CPP 34: Focus: Spatio-Temporal Multiscale Optical Spectroscopy Meets Functional Materials (joint session DF/CPP/O, organized by DF)

In this Focus Session, we want to bring together scientists from transient spectroscopy as well as materials scientists, concentrating on the physics of (light-induced) excitation, transport, and relaxation in functional materials. Besides presentation of experimental techniques for spatio-temporal multiscale spectroscopy, the emphasis is laid on models and analysis tools needed to understand experimental findings beyond time constants. The Focus Session is meant to join insights and interests, potentially establishing new collaborations between "different" communities.

Organizer: Christoph Merschjann TU Berlin

Time: Wednesday 9:30–12:30 Location: GER 37

Topical Talk CPP 34.1 Wed 9:30 GER 37

Mobile electronic excitations studied by ultrafast spectroscopy — •Stefan Lochbrunner, Franziska Fennel, Steffen Wolter, and Tim Völzer — Institute of Physics, University of Rostock, Albert-Einstein-Str. 23, 18059 Rostock, Germany

The design of materials with specific photonic properties attracts currently intense scientific attention since they are the basis for many future applications in the fields of solar cells, detectors or light emitting devices. Understanding the behavior of light induced excitations in the materials is crucial for developing promising strategies. We apply time resolved spectroscopy on time scales ranging from femto- to milliseconds to characterize the sequence of processes and appearing species which result from the absorption of light. Two examples are presented in the talk. The migration of excitons due to Förster energy transfer is studied using a guest-host system based on dye molecules incorporated with a high concentration in PMMA. We characterized the influence of energetic disorder on the exciton mobility and developed a modified Förster theory which takes this effect correctly into account [1]. The dynamics of charge recombination in transition metal dichalcogenides is investigated by means of MoS_2. Evidence for defect assisted, bimolecular recombination is found by modelling the excitation dependent signal decay with corresponding rate equations.

[1] F. Fennel and S. Lochbrunner, Phys. Rev. B **92** (2015), 140301.

CPP 34.2 Wed 10:00 GER 37

Diffuse transient absorption: a tool for investigation of powdery functional materials — •Christoph Merschjann — Freie Universität Berlin, Fachbereich Physik, Arnimallee 14, D-14195 Berlin, Germany — Universität Rostock, Institut für Physik, Albert-Einstein-Straße 23-24, D-18059 Rostock, Germany

Transient absorption (TA) spectroscopy is a wide-spread and versatile tool for investigating dynamic processes induced in functional materials. Typical quantities of interest include generation, spatial transport, separation (e.g. at interfaces), and ultimate recombination of (photo)excited charge-carriers, polarons, excitons, etc.
However, most novel functional solids are initially synthesized in powder form, and many applications (e.g. catalysis) demand for rather sponge-like or otherwise rough morphologies. Standard TA spectroscopy, which typically utilizes either direct transmission or specular reflection geometries, does not perform too well for such highly scattering materials.

Here, we present a femtosecond TA spectroscopy setup using diffuse reflection/transmission geometry, showing good results for a variety of functional materials, like polymeric carbon nitrides and TaON. A major challenge is the analysis of the retrieved data. Contrary to standard TA, a simple application of Lambert-Beer's law is not possible due to a wide distribution of optical pathlengths in the sample. Models and tools for photon propagation and mutual as well as light-matter interaction, to be used alongside those models needed for the kinetics of the excited states themselves, will be discussed.

Topical Talk CPP 34.3 Wed 10:20 GER 37

Time-resolved characterization of photoactive materials using terahertz spectroscopy — •Rainer Eichberger — Helmholtz-Zentrum Berlin für Materialien und Energie, Germany

Charge carrier mobility and lifetime are key properties of photoactive semiconductor materials. To develop and better understand novel functional materials that enable direct conversion of sunlight into electricity or chemical fuels it is essential to characterize charge generation and separation as well as carrier transport and recombination processes with high time resolution and sensitivity. In conjunction, time-resolved terahertz spectroscopy (TRTS) and microwave conductivity (TRMC) can probe a broad photoconductivity time window from a few hundred femtoseconds to milliseconds. An additional advantage of TRTS is the possibility to measure frequency-dependent conductivity or mobility spectra by transferring the signal into the Fourier space, which gives a deeper insight into the nature of carrier transport and localization. We discuss the dynamics of photo-induced processes such as polaron formation in metal oxides and charge transport issues in polycrystalline chalcopyrite and kesterite thin films arising from carrier localization caused by bandgap fluctuations.

20 min. break

Topical Talk CPP 34.4 Wed 11:10 GER 37

Theoretical simulations of pump-probe spectroscopies in solids — •Michael Sentef — Max Planck Institute for the Structure and Dynamics of Matter, Hamburg

In this talk I will discuss recent progress of theoretical simulations of the nonequilibrium dynamics following laser excitations on femtosecond time scales. I will show two examples: (i) the nonequilibrium dynamics following laser stimulation in a cuprate high-Tc superconductor, where an electron-boson dissipation pathway could be identified in a theoretical-experimental collaboration [1,2,3]; and (ii) the proposed generation of Floquet states in solids with the prospect of engineering effective Hamiltonians [4,5]. I will use this to discuss different theoretical approaches to the nonequilibrium many-body problem - from models to materials - and the future opportunities they offer for the field of pump-probe spectroscopies.

[1] M. A. Sentef et al., Phys. Rev. X 3, 041033 (2013)
[2] A. F. Kemper et al., Phys. Rev. B 90, 075126 (2014)
[3] J. D. Rameau et al., arXiv:1505.07055 → Nature Communcations
[4] M. A. Sentef et al., Nature Communications 6, 7047 (2015)
[5] H. Hübener et al., arXiv:1604.03399 → Nature Communications

CPP 34.5 Wed 11:40 GER 37

Time-dependent quantum transport in nanosystems: A nonequilibrium Green's function approach — •Riku Tuovinen — Max Planck Institute for the Structure and Dynamics of Matter, Center for Free Electron Laser Science, 22761 Hamburg, Germany — Department of Physics, Nanoscience Center, FIN 40014, University of Jyväskylä, Finland

Quantum transport is often discussed in the steady-state regime where the characteristics of the system are described in terms of the energy-dependent transmission or conductance. It is not guaranteed, however, that this description would capture the essential physics in, say, atomic-scale junction operating at high frequencies. Therefore, we consider the full time-dependence which also provides us with "transient spectroscopy" giving detailed information about the nanosystems out of equilibrium. A time-dependent extension to the Landauer–Büttiker approach is presented. The nonequilibrium Green's function approach is employed for describing the charge and heat transport dynamics. The importance of the method is that it provides a closed formula for the time-dependent density matrix in both electronic and phononic systems. In the electronic case the nonequilibrium conditions are due to a switch-on of a bias voltage in the leads or a perturbation in the junction whereas in the phononic case the junction is coupled to reservoirs of different temperatures. In both cases time-dependent transport properties, such as local charge and heat currents, may be evaluated without any propagation. Some applications with, e.g., graphene-based

circuitries are presented and discussed.

Topical Talk CPP 34.6 Wed 12:00 GER 37

Accessing micro- and mesoscopic ultrafast electron dynamics in low-dimensional materials — Michele Puppin[1], Christopher Nicholson[1], Melanie Müller[1], Roman Bertoni[1], Hannes Hübener[2], Angel Rubio[2], Claude Monney[3], Cephise Cacho[4], Martin Wolf[1], Alexander Paarmann[1], Laurenz Rettig[1], and •Ralph Ernstorfer[1] — [1]Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14195 Berlin — [2]Max Planck Institute for the Structure and Dynamics of Matter, Hamburg — [3]Universität Zürich, CH — [4]STFC Rutherford Appleton Laboratory, UK

The coupling of electronic, phononic and spin degrees of freedom is at the heart of most phenomena in condensed matter and governs the flow and relaxation of charge carriers in out-of-equilibrium conditions. Microscopic coupling and correlation effects can be accessed quantitatively by time-, energy- and momentum-resolved information on ultrafast electron dynamics as obtained by time- and angle-resolved photoelectron spectroscopy (trARPES). I will discuss the generation of spin-, valley- and layer-polarized excited states in the semiconductor WSe_2 [1], their ultrafast evolution from 2D to 3D states, and the signatures of transient excited-state many-body interactions. In addition, the visualization of the spatio-temporal evolution of photocurrents in a nanowire by means of femtosecond point-projection microscopy will be discussed [2,3].

[1] Bertoni *et al.*, Phys. Rev. Lett., in print, arXiv:1606.03218.
[2] Müller *et al.*, Nature Comm. 5, 5292 (2014).
[3] Müller *et al.*, ACS Photonics 3, 611 (2016).