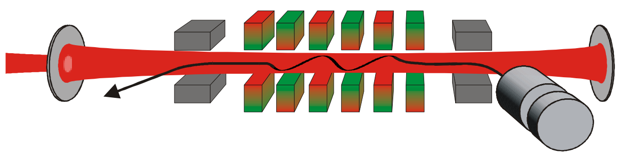
THz2010

International Workshop on

Terahertz spectroscopy and its high-field applications

Forschungszentrum Dresden-Rossendorf (FZD)
Dresden, Germany

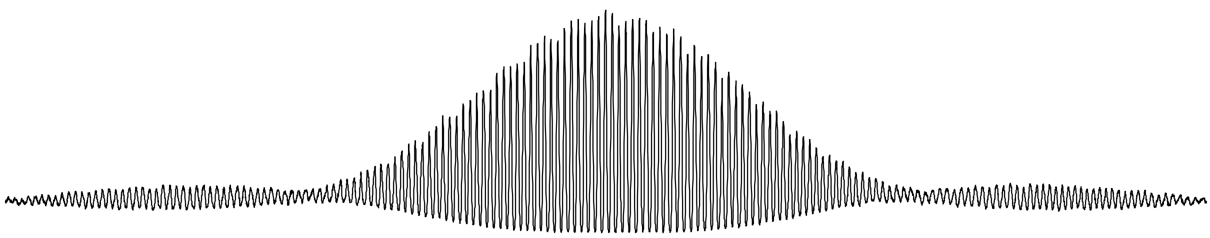
14 – 15 June 2010



Programme

Invited Abstracts

Poster Abstracts



Workshop organizers

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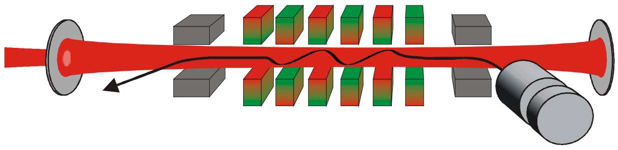

Goal and background:

The goal of this workshop is to bring together researchers working in the field of THz spectroscopy, mainly of solids. This includes spectroscopy using short-pulse, high-power sources as well as broad-band sources, but also microscopy and imaging. A further focus will be THz spectroscopy in high magnetic fields (including high-frequency / high-field ESR).

A motivation for this workshop relates to the fact that at FZD the short-pulse THz/infrared free-electron laser FELBE is being operated as a user facility (funded as a transnational access facility in the EU project ELISA), and is in fact connected to the Dresden pulsed high-magnetic field laboratory HLD (funded as a transnational access facility in the EU project EuroMagNET II). In addition, it is planned to extend the facilities with an accelerator-based broad-band high-power source of coherent THz radiation within the next few years. A further important aspect relates to the fact that presently a new THz free-electron laser (FLARE) is being constructed at the high-magnetic field lab in Nijmegen.

The THz workshop will actually take place the first two days (14-15 June), while the third day (16 June) will be devoted to a EuroMagNET II user meeting, covering all aspects of high-magnetic-field experiments (not only spectroscopy). The first day (14 June) represents a FELBE user meeting as well.

FELBE

Programme

Monday, 14 June 2010:

8:30 Welcome (R. Sauerbrey / M. Helm)

8:40 Dominik Stehr (FZD, Dresden)
The Dresden Free Electron Lasers and future broad-band THz sources

9:00 Rupert Huber (Univ. Konstanz)
THz nonlinear interaction and quantum optics in the sub-cycle regime

9:30 Jure Demsar (Univ. Konstanz)
Dynamics of superconductors driven out of equilibrium by femtosecond optical pulses

10:00 Coffee Break

10:30 Daniele Fausti (CFEL Hamburg):
THz induced phase transitions in manganites

11:00 Paolo Calvani (Univ. La Sapienza, Rome, Italy):
Sub-terahertz spectroscopy in superconductors and charge-ordered materials

11:30 Mischa Bonn (FOM Amsterdam, NL):
THz Studies of dynamics of water around protons and ions

12:00 Erik Bründermann (Univ. Bochum):
Micro-spectroscopy and chemical nanoscopy using infrared and THz radiation

12:30 Lunch provided next to the conference room

13:45 Tobias Kampfrath (FHI Berlin):
THz pulses as probe and driving force of electron and spin excitations

14:15 Mark Sherwin (UCSB, USA):
Electric and magnetic resonance at terahertz frequencies

14:45 Tomas Room (Nat. Inst. Chem. Phys. and Biophysics, Tallinn, Estonia):
Broad-band THz spectroscopy in magnetic fields: applications to the study of multiferroics

15:15 Alexander Schnegg (HZB Berlin):
EPR studies on materials relevant for solar energy conversion: the EPR-solar approach

15:45 Coffee break

16:15 Karsten Holldack (HZB Berlin):
Ultrafast resistive switching in magnetite employing coherent synchrotron radiation at THz frequencies

16:45 Michael Gensch (HZB Berlin):
Coherent THz pulses from linear accelerators: challenges and opportunities

17:15 Hans Sigg (PSI Villigen, Switzerland):
Broadband far- and mid-infrared pump-probe spectroscopy using synchrotron radiation

17:45 Karl Unterrainer (TU Vienna, Austria) :
THz quantum cascade lasers: confinement and dynamics

19:00 Dinner in the restaurant “Fischhaus”, Fischhausstraße 14, 01099 Dresden (transportation by coach provided)

Tuesday, 15 June 2010:

8:30 Andrei Pimenov (Univ. Würzburg):
Magnetic and magnetoelectric excitations in multiferroic manganites

9:00 Hitoshi Ohta (Kobe University, Japan):
Developments of THz ESR systems using a micro-cantilever

9:30 Vladik Kataev (IFW Dresden):
Probing collective spin states in cubic cobaltates by high-frequency ESR spectroscopy

10:00 Coffee break

10:30 Paul Planken (TU Delft, NL):
Imaging the THz electric near-field of sub-wavelength metal structures

11:00 Marek Potemski (LNCMI Grenoble, F):
Infrared and THz magnetospectroscopy of graphene

11:30 Andrei Zvyagin (TU Dresden and ILTP Kharkov, Ukraine):
Features of the high-frequency ESR in one-dimensional quantum spin systems

12:00 Roberta Sessoli (Univ. Firenze, Italy):
High field EPR of single molecule magnets

12:30 Lunch provided next to the conference room

13:45 Hans Engelkamp (Radboud Univ., Nijmegen, NL):
Far infrared spectroscopy at the HFML

14.15 Anne-Laure Barra (LNCMI Grenoble, F):
High frequency EPR studies of iron spin clusters

14.45 Michel Goiran (LNCMI Toulouse, F):
ESR in the THz range, recent results and experimental developments

15:15 Sergei Zvyagin (FZD, Dresden):
High-frequency and high-field ESR in quantum spin systems

15:45 Coffee break

(16:00 EuroMagNET II user selection committee)

16:00 Lab visits ELBE / FEL & HLD

17:00 Poster session

18:30 Dinner at FZD

20:00 Coach to the hotels & city provided

List of posters

1. **Cyclotron resonance in InAs/AlSb QW heterostructures in ultraquantum limit**
A. Ikonnikov, K. Spirin, O. Drachenko*, S. Krishtopenko, V. Gavrilenko, Yu. Sadofyev, M. Helm*
*Institute for Physics of Microstructures RAS, *Forschungszentrum Dresden-Rossendorf*

2. **Terahertz time-domain cyclotron resonance spectroscopy in pulsed magnetic field**
Daniel Molter[a], Sylvie George[b], Michel Goiran[b], Fritz Keilmann[c], Rene Beigang[a], and Jean Leotin[b]
[a]*Fraunhofer Institute for Physical Measurement Techniques IPM, Kaiserslautern, Germany*
[b]*Laboratoire National des Champs Magnétiques Intenses, Toulouse, France*
[c]*Max Planck Institute of Quantum Optics, Garching, Germany*

3. **THz response of a two-dimensional electron gas**
Marco Reuter, Torben Grunwald, Sangam Chatterjee, Daniel Golde, Mackillo Kira, and Stephan W. Koch
Faculty of Physics, Philipps-Universität Marburg, Renthof 5, 35032 Marburg, Germany

4. **Coherent broadband continuous-wave THz spectrometry: A powerful tool for low-energy solid-state spectroscopy**
H. Schmitz[1], K. Thirunavukkuarasu[1] 1, A. Roggenbuck[1,2], A. Janssen[1], A. Deninger[2], I. Camara Mayorga[3], J. Hemberger[1], R. Güsten[3], and M. Grüninger[1]
[1]*II. Physikalisches Institut, Universität zu Köln, D-50937 Köln, Germany*
[2]*TOPTICA Photonics AG, Lochhamer Schlag 19, D-82166 Gräfelfing, Germany*
[3]*Max-Planck Institute for Radio Astronomy, Auf dem Hügel 69, D-53121 Bonn, Germany*

5. **Ultrafast saturable absorption in doped semiconductors at high THz field strengths**
Dmitry Turchinovich[1] and Matthias C. Hoffmann[2]
[1]*DTU Fotonik - Department of Photonics Engineering, Technical University of Denmark, DK-2800 Kgs. Lyngby, Denmark*
[2]*Max Planck Research Department for Structural Dynamics, University of Hamburg, CFEL, 22607 Hamburg, Germany*

6. **Investigation of MIR-pumped quantum-cascade structures as emitters of THz radiation**
M.Wienold[1], M. Giehler[1], L. Schrottke[1], R. Hey[1], S. Winnerl[2], H. Schneider[2], S. G. Pavlov[3], and H. T. Grahn[1]
[1]*Paul Drude Institut für Festkörperelektronik, Hausvogteiplatz 5-7, 10117 Berlin*
[2]*Institute of Ion Beam Physics and Materials Research, Forschungszentrum Dresden-Rossendorf, P. O. Box 510119, 01314 Dresden*
[3]*German Aerospace Center (DLR), Institute of Planetary Research, Rutherfordstr. 2, 12489 Berlin*

7. **1.55 µm photoconductive THz emitters based on ErAs:$In_{0.53}Ga_{0.47}As$ superlattices**
Z.-Y. Zhao, A. Schwagmann, F. Ospald, K. von Klitzing, and J. H. Smet
Max-Planck-Institut für Festkörperforschung Stuttgart, Germany
D. C. Driscoll, M. P. Hanson, H. Lu, and A. C. Gossard
Materials Department, University of California-Santa Barbara, Santa Barbara, USA

8. **Optical conductivity of $LuNi_2B_2C$ in the terahertz range**
T. Fischer[1], A. V. Pronin[1], J. Wosnitza[1], T. Niemeier[2], and B. Holzapfel[2]
[1]Hochfeld-Magnetlabor Dresden (HLD), FZ Dresden-Rossendorf, 01314 Dresden, Germany
[2]Leibniz-Institut für Festkörper- und Werkstoffforschung, 01171 Dresden, Germany

9. **THz-range free-electron laser ESR spectroscopy: techniques and applications in high magnetic fields**
M. Ozerov, E. Cizmar, D. Kamenskyi, S. Zherlitsyn, T. Herrmannsdörfer, J. Wosnitza, and S. A. Zvyagin
Hochfeld-Magnetlabor Dresden (HLD), Forschungszentrum Dresden-Rossendorf (FZD), Dresden, Germany
R. Wünsch and W. Seidel
Institute of Radiation Physics, Forschungszentrum Dresden-Rossendorf (FZD), Dresden, Germany

10. **Fano interference in the intersubband THz response of semiconductor quantum wells after interband photoexcitation**
M. Wagner[1], D. Golde[2], D. Stehr[1], H. Schneider[1], M. Helm[1], A. M. Andrews[3], T. Roch[3], G. Strasser[3], M. Kira[2] and S. W. Koch[2]
[1]Institute of Ion Beam Physics and Materials Research, Forschungszentrum Dresden-Rossendorf, P.O. Box 510119, 01314 Dresden, Germany
[2]Department of Physics and Materials Sciences Center, Philipps-University, Renthof 5, 35032 Marburg, Germany
[3]Micro- and Nanostructure Center, TU Wien, Floragasse 7, 1040 Vienna, Austria

11. **THz sideband generation using intraexcitonic transitions in GaAs/AlGaAs quantum wells**
M. Wagner[1], H. Schneider[1], S. Winnerl[1], M. Helm[1], T. Roch[2], A. M. Andrews[2], S. Schartner[2], and G. Strasser[2]
[1]Institute of Ion Beam Physics and Materials Research, Forschungszentrum Dresden-Rossendorf, P.O. Box 510119, 01314 Dresden, Germany
[2]Micro- and Nanostructure Center, TU Wien, Floragasse 7, 1040 Vienna, Austria

12. **The spatial structure of longitudinal and transversal fields in focussed terahertz beams**
M. Mittendorff, S. Winnerl, R. Hubrich, F. Peter, H. Schneider, and M. Helm
Forschungszentrum Dresden-Rossendorf, Postfach 510119, 01314 Dresden, Germany

13. **Carrier dynamics in graphene excited at low energy**
S. Winnerl[1], M. Orlita[2], P. Plochocka[2], H. Schneider[1], P. Kossacki[2], M. Potemski[2], O. Drachenko[1], M. Sprinkle[3], C. Berger[3], W. A. de Heer[3], and M. Helm[1]
[1]Forschungszentrum Dresden-Rossendorf, Dresden, Germany,
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Invited Abstracts

The Dresden Free Electron Lasers and future broad-band THz sources

Dominik Stehr[1], Stephan Winnerl[1], Wolfgang Seidel[2], Harald Schneider[1], Peter Michel[2] and Manfred Helm[1]
[1] *Institute of Ion Beam Physics and Materials Research, Forschungszentrum Dresden-Rossendorf, Dresden, Germany*
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Since achievement of first lasing in May 2004 the Dresden Free Electron Lasers at the ELBE accelerator have undergone a systematic development towards a highly versatile research instrument, enabling research in the mid-infrared and THz region. Specifically the long wavelength FEL covers a wavelength range that is hardly accessible with table-top laser sources. Thanks to the use of a superconducting RF linac, the Dresden FELs deliver picosecond pulses not only with high peak power but also at high average powers of up to 30 W. These properties make FELBE highly suitable for experimental studies where small signals need to be measured at moderate intensities or a large average power is needed as for imaging experiments.

In this presentation we will give an overview of the experimental techniques that are currently available at FELBE and present a short summary of successful experiments of the last few years. Future developments, in particular in combination with synchronized table-top lasers, will also be discussed.

Along with the laser-driven electron gun and electron-pulse-compression schemes, two new terahertz sources are planned for generation of coherent and broadband THz radiation. While broadband THz radiation will be generated simply from an highly charged (up to 1nC) ultrashort (less than 200 fs) electron bunch in a bending magnet, a slightly more tunable THz source based on a few-period superradiant undulator is also under development. Since these sources will not require an optical cavity, their repetition rate and their peak powers will be adjustable by the laser-driven gun. With installation starting in 2011, these new THz sources will provide intense and phase-stable THz pulses providing new experimental possibilities at ELBE.

THz nonlinear interaction and quantum optics in the sub-cycle regime

R. Huber[1], A. Sell[1], A. A. Anappara[1], A. Leitenstorfer[1],
G. Biasiol[2], L. Sorba[2], A. Tredicucci[2],
T. Kampfrath[3], K. v. Volkmann[3], M. Wolf[3]

Department of Physics and Center for Applied Photonics, University of Konstanz, Germany.
NEST CNR-INFM and Scuola Normale Superiore, Pisa, Italy
Department of Physics, Freie Universität Berlin, Berlin, Germany

We present a novel laser source of intense phase-locked few-cycle THz pulses tunable between 1 and 107 THz [1,2]. Fiber-based few-fs gate pulses [3,4] allow us to trace all THz transients electro-optically. Peak electric and magnetic fields of up to 108 MV/cm and 30 T, respectively, pave the way to extremely nonlinear THz interaction. Examples include:

(i) THz Rabi cycles promoting 1s para excitons in the semiconductor Cu_2O into the 2p state [5]. The results point out a promising route towards ultracold exciton gases and potential Bose-Einstein condensation.

(ii) Intense THz transients coherently control collective magnon oscillations in antiferromagnetic NiO [6]. Being triggered by Zeeman interaction with the magnetic field of the THz pulse, this approach opens up a novel and most direct gateway to the ultrafast dynamics of electron spins in the electronic ground state.

(iii) Finally, we explore a new limit of non-adiabatic quantum electrodynamics: Intersubband cavity polaritons in a semiconductor quantum well waveguide structure are photogenerated by 12-fs near-infrared pulses. Multi-THz transients trace the abrupt conversion of photons into cavity polaritons. Our structure represents the first sub-cycle switching device of ultrastrong light-matter coupling and paves the way towards non-adiabatic quantum optics [7].

Peak electric fields of 1 V/Å reached by our novel high-field THz source even compete with inner atomic potential gradients. We may hence study condensed matter systems under unprecedented conditions of extremely high electric and magnetic fields.

[1] A. Sell et al., Opt. Lett. **33**, 2767 (2008).
[2] A. Sell et al. Appl. Phys. Lett. **93**, 251107 (2008).
[3] A. Sell et al., Opt. Express **17**, 1078 (2009).
[4] G. Krauss et al., Nature Photonics **4**, 33 (2010).
[5] S. Leinß et al., Phys. Rev. Lett. **101**, 246401 (2008).
[6] T. Kampfrath et al., submitted.
[7] G. Günter et al., Nature **458**, 178 (2009).

Dynamics of Superconductors Driven out of Equilibrium by Femtosecond Optical Pulses

M. Beck[(a)], M. Klammer[(a)], M. Beyer [(a)], H. Schäfer[(a)], S. Lang[(a)], D. Städter[(a)], V.V. Kabanov[(b,c)], G. Goltsman[(d)], G. Logvenov[(e)], I. Bozovic[(e)], G. Koren[(f)] and J. Demsar[(a,b,c,*)]

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The study of nonequilibrium phenomena in superconductors has been an important topic of condensed matter physics since the 1960's and the fact that intense laser pulses can non-thermally destroy the superconducting state has been known for a long time. The field of non-equilibrium superconductivity regained a lot of interest in 1990's, on one side due to the discovery of high temperature superconductivity, and on the other side due to the rapid development in the generation of femtosecond laser pulses.

In the last decade or so numerous real-time studies of carrier relaxation dynamics in superconductors have been performed utilizing pump-probe techniques. Most of the work focused on cuprate superconductors, with the large majority of studies performed in the all-optical configuration, where photoinduced changes in the dielectric constant at optical frequencies (far above the characteristic energy gap scale) were studied as a function of temperature, excitation density, or applied magnetic field. Several competing theoretical models have been proposed, yet an overall consensus on the underlying relaxation processes is still lacking. Moreover, no data on the complex conductivity dynamics in conventional superconductors, which could serve as a benchmark when compared to the unconventional high temperature superconductors, exist to date.

To address these questions we have performed first optical pump - THz probe studies of the complex conductivity dynamics in a conventional BCS superconductor NbN. The temperature and excitation intensity studies over large range of excitation densities were performed. These results will be reviewed in view of the existing theoretical models and compared to the results obtained on cuprate superconductor $La_{2-x}Sr_xCuO_4$.

THz induced phase transitions in manganites

Daniele Fausti

CFEL Hamburg

Sub-Terahertz spectroscopy in superconductors and charge-ordered materials

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Measuring the optical gaps of superconductors with low critical temperatures, or the collective excitations of ordered systems of charges like the Charge Density Waves, requires an exceptionally high sensitivity in the sub-Terahertz range, not achievable with the conventional broad-band sources.

Signal-to-noise ratios up to 1000 on reflectivity measurements between 3 and 30 cm^{-1} (at a spectral resolution better than 1 cm^{-1}) can be obtained by use of Coherent Synchrotron Radiation (CSR), like that produced by the storage ring BESSY in Berlin when it works in the so-called low-α mode [1].

These excellent experimental conditions were exploited either to measure the gap of novel superconductors, like boron-doped diamond [2] and CaAlSi [3], or to observe the collective excitations of charge-orders systems like the La-Ca and Nd-Sr manganites [4] or the multiferroic LuFe(2)O(4) [5].

All the above experiments will be reviewed, and the future perspectives of CSR, as applied to solid state physics, briefly discussed.

[1]. M. Abo-Bakr *et al.*, *Phys. Rev. Lett.* **90** 094801 (2003).
[2] M. Ortolani *et al.*, *Phys. Rev. Lett.* **97** 097002 (2006).
[3] M. Ortolani *et al*., *Phys. Rev. B* **73**, 184508 (2006).
[4] A. Nucara *et al., Phys. Rev. Lett.* **101** 066407 (2008).
[5] F. Vitucci *et al*., submitted *to Phys. Rev. B* (2010).

Klaas-Jan Tielrooij, Huib Bakker and Mischa Bonn

THz Studies of Dynamics of Water around Protons and Ions

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Despite many studies on water, many questions remain, for instance about the interaction of water with charged entities, such as proteins, protons and ions. We study water around protons and ions using TeraHertz and femtosecond mid-infrared spectroscopy.
For water around protons, we show that as many as ~20 water molecules are involved in proton transport [1]. These results are consistent with the notion that picosecond reorientational motion of water in the hydrogen bonded network close to the proton is the rate limiting step in proton transfer. For water around ions, we introduce the concept of semi-rigid hydration shells, and show that these semi-rigid hydration shells can become rigid when strongly hydrating anions and cations act cooperatively [2].

[1] K.J. Tielrooij, R.L. Timmer, H.J. Bakker & M. Bonn, Phys. Rev. Lett. 102, 198303 (2009).
[2] K.J. Tielrooij, N. Garcia-Araez, M. Bonn & H.J. Bakker, Science, in print (2010).

14-16 June 2010: Workshop at Forschungzentrum Dresden-Rossendorf (FZD), Dresden, Germany

Micro-spectroscopy and chemical nanoscopy using infrared and THz radiation

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Honorable Guest Professor of Shizuoka University
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Abstract

A new experimental station will be integrated into the new ANKA-IR2 beamline to combine broadband synchrotron radiation with near-field microscopy, among other microscopy techniques also available in the same instrument [1]. This will foster studies of the same sample at the same position with a variety of microscopy techniques currently including conventional confocal visible microscopy, atomic force based microscopy (AFM), confocal Raman microscopy, aperture based scanning optical near-field microscopy (SNOM), and apertureless scattering scanning nearfield infrared (s-SNIM, [2]) and near-field terahertz microscopy. Infrared spectroscopy combined with tip enhanced near-field techniques is used to reveal chemical fingerprints and to map the spatial distribution of chemicals with a resolution below the diffraction limit, reaching a few tens of nanometer or less leading in analogy to well-known chemical microscopes to chemical nanoscopes.

The presentation will show applications using tunable IR lasers as light sources for near-field microscopy. We have performed a variety of applications investigating biomaterials like self-assembled monolayers with zeptomolar concentration [3], DNA nanometric biosensor arrays [4], and 5 nm thick nanoscale lipid membranes [5,6] as well as solid state samples like single nanoparticles formed in dusty plasma [7] and implanted nanostructured dopants in seminconductors [8]. Tip enhanced near-field spectroscopy can obtain and map infrared spectra for a few 100 DNA molecules [4]. Coherent synchrotron radiation (CSR) also available at synchrotron sources has been measured at ANKA to determine power and beam profile [9] for the coupling of terahertz radiation to the nanoscope. Applications known in ensemble THz spectroscopy could then be transferred to nanoscale samples and domains. The status of the project supported by BMBF05KS7PC2 for innovative instrumentation of the synchrotron ANKA will be discussed [1].

References

1. E. Bründermann, D. A. Schmidt, I. Kopf, and M. Havenith, Nano-spectroscopy and chemical nanoscopy of biomaterials, AIP Conf. Proc. 1214, 7 (2010), DOI:10.1063/1.3326355
2. E. Bründermann, M. Havenith, SNIM-Scanning near-field infrared microscopy, Annu. Rep. Prog. Chem., Sect. C: Phys. Chem., 104, 235 (2008). Review and cover article.
3. I. Kopf, J.-S. Samson, G. Wollny, Ch. Grunwald, E. Bründermann, and M. Havenith, Chemical Imaging of Microstructured Self-Assembled Monolayers with Nanometer Resolution, J. Phys. Chem. C 111, 8166-8171 (2007).
4. I. Kopf, C. Grunwald, E. Bründermann, L. Casalis, G. Scoles, M. Havenith, Detection of hybridization on nanografted oligonucleotides using scanning near-field infrared microscopy, J. Phys. Chem. C 114 (2), 1306–1311 (2010).
5. G. Wollny, E. Bründermann, Z. Arsov, L. Quaroni, M. Havenith, Nanoscale depth resolution in scanning near-field infrared microscopy, Optics Express 16, 7453 (2008).
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7. J.-S. Samson, R. Meißner, E. Bründermann, M. Böke, J. Winter, M. Havenith, Characterization of single diamond-like and polymer-like nanoparticles by mid infrared nanospectroscopy, J. Appl. Phys. 105, 064908 (2009).
8. J.-S. Samson, G. Wollny, E. Bründermann, A. Bergner, A. Hecker, G. Schwaab, A.D. Wieck, and M. Havenith, Set-up of a scanning near field infrared microscope (SNIM): Imaging of sub-surface nano-structures in gallium-doped silicon, Phys. Chem. Chem. Phys. 8, 753 - 758 (2006).
9. A.-S. Müller, I. Birkel, S. Casalbuoni, B. Gasharova, E. Huttel, Y.-L. Mathis, D.A. Moss, N. Smale, P. Wesolowski, E. Bründermann, T. Bückle, M. Klein, Characterizing THz coherent synchrotron radiation at the ANKA storage ring, Proceedings of EPAC08, European Particle Accelerator Conference, WEPC046, 2091-2093 (2008).

THz pulses as probe and driving force of electron and spin excitations

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Many fundamental elementary excitations of physical systems exhibit transition energies of the order of 10meV. Electromagnetic pulses with frequencies in the terahertz (THz) range have a photon energy of 4.1meV per THz and are, thus, perfectly suited to probe or even drive such excitations. In this talk, we present two examples to illustrate the flavors of THz spectroscopy.

First, THz pulses are used as an "ultrafast Ohm-meter" of carbon-nanotube films to reveal the mechanism of the broad absorption resonance at 4THz. We observe an overall depletion of this peak when the tubes are excited by a femtosecond optical pulse. This finding excludes THz absorption through a collective electron oscillation (particle-plasmon polariton) along the tube axis. It instead shows that interband transitions in tubes with an energy gap of $\sim$ 10meV dominate the THz conductivity. Temperature-dependent measurements suggest that the chemical potential has a tube-to-tube variation of $\sim$ 0.1eV in our sample [1].

Second, THz pulses serve as an "ultrafast magnet" to launch a spin wave with a frequency as high as 1THz in the antiferromagnet NiO. The induced magnetization dynamics is monitored by the transient Faraday rotation experienced by an optical probe pulse. We show that the spin precession is driven by straightforward Zeeman coupling between the spins and the THz *magnetic* field. This mechanism affords a novel approach of ultrafast coherent spin control in the electronic ground state [2].

[1] T. Kampfrath, K. von Volkmann, C.M. Aguirre, P. Desjardins, R. Martel, M. Krenz, C. Frischkorn, M. Wolf, and L. Perfetti, Phys. Rev. Lett. **101**, 267403 (2008)
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Electric and Magnetic Resonance at Terahertz Frequencies

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Nonlinear and time-resolved spectroscopies based on manipulating electric charges and magnetic dipoles in condensed matter are relatively widespread at radio and microwave frequencies ($<10^{11}$Hz = 0.1 THz), as well as at infrared and optical frequencies (> 30 THz). The difficulty of generating high-power, coherent radiation between ~0.1 and 30 THz has limited such spectroscopies in this so-called "terahertz gap." The first part of this talk will introduce the 0.24 THz free-electron-laser-driven pulsed EPR spectrometer that is under development at UCSB. The remainder of the talk will focus near-ir spectroscopy of undoped quantum wells driven by intense terahertz fields. The lowest excitonic absorption splits and shifts under the influence of strong terahertz excitation. A full explanation of the data requires a theoretical treatment that goes beyond the rotating wave and two-level approximations that are familiar from standard treatments of quantum systems in oscillating fields.

Broad-band THz spectroscopy in magnetic fields: applications to the study of multiferroics

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EPR Studies on Materials Relevant for Solar Energy Conversion: The EPR-Solar Approach

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Materials applicable for photovoltaics and catalytic solar energy conversion frequently exhibit function determining paramagnetic states. This holds true for such dissimilar materials like photovoltaic semiconductors, where the genesis and structure of defect states determines charge transport and loss mechanisms, or photocatalytic transition metal ion complexes, where the catalytic cycle often involves paramagnetic states. Knowledge about the spin coupling parameters of theses states thereby provides highly desired pieces of information, which help to understand and control the structure function relationship in photovoltaic and photocatalytic materials. The method of choice to extract the relevant parameters is electron paramagnetic resonance (EPR). However, recent EPR studies, typically carried out at X-band frequencies, suffered from instrumental restrictions with respect to detection sensitivity, spectral resolution and the availability of broad band THz sources. This situation dramatically changed with the advent of advanced multi-frequency EPR methods and the implementation of novel THz-EPR techniques based on coherent synchrotron radiation in the THz range. With the aim to further exploit the increased capabilities of advanced EPR to study materials relevant for future photovoltaic and photocatalytic devices; in 2008 the interdisciplinary German network *EPR-Solar* was formed. Here we present recent multifrequency EPR and electrically detected magnetic resonance (EDMR) studies on defect states in amorphous and microcrystalline solar cells as well as THz-EPR studies on high spin Mn complexes, which have been carried out by the network partners.

Acknowledgment: This work is funded by the German Federal Ministry of Education and Research (network project EPR-Solar 03SF0328).

Ultrafast Resistive Switching in Magnetite employing Coherent Synchrotron Radiation at THz frequencies

K. Holldack

The photo-induced transient charge carrier dynamics in magnetite (Fe_3O_4) was probed by monitoring the THz absorption of a 400 nm-thick single crystalline layer [1]. This was carried out using an optical pump-THz probe setup in transmission mode employing ultrashort THz pulses emitted from laser-energy modulation of relativistic electrons in the BESSY II storage ring [2].
The experiments were carried out at the dedicated THz beamline which was installed in the slicing section of BESSY II in 2004. A general overview of the setup and a variety of scientific THz applications there will be briefly reported.

In Magnetite, the transient change of the spectrally integrated low frequency (>2.4 THz i.e. 80 cm^{-1}) THz transmission reveals that the carrier generation is accomplished within 1 ps for sample temperatures spanning from 80 K to 300 K. This indicates that this change reflects the temperature-dependent semiconducting behaviour of magnetite which almost behaves as an insulator at the lowest temperatures but tends to a half-metal at room temperature. No cooperative laser-induced phase transition (Verwey transition in Fe_3O_4 at 120 K) has been observed contrary to what has often been reported in other transition metal oxides. Considering the fluence and temperature dependence of the observed transient change, the ultrafast transmittance variation is found to be mainly driven by the photo-injected charge carrier density and the Verwey transition seems not “switchable” by the laser on ps timescales but occurs statically crossing the Verwey temperature.

The results have clearly demonstrated the feasibility to perform condensed matter experiments of high quality employing THz pulses being naturally synchronized to an optical laser pulse emitted from an accelerator-based source.

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Coherent THz pulses from linear accelerators: challenges and opportunities

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The potential and perspectives of experiments with coherent THz pulses from ultra short electron bunches are presented based on the commissioning results from a unique instrumentation situated at the soft x-ray free electron laser FLASH. This particular set up provides radiation throughout the THz and in the XUV to soft x-ray spectral range. The THz pulses and the XUV/soft x-ray pulses are generated from the same electron bunch and thereby naturally synchronized. This natural synchronization between the THz and soft-X-ray pulses is an intrinsic property of the *cascaded-undulator* design, where a long-period THz undulator immediately follows the short-period x-ray undulators. The electric field within the THz pulse furthermore is a Lorentz transformed image of the periodicity of the THz undulator and fixed with respect to its envelope, thereby generating naturally phase stable radiation. Due to the only few micron long electron bunches the THz pulse energy scales quadratically with the bunch charge and at FLASH routinely reaches the μJ range [1] providing for THz electric fields in the experiment of up to 100 MV/m. The instrumentation has been benchmarked and analyzed by novel NIR/THz and XUV/THz cross correlation techniques [2] in the time and frequency domain. It is shown, that combination of the THz pulses with the fully synchronized femtosecond soft x-ray pulses enables a new class of experiments that directly probes the sub cycle dynamics of e.g. highly vibrationally perturbed molecules and solids. Based on these results the general potential of science with THz radiation from ultra short electron bunches will be discussed in hindsight that these electron bunch forms are typical for all currently developed or already operating so called 4_{th} generation X-ray light sources. A perspective on the most promising fields of research is given and possibilities for transfer of the novel conceptional design of the FLASH THz beamline to other 4_{th} generation x-ray light sources or linear accelerators working with ultra short electron bunches are outlined.

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Broadband far- and mid infrared pump-probe spectroscopy using synchrotron radiation

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In spite of the increasing availability of pulsed infrared and THz laser light sources, trans_mission and reflection spectroscopy with "non-coherent" sources remain attractive for time resolved, or steady-state, studies in solid state and surface chemistry. A good example is that of Kramers-Kronig transformation determinations of the frequency dependent complex surface conductivity, deduced from spectra taken over large wavelength ranges.
For such broadband applications, FTIR spectroscopy is found to be the most useful method. Practical sources consist of thermal lamps (globar etc.) and with increasing importance, IR synchrotron based sources, in the case of investigations of small sized (diffraction limit) samples.
Here, the broadband pump/probe set-up as realized recently at the IR-synchrotron beamline of the SLS, is presented. Time dependent phenomena triggered by an optical or electrical pulse are investigated. The available spectral window covers about a decade, typically from $< 10^3$ cm^{-1} to 10^4 cm^{-1}.
We give the example of electron-hole carriers plasma reflection, optically induced at an germanium/air surface, and we discuss the prospect to generate a strong carrier population inversion at the direct band gap of Ge, for interband lasing application. Finally, we consider extension of this method to the THz range, using coherent light from fs-slicing of the storage-ring electron bunches, and operation of the SLS in the low-alpha mode.

THz Quantum Cascade Lasers: Confinement and Dynamics

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Nowadays terahertz (THz) quantum-cascade lasers (QCLs) cover a broad frequency region and are capable to deliver peak powers up to several 100 mW as a consequence of refined band structure designs. In addition, THz quantum cascade lasers show exciting novel properties when the carrier or photon confinement is changed.

Carrier confinement by a magnetic field leads to large performance increases and to new striking physical effects. The magnetic field induced reduction of the dimensionality changes the carrier dynamics in favorable way i.e. by reducing non-radiative relaxation channels with important consequences for novel lower dimensional quantum devices.

Photon confinement by photonic crystal resonators or novel micro cavities allows sub-wavelength confinement resulting in single mode emission and ultralow threshold current.

The combination with time-resolved THz spectroscopy allows to study stimulated emission and amplification with a phase-resolved methods.

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Magnetic and magnetoelectric excitations in multiferroic manganites

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Multiferroics are materials simultaneously showing ferromagnetic and ferroelectric order. Two order parameters are coupled in these materials, which leads to such unusual effects like magnetic switching of electric polarization and dielectric constant. As can be expected already from the first principles, changes in the static properties of multiferroics must be accompanied by dynamic effects like characteristic magnetoelectric excitations. Indeed, such excitations could be recently observed in the spectra and were called electromagnons. Contrary to the conventional magnons, the electromagnons are excited by the electric component of the electromagnetic wave and contribute to the static dielectric permittivity. The suppression of electromagnons in external magnetic fields provides a natural explanation for the magnetoelectric effects in broad frequency range between dc and terahertz.

Developments of THz ESR Systems Using a Micro-Cantilever

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Developments of our highly sensitive THz ESR systems will be presented. THz ESR has several advantages, such as the high spectral resolution, the observation of the zero field gap or the measurement beyond the magnetic phase transition. However, the typical sensitivity of conventional THz ESR using the transmission method is about 10^{15} spins/G and it is not applicable to the detection of micrometer size sample. In order to overcome this difficulty, we applied the torque measurement using a micro-cantilever for ESR detection [1-6]. We have achieved the sensitivity of 10^{11} spins/G using Gunn oscillators up to 315 GHz. The applications of technique to both static and pulsed magnetic fields will be discussed.

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Probing collective spin states in cubic cobaltates by high-frequency ESR spectroscopy

Vladik Kataev

IFW Dresden

Imaging the THz electric near-field of sub-wavelength metal structures

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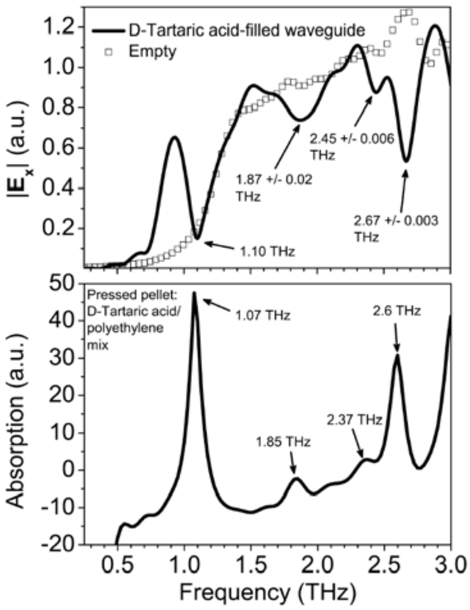
We present results of measurements of the terahertz electric near field of small, sub-wavelength sized metal structures, such as holes, antennas and split-ring resonators. We also show how the measured near fields change when, for example, the holes are illed with a dielectric material. The latter allows us to measure the absorption spectrum of a tiny volume of a sample, such as D-tartaric acid.

In the figure we show the transmitted, normalized near-field spectrum of an empty hole with a diameter of about 150 μm, and an identical hole filled with crystalline D-tartaric acid powder. In addition to a shift of the aperture cut-off frequency towards lower frequencies, the D-tartaric acid-filled aperture shows dips at frequencies where the D-tartaric acid is known to have absorption lines (bottom graph, quasi near-field measurement of pressed pellet). For THz frequencies, this volume of sample is actually quite small and a far-field measurement on such a small volume sample would be extremely difficult, if not impossible. The advantage being that all the light that emerges from the hole or waveguide must have interacted with the sample, making this a background-free measurement.



Top: THz near-field spectra measured behind an empty aperture, and an aperture filled with D-tartaric acid.
Bottom: Quasi near-field THz spectrum of a pressed polyethylene/D-tartaric pellet.

Infrared and THz magnetospectroscopy of graphene

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Features of the high-frequency ESR in one-dimensional quantum spin systems

A.A. Zvyagin

The theory of the electron spin resonance is developed for quantum spin chains with the alternation of exchange constants.
It is shown that only magnetically anisotropic relativistic interactions or Dzyaloshinskii-Moriya exchange-relativistic couplings can produce shifts of resonance positions (comparing to the ESR position of a single spin), and cause the broadening of the ESR linewidth due to spin-spin interactions. The alternation of the spin-spin coupling along chains together with the mentioned magnetic anisotropy cause two resonance branches, in general case with gaps. The values of those gaps are determined by the values of exchange constants (and their magnetic anisotropy) between nearest neighbor spins, and, if present, by the values of the exchange and the anisotropy of next-nearest-neighbor interactions between spins. For usual spin chains ESR frequencies related to those gaps can be in the THz region. The intensities of the absorption of two resonance lines are different; they depend on the interaction constants. Next-nearest-neighbor interactions can produce asymmetry of the ESR lines with respect to the gapless line of a single spin. We discuss possible temperature dependencies of the ESR shifts, and analyze influence of mentioned spin-spin interactions on the ESR linewidths.

High Field EPR of Single Molecule Magnets

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Molecules showing slow relaxation of the magnetization at low temperature, commonly known as single molecule magnets (SMMs), have represented a major breakthrough in nanomagnetism providing evidence of the coexistence of classical and quantum effects in the dynamics of the magnetization. HF-EPR spectroscopy has demonstrated to be a key tool to characterize these quantum systems and provide information on their magnetic anisotropy. While longitudinal anisotropy is responsible of the height of the barrier, transverse magnetic anisotropy, in particular high order terms, influence the quantum tunnelling of the magnetization. In general the low-temperature magnetic properties of SMMs have been interpreted by attributing a well-defined total spin value (*S*) to the ground state and by using a so-called “giant spin Hamiltonian” (GSH). However, to justify the single crystal HF-EPR spectra recorded on a truly tetragonal Mn_{12} cluster compound a simplified multispin Hamiltonian, which takes into account the exchange interactions and the single ion magnetic anisotropy of the Mn^{III} centers, has been used. Transverse anisotropy in axial single molecule magnets has been found to originate from the multispin nature of the system and from the breakdown of the strong exchange approximation. The tilting of the single-ion easy axes of magnetization with respect to the 4-fold molecular axis of the cluster plays the major role in determining the transverse anisotropy. These investigations have thus provided precious magneto-structural correlations for the design of novel SMMs.

Far infrared spectroscopy at the HFML

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High Frequency EPR studies of iron spin clusters

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We will present recent results obtained from the use of High-Frequency Electron Paramagnetic Resonance (HF-EPR) spectroscopy for the investigation of iron magnetic nano-systems. The studies reported will start from small spin clusters, involving only four Fe(III) ions, to much larger entities comprising thirty magnetic ions. Besides the analysis of the magnetic anisotropy of these complexes obtained from HF-EPR, the aim is also to point out that the same approach is used from small complexes up to large complexes which are reaching the sizes of magnetic nanoparticles. Whereas the size of the systems is becoming the same (a few nanometers), the approaches to the interpretation of their properties are still different, being bottom up for the spin clusters and top down for the magnetic nanoparticles. We want also to illustrate the need for a common view point.

ESR in the THz range, recent results and experimental developments

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High-Frequency and High-Field ESR in Quantum Spin Systems.

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Quantum fluctuations, significantly enhanced in spin systems with reduced dimensionality, give rise to a variety of strongly correlated states, making low-dimensional magnets an ideal ground for testing various theoretical concepts. In my presentation, I will focus on recent results of high-frequency and high-field electron spin resonance (ESR) studies of quantum spin systems, allowing us to study the low-energy spin dynamics in spin-1/2 and spin-1 chain systems, spin-ladders, and quasi-2D magnets. I am also going to briefly discuss the recent development of the high-field ESR Program at the Dresden High Magnetic Field Laboratory (HLD) at FZD.

Poster Abstracts

Cyclotron resonance in InAs/AlSb QW heterostructures in ultraquantum limit

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Cyclotron resonance (CR) in InAs/AlSb QW heterostructures in ultraquantum limit in the magnetic field up to 50 T has been studied. Free electron laser has been used as a radiation source. We observed a single line in CR spectra which correspond to transition from the lowest Landau level. According to the single-electron theoretical model energy of this transition shouldn't depend on electron concentration, however, experiments have yielded a different result: cyclotron line shifts to the lower magnetic fields with concentration decreasing. We speculate that this effect results from electron-electron interaction and from the broadening of Landau levels. The above effects will be discussed in the talk.

Terahertz time-domain cyclotron resonance spectroscopy in pulsed magnetic field

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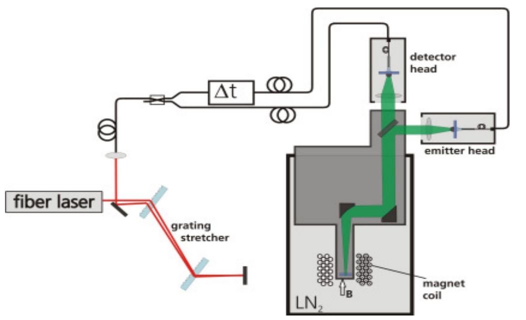
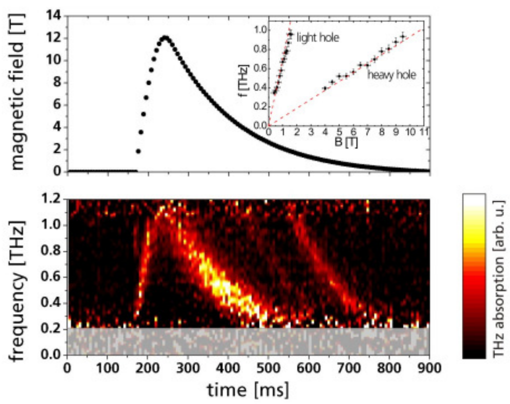
For the first time, energy dispersive cyclotron resonance (CR) spectra were measured during a single magnetic field pulse by using a rapid-scanning, fiber-coupled terahertz (THz) time-domain spectroscopy (TDS) system [1, 2]. The experimental setup, actually compact and portable, is depicted in the figure below. Broadband THz ultrashort pulses were generated and detected by using a customized photoconductive emitter-detector setup pumped by a femto-second fiber-laser. The emitter and the detector, based on low-temperature GaAs, were kept far enough from the magnet coil. Then, free-space propagation of the THz signal was used inside the cryostat that includes a high resistivity silicon beam splitter and an off-axis parabolic mirror for focussing the beam on a p-type germanium sample that sits on a mirror. In the upper plot of the right figure, magnetic fields at which spectra were measured are shown by dots as a function of time. The lower plot shows THz transmission spectra measured at 77K during a single shot and obtained at the frequency of 150 Hz, actually the scanning rate of THz pulses by the delay line (Δt) in the figure below. Absorption is decoded in colour (the low





frequency part is greyed out because of poor S/N). Strong spectral light and heavy holes CR absorption peaks are displayed on the down sweep of the field at frequencies that follow linearly the magnetic field ($2\pi\ \nu_c = eB/m^*$). The peaks provide the light and heavy holes cyclotron effective masses m* of $0.04m_0$ and $0.3\ m_0$ expected for a crystal with magnetic field parallel to the 100 axis. In addition, a weaker line related to quantum effects is observed. Further developments now in progress are aimed at increasing the scanning speed, the intensity and the frequency range of THz pulses. This work paves the way to routine terahertz time-domain-spectroscopy at low temperatures and magnetic fields supplied by 60 Tesla coils.

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THz response of a two-dimensional electron gas

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We investigate the THz transmission through a two dimensional electron gas (2DEG) in a high electron-mobility transistor-like device [1]. A clear resonance is found in the imaginary part of the inverse dielectric function similar to previous measurements in a three-dimensional system [2]. We vary the charge density in the 2DEG by applying different voltages. The square of plasma peak frequency increases linearly with density. Other contributions besides the 2DEG are carefully excluded by investigating various reference structures showing no THz response. The results are analyzed using a microscopic analysis.

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Coherent broadband continuous-wave THz spectrometry: A powerful tool for low-energy solid-state spectroscopy

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We present the development of a continuous-wave THz spectrometer and its application to solid-state spectroscopy. This spectrometer is based on the principle of THz generation using frequency mixing of two near-infrared distributed feedback diode lasers with frequency stabilization. The laser beat is converted into THz radiation by a photomixer, which efficiently generates THz radiation from 60 GHz to 1.8 THz. The THz radiation is detected by a second photomixer via homodyne mixing of the THz signal and the laser beat. A phase modulation technique is used to accurately determine the amplitude and the phase at a given frequency. Also, a photocurrent correction is implemented to account for the drifts in the THz intensity using the dc photocurrents measured at the photomixers. The complex optical functions can then be evaluated from the full phase information of the THz beam, and a very high spectral resolution in the MHz range can be achieved. Furthermore, this compact spectrometer can be integrated within a magnetic cryostat eliminating the need for optical windows. In this way, investigations at high magnetic fields up to 16 T and low temperatures down to 2 K can be performed without loss of intensity. Thus, a new door is opened for exploring low-energy electronic excitations of novel materials, lying in the sub-phonon energy regime.

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Ultrafast saturable absorption in doped semiconductors at high THz field strengths

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Abstract: We demonstrate saturable absorber behavior of n-type semiconductors GaAs, GaP, and Ge in the THz frequency range using nonlinear THz spectroscopy, with the THz pulses reaching 300 kV/cm in electric field strength. Apart from classical saturable transmission, we observe THz pulse shortening and increase of the group refractive index at high field strengths.

1. Introduction

Semiconductor saturable absorbers and saturable absorber mirrors (SESAMs) are routinely used for ultrafast laser modelocking and ultrafast signal control [1]. Saturable absorbers operating in the visible and infrared wavelength ranges rely on bleaching of two-level electronic systems, usually realized by an interband transition in semiconductor quantum wells or quantum dots [2, 3]. Clearly, such quantum confined semiconductor systems are not suitable for applications in the far-infrared (THz) range, where the photon energy is much smaller than the bandgap energy of most semiconductors, and where thermal population of such closely spaced electronic levels dominates. In doped bulk semiconductors the dominant loss factor for THz frequency range is free-carrier absorption which can be saturated by hot carrier effects leading to reduced carrier mobility. Detailed studies of time-resolved high-THz-field transport in bulk semiconductors were recently published in Refs. [4,5]. Here, we demonstrate saturable absorbers for the THz frequency range, based on n-type bulk semiconductors, where the carrier mobility is modulated by nonlinear electron transport caused by the THz electric field, thus affecting the conductivity of the sample.

2. Experimental Set-Up

In our experiment, we generated high-power single-cycle THz pulses by tilted pulse-front optical rectification in a lithium niobate crystal of 800-nm, 80-fs laser pulses provided by a 1-kHz repetition rate Ti:Sapphire amplifier [6]. The strongest THz signal used in our measurements, with the peak electric field reaching 300 kV/cm and instantaneous intensity reaching 120 MW/cm^2, is shown in Fig.1 THz pulses were collimated and then refocused onto a sample point using a set of off-axis paraboloidal mirrors. A pair of broadband wiregrid polarizers was used to controllably attenuate the THz pulses. After propagation through the sample point, the THz pulses were detected by standard free-space electro-optic sampling. Direct and indirect n-type semiconductors GaAs, GaP, and Ge with different doping levels were studied at room temperature.

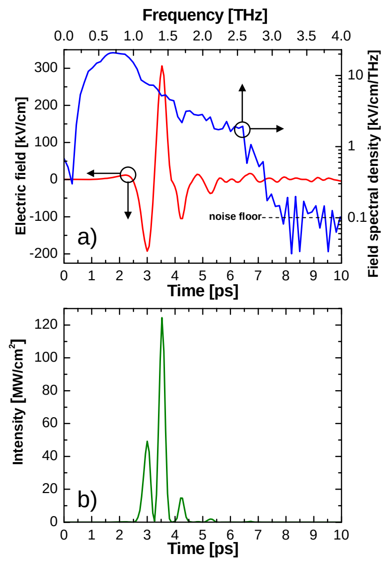

Fig 1. (a) Electric field and (b) instantaneous intensity of the strongest THz pulse used in the experiment.

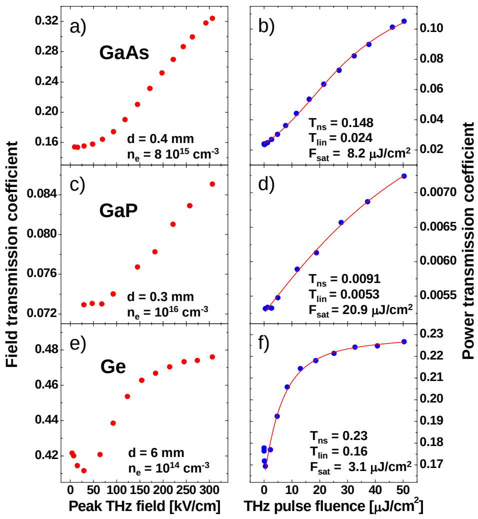

Fig. 2. Field and power transmission coefficients for GaAs (a,b), GaP (c,d) and Ge (d,e). Solid lines: fit with saturable transmission function

3. Results and Discussion

Fig. 2 shows the field and power transmission coefficients as a function of THz pump field and fluence respectively in GaAs (d = 0.4 mm, $n_e = 8\times10^{15}$ cm^{-3}), GaP (d = 0.3 mm, $n_e = 10^{16}$cm^{-3}) and Ge (d = 6 mm, $n_e = 10^{14}$ cm^{-3}). The field and power transmission coefficients were obtained by integrating either the modulus or the square, respectively, of the THz fields transmitted through the sample, and dividing them by reference values recorded without the sample in the beam path. In all our samples we observed increased transmission at higher pump fluences. In particular, we observed a nearly five-fold increase in power transmission coefficient for GaAs sample in the full THz pulse fluence range of our experiments. The solid lines in Figs. 2 (b,d,f) are fits to measured data using a saturable power transmission function, defined after Ref. [1] as

$$T(F_p) = T_{ns}\frac{\ln\left[1+T_{lin}/T_{ns}\left(e^{F_p/F_{sat}}-1\right)\right]}{F_p/F_{sat}} \qquad (1)$$

where T_{lin} and T_{ns} are linear and non-saturable power transmission coefficients, F_p is the pump fluence, and F_{sat} is the saturation fluence. Using fits with Eq. 1 we were able to extract saturable absorber parameters for our semiconductor samples, as indicated in Fig 1 and Ref [7]. In particular, the saturation fluence F_{sat} was found to be 8.2 µJ/cm^2 for GaAs, 20.9 µJ/cm^2 for GaP, and 3.1 µJ/cm^2 for Ge.

Saturable absorption is accompanied by pulse shortening and by an increase in group refractive index in all three samples, as calculated from the Hilbert transforms (HT) of THz pulses, as shown in Fig. 3. The pulse shortening factor was calculated as the ratio of FWHMs of HT's modula of a sample and reference THz pulses. The group index n_g was calculated using the difference of arrival times $\Delta\tau$ between sample and reference pulses and taking into account the sample thickness. The arrival times were obtained from mean-weighted maxima of the modula of THz pulses' HTs.

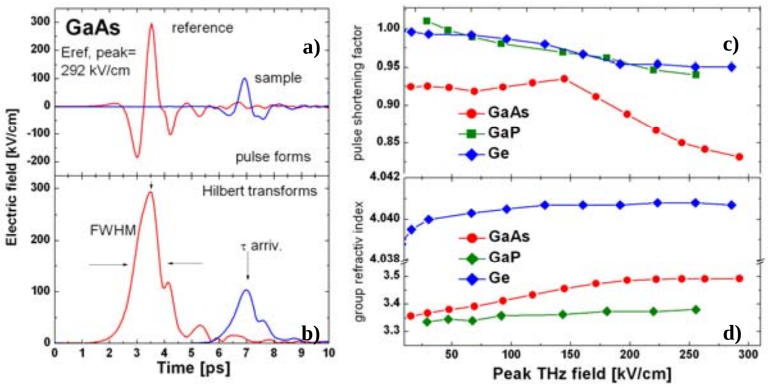

Fig. 3. a) Shape of the THz pulses before (reference) and after (sample) propagating through the 0.4-mm thick GaAs sample. This reference pulse had peak electric field strength of 292 kV/cm. (b) Modulus of Hilbert transforms of these THz pulses. (c) Pulse shortening factors and (d) group refractive indices the samples as functions of peak electric field of incident THz pulse.

The pulse shortening is a typical effect of pulse propagation through the saturable-absorption medium. The increase in group index here can be explained by the phase shift at the saturable-absorbing sample interface [8], and by the propagation through the bulk of medium with steepening phase index dependency $n_\varphi(\omega)$ below plasma resonance. Indeed, stronger THz pump results in larger carrier effective mass, or in equivalent decrease in density of highly mobile carriers in the Γ-valley. This ~~decreases the plasma frequency~~ ω_p, making the phase index dependency $n_\varphi(\omega < \omega_p)$ steeper, and leading to a higher value of a group index for the stronger excited samples. Even though the origin of absorption mechanism in the THz and in the optical frequency ranges is completely different, we note that the saturation fluences observed here are within the same order of magnitude (i.e. few to tens of µJ/cm^2) as the values reported for SESAMs in the optical range, such as e.g. quantum dot SESAMs [2,3].

Possibilities of nonlinear THz experiments on doped semiconductors, combining quasi-monochromatic THz-excitation by free-electron laser sources with a broad-band ultrafast THz probe as well as possible applications of saturable absorbers for the THz range will be discussed.

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Investigation of MIR-pumped quantum-cascade structures as emitters of THz radiation

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One proposal to obtain THz radiation from a compact semiconductor source at room temperatures is based on an optically pumped, electrically driven (OPED) quantum-cascade structure [1]. It has been theoretically shown that such a structure may allow for overcoming the fundamental Manley-Rowe limit of frequency conversion by recycling the pump photons. Furthermore, quantum coherence effects and gain without inversion have been predicted. However, the realization of such a structure has proven to be difficult, which is due to nonlinear transport effects such as the formation of electric-field domains. We investigate the influence of MIR pumping on the electrical transport properties in different OPED quantum-cascade structure. Experiments are performed using free-electron laser (FEL) radiation in the range of 10 to 12 µm. Although the effect of the FEL pumping on the transport properties is rather small and ambiguous, in particular with respect to different polarization directions, a small emission signal in the THz range is observed. Additional experiments with a pulsed CO_2 laser show a clear influence of the pumping being on or off and of its polarization direction on the dc current-voltage characteristics.

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1.55 µm photoconductive THz emitters based on ErAs:$In_{0.53}Ga_{0.47}As$ superlattices

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ABSTRACT

$In_{0.53}Ga_{0.47}As$ with periodically arranged layers of self-assembled ErAs nanoislands is studied as a photoconductive material for THz emitters at an excitation wavelength of 1.55 µm. The ErAs:InGaAs superlattices were grown at 490 °C with molecular beam epitaxy on top of a semi-insulating (100) InP:Fe substrate and a 250 nm thick $In_{0.52}Al_{0.48}As$ buffer. Each superlattice consists of a periodic sequence of one layer of self-assembled ErAs islands and an $In_{0.53}Ga_{0.47}As$ spacer of thickness *L*. The ErAs islands act as trap sites for photoexcited charge carriers. They also donate free electrons to the $In_{0.53}Ga_{0.47}As$ which pushes the Fermi level closer to the conduction band. Be dopants are incorporated in the vicinity of the ErAs islands to compensate these free electrons and enhance the dark resistance. A bow-tie antenna structure is patterned with photo-lithography on top of these samples (20 nm Ti adhesion layer, 100 nm Au metallization). A 20 µm × 20 µm active mesa was etched where the two electrodes of the antenna structure approach each other in order to reduce the dark current further. By varying the distance *L* separating two adjacent ErAs island layers, the lifetime of the photoexcited charge carriers can be tuned. The carrier lifetime ranges from 0.2 to 6.3 ps when changing the period from 5 nm to 100 nm. The larger period superlattices generally exhibit higher photocurrents. The THz emission properties of the samples are measured with a THz time domain spectrometer driven with 1.55 µm laser light from a femtosecond fiber laser (TC-1550, MenloSystems GmbH). The THz waveforms were recorded with free-space electro-optic sampling using a <110>-oriented GaAs crystals. The maximum bandwidth exceeds 3 THz. There is no close correlation between the measured carrier lifetime and the observable bandwidth. We conclude that the lifetime does not have a dominant impact on the bandwidth. Instead, the bandwidth of the THz output is mainly determined by the dark resistivity of the material and the maximum bias voltage which can be applied.

Optical conductivity of $LuNi_2B_2C$ in the terahertz range

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Abstract

Using a backward-wave-oscillator-based setup in a Mach-Zehnder interferometer arrangement, we have measured the temperature- and frequency-dependent transmission and phase-shift spectra of $LuNi_2B_2C$ films on MgO substrates in the range 200 GHz - 1.4 THz. From the measured spectra, we have directly calculated the complex optical conductivity. We observe a clear signature of the superconducting energy gap in the spectra. In the poster, a comparison of the experimentally obtained spectra with theoretical predictions for a multi-band superconductor will be given.

THz-range free-electron laser ESR spectroscopy: techniques and applications in high magnetic fields

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The successful use of picosecond-pulse free-electron-laser (FEL) radiation for the continuous-wave THz-range electron spin resonance (ESR) spectroscopy has been demonstrated. The unique combination of two linac-based FELs (covering the wavelength range of 4 - 250 μm) with high magnetic fields at the Research Center Dresden-Rossendorf (FZD) allows for tunable-frequency ESR spectroscopy in an extraordinary broad frequency range of 1.2 - 75 THz in magnetic fields up to $\sim$70 T. The new approach is of particular importance for studying magnetic excitations in materials exhibiting field-induced phenomena (including magnetic phase transitions) and in spin systems with a large zero-field splitting. The performance of the spectrometer is illustrated with ESR spectra obtained in the low-dimensional organic material $(C_6H_9N_2)CuCl_3$ and the multiferroic compound $YMnO_3$. This work was made in collaboration with H.D. Zhou, C. Wiebe.

Fano interference in the intersubband THz response of semiconductor quantum wells after interband photoexcitation

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THz spectroscopy on semiconductor heterostructures has revealed new insight in low-energy excitations such as intersubband transitions (ISTs). A microscopic analysis has to consider the true THz transitions, but also the so-called ponderomotive response as a charge current of carriers generated by the alternating classical electric field. Its response can be simply described by $\chi = -\omega_p^2/\omega^2$ with the plasma frequency ω_p. Due to its ω^{-2} dependency it becomes relevant in the THz range where ISTs occur. In former experiments the ponderomotive contribution could only be seen rather indirectly [1].

Here, we directly study the interplay between ponderomotive contribution and true IST [2]. In our experiment we create electrons in the conduction band of an undoped $GaAs/Al_{0.34}Ga_{0.66}As$ multiple quantum well by resonant interband excitation at the 1s heavy-hole exciton, using either a broad- or narrowband laser. Broadband THz pulses probe the IST and are detected by electro-optic sampling. The observed differential THz transmission transient as the pump-induced change in the transmitted THz field shows a strong beating (inset of Fig. 1 a)). In the frequency domain this results in an intersubband resonance with a broad additional low-frequency peak (Fig. 1 a)). The line shape of this intersubband resonance strongly resembles a Fano-resonance with an undershoot at the low-frequency side and an asymmetric peak to higher frequencies. However, since we are able to measure differential transmission and total THz absorption separately, we find that the absorption does not show such a Fano-asymmetry, but reveals the expected Lorentzian-like line shape of the intersubband resonance (Fig. 1 b)). Especially, it is a single peak and therefore the beating in the time domain cannot originate from an adjacent second absorptive resonance next to the true intersubband resonance.

In our microscopic theory these features can be explained unambiguously by a phase-sensitive superposition of the true THz intersubband current and the ponderomotive current. This results in a Fano-like line shape of the differential transmission (Fig. 1 a)). However, since the ponderomotive current alone leads to a real-valued linear susceptibility, it does not contribute to the total THz absorption where only the imaginary part of the susceptibility enters. Therefore only the symmetric intersubband absorption line appears in the total THz absorption (Fig. 1 b)). Here the Fano-resonance does not originate from a coupling of the sharp intersubband resonance to an absorptive continuum, but to a continuum provided by the light-matter interaction in form of the ponderomotive current.

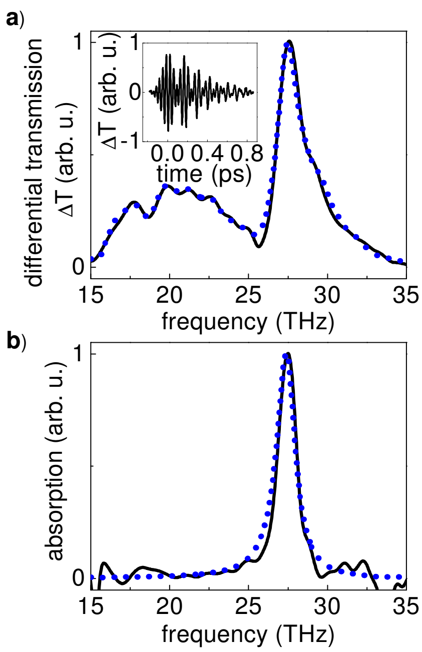

Fig. 1. a) Measured (black line) and computed (blue dotted line) differential transmission spectrum after broadband photoexcitation at the heavy hole 1s exciton. The inset shows the corresponding time transient. b) Measured (black line) and computed (blue dotted line) total THz absorption.

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THz sideband generation using intraexcitonic transitions in GaAs/AlGaAs quantum wells

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We have investigated the third-order nonlinear mixing process between a near-infrared laser and a free-electron laser in GaAs/AlGaAs multi quantum wells.

AC THz electric fields which couple strongly with intraband excitations in semiconductors can lead to spectral sidebands of a simultaneous interband excitation. In this nonlinear mixing process a near-infrared (NIR) laser beam is mixed with the THz beam to generate sidebands around the NIR frequency with a frequency spacing equal to the THz frequency or multiples of it: $\omega = \omega_{NIR} \pm n \times \omega_{THz}$ (with integer n). In recent years this effect has been investigated in various semiconductor systems (i.e. in bulk GaAs [1] and in multi quantum wells [2]). Here, we demonstrate efficient n=2 sideband generation in an undoped symmetric GaAs/AlGaAs multi quantum well film (substrate etched away) using an all-normal-incidence geometry for the picosecond NIR and THz laser pulses [3], the latter ones delivered by the free-electron laser (FEL) FELBE at the Forschungszentrum Dresden-Rossendorf. We use the tunability of the FEL to study the dependence of the mixing efficiency on THz wavelength. We find resonances related to heavy- to light-hole transitions, but also to the heavy-hole intraexciton 1s-2p transition. It turns out that the conversion efficiency of the n=+2 sideband is largest (up to 0.1 %) for the 1s-2p intraexciton transition, which is comparable to values previously reported for n=+1 [2]. The corresponding NIR transmission spectrum at 10 K (Fig. 1) shows the NIR fundamental at the hh(1s) state and the surrounding even-order sidebands.

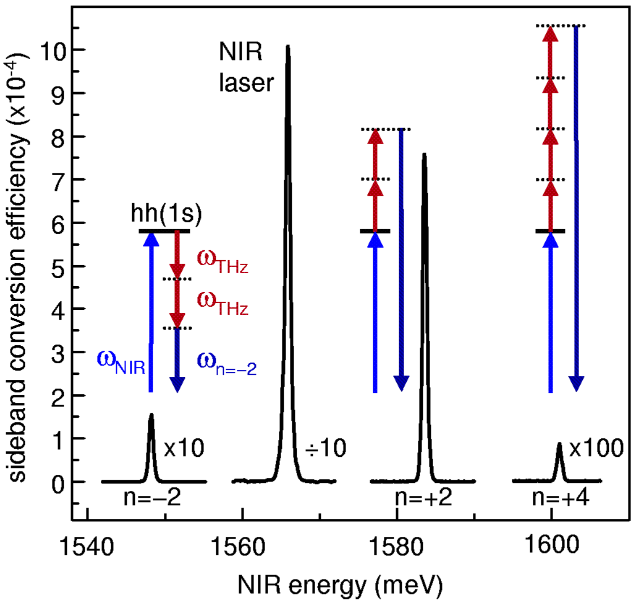

Fig. 1: Typical sideband spectrum. Around the NIR-laser line, centered at the hh1(1s) transition, different sidebands appear for an FEL energy close to the hh1(1s) → hh1(2p) transition at 9 meV. Energy level diagrams illustrate the involved transitions schematically with respect to the hh(1s) transition.

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The spatial structure of longitudinal and transversal fields in focussed terahertz beams

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Longitudinal and transverse terahertz fields in the focus of freely propagating terahertz beams of radial and linear polaraziation are investigated. The THz beams are generated by scalable photoconductive emitters of appropriate electrode geometry. The radiation is focused and detected with ZnTe crystals. Crystals oriented in the (110) direction serve as sensors for the transverse THz field components, (100) oriented crystals are applied for sensing of longitudinal field components. We measured the beam profiles and show that radially polarized beams can be described well as Bessel Gauss beams, which are solutions of the vector Helmholtz equation. Consistent with the theory we observe a smaller spot size for the longitudinal component of a radially polarized beam as compared to a similarly focused linearly polarized beam. A phase difference of $\pi/2$ is found for the longitudinal field components with respect to the transverse field components. It is a fundamental consequence of Maxwell's equation div $\vec{E}$ = 0 and observed in these experiements for the first time.

Carrier dynamics in graphene excited at low energy

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The relaxation dynamics of carriers is of key importance for understanding fundamental material properties of graphene and for device applications. It has been investigated by different single color and two color pump-probe studies [1-5]. Common to all these experiments is a pump wavelength of around 800 nm (1.55 eV). Relaxations times between 0.1 and 10 ps were observed and attributed to carrier-carrier intraband scattering and a cooling of the carrier distribution by phonons. We report on degenerate pump-probe experiments in the energy range from 30 meV – 250 meV. The radiation is provided by the free-electron laser FELBE. The graphene samples were grown on the carbon-terminated surface of 4H-SiC by thermal decomposition and characterized by Raman spectroscopy and magneto-spectroscopy. They consist of ~70 electronically uncoupled graphene layers.
For excitation energies below 70 meV the relaxation dynamics is determined by an initial decay with a time constant of 25 – 40 ps and a slower component of the order of 200 ps. At higher photon energies (245 meV) the relaxation dynamics is significantly faster with a fast component of 1.2 ps and a slower component in the range from 4 – 8 ps, depending on the substrate temperature. We attribute the faster decay and the different temperature dependence of the pump-probe signals to contribution of optical phonons at the higher energy. At a photon energy of 245 meV the zone center LO phonon (196 meV) can contribute to *interband* recombination, the out of plane flexure phonons can contribute to the *intraband* relaxation.

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