THz}$

O. D. Mücke *et al.*, *PRL* **87**, 057401 (2001)

HZDR