Progresses in NonEquilibrium Green's Functions VII

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National Laboratories of Frascati
27-31 August 2018

Organizers :

- Gianluca Stefanucci, Physics Department Tor Vergata University, Rome, Italy

- Andrea Marini, CNR-ISM, Division of Ultrafast Processes in Materials (FLASHit), Rome, Italy

- Stefano Bellucci, INFN-Laboratori Nazionali di Frascati, Frascati, Italy

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Introduction

With the advent of nanoscale physics and ultrafast lasers it is now possible to directly probe real-time the correlated motion of electrons and nuclei in excited quantum states. In addition, the intensity and profile of the laser field can be tuned to control and manipulate the opto-electronic properties of a wide range of molecules and materials. All these progresses have opened new fields of research like, e.g., molecular transport, nanoelectronics, atto-physics/chemistry, nonequilibrium phase transitions, ultracold atomic gases, optimal control theory, etc.

Experiments are usually carried on large molecules, biological systems and nanostructures whose peculiar dynamical properties are inevitably linked to their atomistic structure. Thus, an ab-initio, time-dependent and quantum-mechanical approach is required for reliable calculations. The aim of this workshop was to

gather together many of the most prominent theoretical and experimental scientists to advance our fundamental understanding of matter under extreme nonequilibrium conditions. Particular emphasis was given to many-body methods like Nonequilibrium Green's Functions Theory (NEGF) and how to combine NEGF with ab initio methods like Density Functional Theory.

Summary

The participants gathered at 2:00 pm in the Salvini lecture hall of the INFN National Laboratories of Frascati, Rome (Italy) on August 27th. Gianluca Stefanucci welcomed everyone and gave a brief introductory speech about the location, facilities, social event as well as the format of talks and posters.

The talks have been grouped in four different topics: Transient Spectrocopies, Electron-Nuclei Interaction, Transport and Cold-Atoms, Nuclear Physics.

The first session was on Transient Spectroscopies and it was opend by on **Monday August 27-th** at 2:20 pm by Stefano dal Conte from Politecnico di Milano who presented experimental results on the ultrafast optical response of transition metal dichalcogenides, focusing on two aspects: time-resolved measurements of exciton dynamics and spin/valley relaxation processes. Two theoretical contributions followed on the same day. The first contribution was from Davide Sangalli from CNR-ISM, Monterotondo — Roma with a talk on how to describe and calculate exciton dynamics and exciton lifetimes starting from the non-equilibrium Green's function equations for the carriers dynamics of semiconductors and 2D materials. The second contribution was given by Enrico Perfetto from CNR-ISM, Monterotondo — Roma with a talk on non-equilibrium Green's function applications to finite systems, specifically on time-resolved Auger decays and charge migration processes in organic molecules.

The first session terminated around 6:00 pm and the participants had a chance to go back to their hotels to then meet again in subgroups and have dinner in the cozy historical center of Frascati which, with more than hundred restaurants at 10 mins walking distance, offered a large variety of food and drinks options.

The participants gathered again the day after, **Tuesdsay August 28-th** at 9:00 am for the morning session on Electron-nuclei interaction. The session was opened by E.K.U. Gross from the Max Planck Institute of Microstructure Physics in Halle who presented a novel approach to go beyond the adiabatic approximation starting from the exact factorization of the full electron-nuclear wave function into a purely nuclear part and a many-electron wave function. Another important aspect of the talk was the perturbative treatment in terms of the nuclear velocities as an alternative to electron-phonon diagrammatics. The session continued with the talk

by Yaroslav Pavlyukh from the University of Kaiserslautern. Pavlyukh presented results for the one-body Green's function at finite temperatures using the Matsubara formalism and discussed the quality of the underlying many-body approximation. He also extended the Koopmans' theorem and connected this extension to optical excitations and transient scenarios. The last talk of the morning was given by Michael Sentef from the Max Planck Institute — Hamburg. Sentef presented recent theoretical progresses in topics of current interest for ultrafast materials science, namely light-enhanced superconductivity and ultrafast switching to hidden ordered states.

After the first morning session we all went to lunch and the participants had a chance to walk through the National Laboratories and get acquainted with the internal streets and buildings.

The afternoon session was again dedicated to Transient Spectroscopies and it was open by Patrick O'Keeffe from CNR-ISM — Roma who introduced us to a new femtosecond laser laboratory and to the present and future pump-probe spectroscopies employed in it. These spectroscopies have been shown to be useful to investigate the ultrafast carrier dynamics in materials and composite systems such as layered solar cells. The second talk was given by Nina Rohringer from DESY— Hamburg. Rohringer introduced us to the physics of high-intensity x-ray and the consequent macroscopic number of core-inverted atoms in a gas, posing interesting challenges to the nonequilibrium Green's function community for the description of these entangled highly excited many-body photon-electron state. The session was closed by Ralph Ernstorfer from Fritz Haber Institute — Berlin with novel experimental time-resolved ARPES spectra of WSe_2. Ernstorfer showed that pumping electrons at the excitonic (subgap) energy a transient photoemission subgap peak was visible. This talk too stimulated many discussions among the participants.

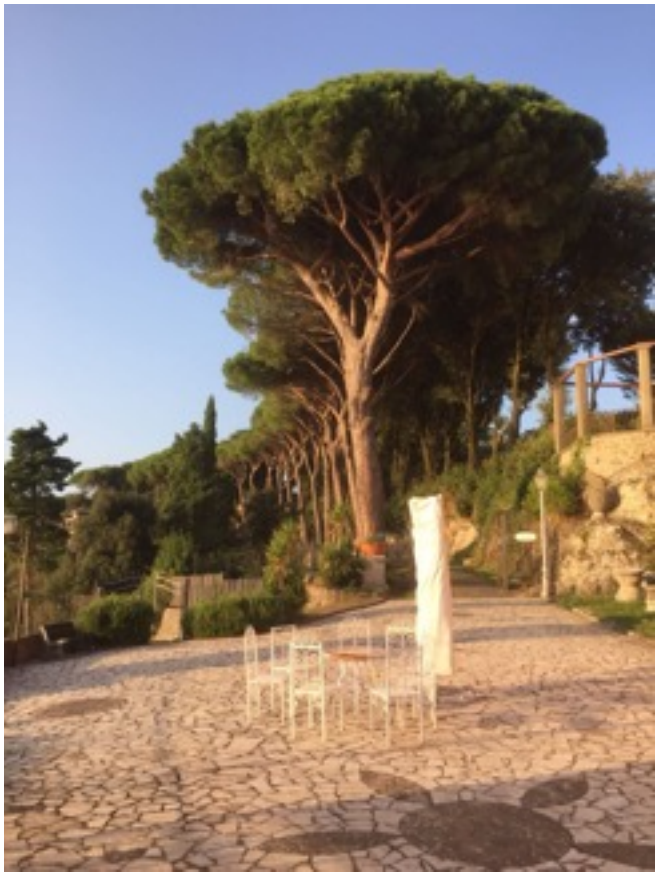
The third day **Wednesday 29-th** started with a morning session on numerical and theoretical developments of the Green's function formalism. The first talk was given by Claudio Verdozzi from the University of Lund and dealt with combining NEGF with Ehrenfest dynamics to assess the role of correlation in molecular motors, proposing an ultrafast correlation induced mechanism to charge separation in donor-acceptor complexes and work in progress on skyrmions. A novel effect on

spin precession was presented by Michael Potthoff from the University of Hamburg. Potthoff showed that conduction-electron correlations as well as energy and spin dissipation result in incomplete spin relaxation, ultrafast damping of quantum nutation and, remarkably, to a novel geometrical spin torque. The morning session was closed by Horia Cornean from the University of Aalborg who presented a mathematically rigorous formulation of the quantum transport problem using NEGF.

After lunch we opened the second session on electron-nuclei interaction. Daniel Karlsson from the University of Jyvaskyla presented a general many-body perturbation theory for electron-boson systems and an advance in the NEGF theory within the Generalized Kadanoff-Baym Ansatz, namely the generalization of the equations to include initial correlations. The second talk was given by Frank Jahnke from the University of Bremen. Using a many-body theory based on the Green's functions technique, Jahnke showed that the thermodynamic fission-fusion balance of excitons and electron-hole plasma can be efficiently tuned via the dielectric environment as well as charge carrier doping. The session was closed by two contributed talks from early stage researchers. Denis Golež presented a talk on the Dynamics of photo-doped charge transfer insulators whereas Yuta Murakami presented a talk on high-harmonic generation in Mott insulators.

At 6:00 pm we all gather in the open space outside the conference building where a bus was waiting for us to go to Villa del Cardinale, see https://www.hotelcastelvecchio.com/villadelcardinale/en/. After the social aperitif in the

breathtaking terrace facing the lake of Castel Gandolfo we moved in the ancient rooms of the villa where the dinner was served.



The fourth day **Thursday 30-th** opened with a session on Transport and Cold Atoms. The first talk was given by Michael Bonitz from Kiel University who presented an overview on his recent developments in NEGF simulations for finite strongly correlated lattice systems. Bonitz showed the time-dependent response of correlated fermionic atoms following a confinement quench and found results in excellent agreement with cold atom experiments. A second application was on the doublon formation in graphene nanoribbons due to the impact of energetic ions. The second talk of the session was given by Andela Kalvova from the Institute of Physics — Praha who put forward a theoretical scheme to go beyond the widely used Generalized Kadanoff-Baym Ansatz. She also demonstrated the accuracy of the scheme in quantum transport with sharp structures in the leads density of states. The session was then closed by three contributed talks from early stage researchers. Christian Schäfer gave a talk on the significance of vertex-corrections on the interface between non-equilibrium greens functions and time-dependent density-functional theory for cavity quantum electrodynamics, Stanislav Kruchinin on Non-Markovian pure decoherence in a dielectric excited by a few-cycle laser pulse and Jan-Philip Joost on carrier multiplication in UV excited graphene nanoribbons.

After lunch the workshop continued with the session on Transport and Cold Atoms. The opening talk was given by Michael Galperin from the University of California San Diego. Galperin presented a formulation of transport and optical response with pseudo-particle NEGF as well as the derivation of a general expression or current-induced forces, showing also the connection to previous studies. Then we heard the talk by Urs Aeberhard from Forschungszentrum — Juelich who reviewed the progress and actual challenges in an alternative framework for theory and simulation of nanostructure photovoltaics. The third talk of the session was given by Tue Gunst from the Technical University of Denmark. Gunst presented a new approach for first-principles modeling of inelastic transport in devices with thousands of atoms. The basic idea consisted in including phonon-assisted tunneling through a single “special thermal displacement” of the atomic coordinates. Gunst also provided evidence of phonon-driven rectification ratio suppression in a diode in good agreement with experiments. The last talk of the session (which was the longest session of the workshop) was given by Antti-Pekka Jauho, also from the Technical University of Denmark. Jauho discussed two recent applications of NEGF. The first was on the Coulomb drag effect in a capacitively

coupled double-quantum-dot where he proposed a theory to go beyond the standard rate equation approach. The second application was the study of a Cooper pair pumping device.

In the last day **Friday 31-st** we had a session on the applications of NEGF in Nuclear Physics. The session was opened by Pawel Danielewicz from Michigan State University who used the Kadanoff-Baym equations to investigate nuclear reactions. The second talk was given by Aurel Bulgac from the University of Washington. Bulgac put forward a Time-Dependent Density Functional Theory for fermionic superfluids and applied successfully to cold atoms, nuclei and neutron star crust. The session and with it the entire workshop was closed by Carlo Barbieri from the University of Surray. Barbieri discussed the crucial parts of a formalism based on NEGF for particle-vibration couplings and three-body forces to account for pairing effects which can be applied to extended systems.

Program

	Monday	Tuesday	Wednesday	Thursday	Friday
	27/8	**28/8**	**29/8**	**30/8**	**31/8**
		Electron-nuclei interaction	**Numerical/theoretical methods**	**Transport and cold atoms**	**Nuclear physics**
09:00 - 09:50		E. K. U. Gross	C. Verdozzi	M. Bonitz	P. Danielewicz
09:50 - 10:40		Y. Pavlyukh	M. Potthoff	A. Kalvova	A. Bulgac
10:40 - 11:10		BREAK	BREAK	BREAK	BREAK
11:10 - 12:00		M. Sentef	H. Cornean	Christian Schäfer Stanislav Kruchinin	C. Barbieri
14:00 - 14:20	Welcome Intro			Jan-Philip Joost	
	Transient spectroscopies	**Transient spectroscopies**	**Electron-nuclei interaction**	**Transport and cold atoms**	
14:20 - 15:10	S. Dal Conte	P. O'Keeffe	D. Karlsson	M. Galperin	
15:10 - 16:00	D. Sangalli	N. Rohringer	F. Jahnke	U. Aeberhard	
16:00 - 16:30	BREAK	BREAK	BREAK	BREAK	
16:30 - 17:20	E. Perfetto	R. Ernstorfer	Denis Golež Yuta Murakami	T. Gunst	
17:20 - 18:00				A. P. Jauho	
18:00 ——			Social Dinner		

The program can be found in the table below.

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Abstracts

Stefano Dal Conte

Non-equilibrium physics of single layer transition metal dichalcogenides and related heterostructures

In layered semiconductors, such as transition metal dichalcogenides (TMD), the electron-electron interaction is strongly enhanced by quantum confinement effect [1]. Furthermore, in these materials the valley polarization can be optically controlled by means of circularly polarized light [2]. Here we discuss the ultrafast optical response of TMDs, focusing on two aspects: time-resolved measurements of exciton dynamics and spin/valley relaxation processes. We first study exciton dynamics in single-layer (1L) MoS_2 by broadband femtosecond transient absorption spectroscopy combined with time- resolved ab-initio simulations [3] based on the non-equilibrium Green's functions and density- functional theory. This study shows that the non-equilibrium optical response of TMDs is determined by the renormalization of the Coulomb screening induced by photoexcited carriers. This effect causes a shift of both the band gap and the exciton binding energy and gives rise to the characteristic derivative-like line shape of the transient optical response. We further use sub-20-fs pump pulses to study the temporal dynamics of the A/B excitons of 1L-MoS_2. We find that the rise time τ_{rise} of the exciton dynamics has an extremely fast but finite value, which raises at increasing energy of the pump excitation, reaching a maximum value of ~35 fs when the pump

energy is tuned above the quasi- particle bandgap. This result is consistent with a scenario where free carriers, initially excited above the quasi-particle gap, relax towards lower energy states and finally form the exciton state via the strong Coulomb interaction. The exciton valley relaxation dynamics of 1L-MoS_2 is investigated by time-resolved Faraday rotation and circular dichroism[4]. We observe a double exponential decay, with an initial fast (~200 fs) decay due to the scattering of spin-polarized excitons from K to K'. An efficient scattering mechanism mediated by phonons and an electron/hole exchange interaction mechanism have been proposed to explain the fast intervalley scattering dynamics. Furthermore, we use two-colour helicity-resolved pump-probe spectroscopy in order to disentangle the intervalley and intravalley spin-flip processes of electrons in the conduction band of 1L-WS_2[5]. Spin-polarized carriers are injected by a circularly polarized pump pulse resonant with the A exciton, while the co-circularly polarized probe pulse is tuned around the B excitonic peak. In this configuration, the scattering of the electrons from the upper to the lower conduction band level (where they cannot radiatively recombine) is detected by measuring the build-up dynamics of the bleaching signal around the B exciton caused by Pauli blocking. We also show that spin-conserving intervalley scattering dynamics occur on a faster time-scale than the intraband spin-flip process. Finally, we study ultrafast charge transfer process in type II heterostructures based on 1L-TMDs, showing that the photoexcited holes can efficiently scatter from $MoSe_2$ layer to WSe_2 layer within 1 ps.

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[3] E.A.A. Pogna *et al.* ACS Nano **10**, 1182–1188 (2016).
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Davide Sangalli

Non equilibrium dynamics and lifetimes within many-body perturbation theory: from free carriers to excitons

The Kadanoff-Baym equation reduces to an equation of motion for the one-body density matrix within the Generalized Kadanoff Baym ansatz. Moreover, introducing a Markovian and a Boltzmann like approximation for the collision integral, its ab-initio implementation becomes feasible also for extendend systems [1-3]. Indeed we have shown it well describes carriers dynamics, lifetimes and transient spectroscopy in bulk silicon[4-6]. The interpretation of the physics captured is strightforward, in terms of coherent and the non-coherent dynamics of electrons and holes, in particular if the static part of the self-energy is kept at equilibrium. Correlation however need to be switched on to describe renormalization effects, coherent processes and decoherence, the absorption of light at excitonc resonances [1] and the physics of non-coherent excitons [7-8]. It becomes then non trivial to give a simple physical interpretation to the resulting equations. Starting from carriers dynamics I'll discuss how such different physical aspects can be tackled within the non-equilibrium Green's function formalism, still connecting the equations to clear physical concepts. I'll focus in particular into the concept of exciton dynamics and exciton lifetimes [8].

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Enrico Perfetto

First-principles nonequilibrium Green's function approach to real-time simulations of correlated electrons in molecular systems

We show how to describe the correlated electron dynamics of molecules and molecular complexes driven by ultrafast laser pulses using the first-principles nonequilibrium Green's function (NEGF) method [1-5]. The method is currently implemented in the CHEERS code [6] which allows for addressing a broad variety of time-resolved phenomena such as transient photoabsorption and photoemission, time-resolved Auger decays as well as ultrafast charge migration. CHEERS is compatible with all main single-particle basis functions (plane-waves, Gaussian type orbitals, Slater type

orbitals, real-space grid etc.) and it accounts for both static and dynamical correlation effects.
We present proof-of-concept results for the ultrafast Auger process and highlight universal features of the outgoing wavepacket, namely a highly asymmetric profile with a long tail exhibiting ripples temporally spaced by the inverse of the Auger energy [3]. As a second application we focus on molecules of biological relevance and address the charge migration process initiated by attosecond ionizing pulses [7], a topic of high current interest to unravel the fundamental mechanisms at the basis of photo-protection or photo-damage. The role played by correlations and memory effects is crucial to achieve an excellent agreement with available experimental data [7-8].

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E.K.U. Gross

Nuclear velocity perturbation theory within the exact

factorization: An alternative to electron-phonon diagrammatics?

The starting point of essentially all modern electronic-structure techniques is the Born-Oppenheimer (BO) approximation. It not only makes calculations feasible, the motion of nuclear wave packets on the lowest BO potential energy surface often provides us with an intuitive picture of chemical reactions. To go beyond this limit is notoriously difficult. Here we present a novel approach to non-adiabatic effects that is based on the exact factorization [1] of the full electron-nuclear wave function into a purely nuclear part and a many-electron wave function which parametrically depends on the nuclear configuration and which has the meaning of a conditional probability amplitude. The variational principle leads to two equations of motion., one for the nuclear wave function and one for the electronic conditional amplitude. The equation for the nuclear factor is a standard Schrödinger equation which describes the nuclear motion – formally exactly- on a single time-dependent potential energy surface. When there is a splitting of the nuclear wave packet in the vicinity of an avoided crossing of BO surfaces, this exact time-dependent surface shows a nearly discontinuous step [2] connecting different BO surfaces, thus incorporating the influence of multiple adiabatic surfaces. The electronic equation of motion is not a standard Schrödinger equation: In addition to the usual BO Hamiltonian it contains terms which are non-linear, non-local and non-Hermitian. These terms are responsible for the proper description of decoherence [3,4]. Here we treat these additional terms by perturbation theory, the small parameter being the ratio of electronic over nuclear mass (m/M). These terms contain the standard electron-phonon interaction, but include other terms as well in lowest order of (m/M). First-order perturbation theory in these terms provides a successful description of

electronic flux densities [5] and vibrational circular dichroism [6]. Furthermore, we explore the possibility of describing non-adiabatic effects in, e.g., proton transfer by R-dependent nuclear masses [7].

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Yaroslav Pavlyukh

Benchmarking approximations of many-body perturbation theory for molecular systems

I will present results for many-body perturbation theory for the one-body Green's function at finite temperatures using the Matsubara formalism. Our method relies on the accurate representation of the single-particle states in standard Gaussian basis sets, allowing to efficiently compute, among other observables, quasiparticle energies and Dyson orbitals of atoms and molecules. In particular, we investigate the second-order treatment of the Coulomb interaction by benchmarking its accuracy for a well-established test set of molecules, which includes also systems where the usual Hartree-Fock treatment encounters difficulties. We discuss

different schemes such as Padé approximation [1] and the extended Koopmans' theorem to extract quasiparticle properties, and assess their range of applicability [2]. For the latter, generalisations to describe optical excitations and transient scenarios via the Floquet approach are presented, connections to the reduced density matrix formalism [3] are given. With an accurate solution and compact representation, our method represents a starting point to study electron dynamics in time-resolved experiments by the propagation of the Kadanoff-Baym equations [4].

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Michael Sentef

Theory of pump-probe spectroscopy: Ultrafast laser engineering of ordered phases and microscopic couplings

Intense femtosecond laser pulses, spanning a large range of photon energies from the X-ray to the THz regime, allow for controlled excitations ("pump") and monitoring ("probe") of the nonequilibrium dynamics of all the relevant microscopic degrees of freedom in solids. The field of ultrafast materials science is currently evolving from measuring time constants - for

instance for the decay of hot electrons via phonon emission - towards ultrafast laser engineering of nonthermal phases of matter with novel properties. Notable examples include light-induced superconducting-like behavior [1], ultrafast switching to hidden ordered states [2], or time-reversal symmetry-broken Floquet states in topological insulators [3].
I will discuss recent theoretical progress in understanding these diverse phenomena from microscopic models and nonequilibrium simulations. I will show examples of light-enhanced superconductivity in an electron-phonon system from classical nonlinear phononics [4,5] and laser-controlled order competition between superconductivity and charge-density waves [6]. I will discuss laser engineering of microscopic couplings in graphene [7] based on quantum nonlinear phononics [8,9]. I will also show ab initio time-dependent density functional theory results for laser-engineered Hubbard U in NiO [10], with a recent application to light-induce the elusive magnetic Weyl semimetal in pyrochlore iridates [11].
I will also highlight some recent developments towards cavity quantum electrodynamical environments enhancing electron-phonon coupling in 2D materials [12], and efforts to make nonequilbrium Green's functions fast for correlated ordered phases (excitonic condensates) using the generalized Kadanoff-Baym ansatz [13].

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Patrick O'Keeffe

The Use of Optical Pump-Probe Spectroscopies to interrogate Hot Carrier Production in Materials

A new femtosecond laser laboratory, situated in the ISM institute of the CNR at the Research Area of Tor Vergata near Rome, will be presented. The present and future pump-probe spectroscopies employed in the laboratory such as transient absorption, fluorescence up-conversion, pump-probe SNOM microscopy and high harmonic generation, will be outlined. These spectroscopies have been used to investigate the ultrafast carrier dynamics in materials and composite systems such as layered solar cells. A particular field of interest for the group is the photogeneration hot non-thermal electron distributions in metal nanostructures and the effect of the type of excitation be it plasmonic excitation or direct photexcitation on the generation and thermalisation of these carriers. The possible applications of these short lived hot electrons distributions will also be discussed. Two examples will be cited the first of which is the sensitization of wide band gap semiconductors to visible light by the incorporation of metal nanostructures with particular reference to silver nanoparticles embedded in a film of CeO_2. The second example is the morphologically controlled melting of metal nanostructures which has allowed us to shape metal structures starting from an aggregate of spherical gold nanoparticles [1,2]. Finally, the effect of the incorporation of graphene into a perovskite cell during the production process on production of hot carriers in the perovskite layer will be discussed with reference to recent experiments performed by the group [3].

[1] *"Plasmon Controlled Shaping of Gold Nanoparticle Aggregates by Femtosecond Laser Induced Melting"*, by D. Catone, A. Ciavardini, L. Di Mario, A. Paladini, F. Toschi, A. Cartoni, I. Fratoddi, I. Venditti, A. Alabastri, R. Proietti Zaccaria, and P. O'Keeffe (submitted).

[2] *"Gold nanoparticles functionalized by rhodamine B isothiocyanate: A new tool to control plasmonic effects."*
Fratoddi I, Cartoni A, Venditti I, Catone D, O'Keeffe P, Paladini A, Toschi F, Turchini S, Sciubba F, Testa G, Battocchio C, Carlini L,
Proietti Zaccaria R, Magnano E, Pis I, Avaldi L, J. Colloid Interface Sci. **513**, p10-19 (2018).

[3] "*Graphene preserves the hot-carrier temperature of perovskite solar cells*", P. O'Keeffe, D. Catone, A. Paladini, F. Toschi, S. Turchini, L. Avaldi, F. Martelli, A. Agresti, S. Pescetelli, A. Esaú Del Rio Castillo, F. Bonaccorso and A. Di Carlo (submitted).

Nina Rohringer

Collective decay mechanisms following saturated inner-shell x-ray excitation

X-ray free-electron lasers are available for experiments for almost a decade and allow for hitherto unachievable x-ray intensities on sample, reaching up to 10^{21} W/cm^2 for hard x-rays. At these intensities, the interaction probability for photo absorption of a single atom or molecule with one x-ray pulse reaches unity. Moreover, several interactions within one pulse – nonlinear x-ray mater interactions – become possible, opening the pathway to nonlinear x-ray optics. For a macroscopic ensemble of atoms, molecules, nanometer-sized clusters, or a solid the interaction with a strongly focused x-ray beam can create a macroscopic, highly excited state of matter, far from equilibrium. In particular, saturated absorption with a high-intensity

x-ray pulse can result in transient states, for which every single atom in the interaction region is in a core-inverted state. This macroscopic population inversion can lead to collective radiative decay mechanisms, such as amplified spontaneous emission or superradiance. I will present recent experimental results of amplified spontaneous K-α emission in transition metal compounds in solution, and superradiance of Xenon atoms following 4d-shell ionisation. Currently we have two theoretical approaches to describe the collective, radiative relaxation dynamics of the system. The conventional approach relies on the solution of generalised Maxwell-Bloch equation – we solve the Liouville - von Neumann equation of motion for the density matrix of a few-level system coupled to a classical radiation field. In this approach, stochastic source terms have to be introduced, to mimic spontaneous decay. A more sophisticated quantum-electrodynamics approach for an atomic few-level system relies on the Heisenberg-operator equation of motion and after a series of approximations solves for the coupled equation of motion of two-point correlation function of the electronic coherences and field correlation functions. This approach treats amplified spontaneous emission and superradiance on the same footing. In both approaches, electronic decay channels (Auger decay) are treated phenomenologically, by appropriate decay terms of the coherences and population. In this forum, I would like to address the question, to which extend non-equilibrium Greens function approaches could be appropriate to treat electronic decay in a dynamic, ab-initio way. In a highly excited macroscopic system, collective electronic decay channels might arise, but currently we have no means to describe these decays. Motivated by our experiments, I will present a few model systems and Hamiltonians, that could serve as the basis for considering collective electronic decays and I am looking forward to an open discussion, if non-equilibrium Greens function techniques could provide a tool set to tackle these difficult problems.

Ralph Ernstorfer

Accessing electron- and exciton-phonon coupling in solids with momentum-resolving ultrafast techniques

We investigate basic material properties like electronic and atomic structure of crystalline materials in laser-prepared non-equilibrium states with time- and angle-resolved photoelectron spectroscopy (trARPES) and femtosecond electron diffraction (FED). The momentum-resolved view of the structure and dynamics of excited electrons and phonons provides information on microscopic coupling phe- nomena, e.g. the coupling of electronic and vibrational degrees of freedom. We aim for a quantum- state-resolved picture of coupling on the level of quasi-particle self-energies, which goes beyond established ensemble-average descriptions. I will exemplify this experimental approach by discussing electron, exciton and phonon dynamics in the semiconducting transition metal dichalcogenide WSe_2. TrARPES reveals the excited state band structure, the k-space distribution, energy and evolution of excitons and single-particle excited states. In addition, we demonstrate the optical preparation of spin- and pseudospin-polarized excited states in this centrosymmetric semiconductor [1]. The complementary momentum-resolved view on phonon dynamics is obtained by FED [2]. By combining this information, a microscopic picture of electron-lattice coupling and energy dissipation emerges.

[1] R. Bertoni et al., Phys Rev. Lett. **117**, 277201 (2016).
[2] L. Waldecker et al., Phys. Rev. Lett. **119**, 036803 (2017).

Claudio Verdozzi

Merging at different levels NEGF and Ehrenfest dynamics for electronic charge-transfer, electronic friction, and work in progress

We use NEGF or NEGF in the GKBA coupled to the Ehrenfest Dynamics (EA) to investigate electron and nuclear dynamics in donor-acceptor complexes and in molecular motors in quantum transport setups. For donor-acceptor complexes, we show that ultrafast charge separation driven by correlation-induced decoherence can in principle occur in organic nanoscale systems, but it will only be correctly predicted by theoretical treatments that include time-nonlocal correlations. Within the EA, these results are largely unaffected by nuclear vibrations; the latter become however essential whenever level misalignment hinders the CT process. For molecular motors, we use the adiabatic limit of the KBE combined with the EA to obtain the current-induced forces on the nuclei. Results for a model molecular motor show that correlations can change dramatically the physical scenario by, e.g., introducing a sizable damping in the self-sustained van der Pol-like oscillations due to positive electronic friction. Finally, we present work in progress, where EA and NEGF-GKBA are used for spin-spin interactions, and address the plausibility of hybrid NEGF+ DFT schemes for electron-phonon interactions.

Michael Potthoff

Effects of electron correlations and topology in the real-time dynamics of the Kondo model

We study a single spin in an external magnetic field which is antiferromagnetically coupled to an extended system of conduction electrons. Real-time dynamics is initiated by a sudden switch of the field direction. For fields stronger than the Kondo temperature, the problem is

solved numerically by means of tight-binding spin dynamics treating the spin as a classical observable, by time-dependent density-matrix renormalization group (t-DMRG) [1], by t-DMRG in combination with linear-response theory [2], and by adiabatic spin dynamics [3] enforcing the conduction-electron system to be in its instantaneous ground state.
Conduction-electron correlations as well as energy and spin dissipation are shown to result in various anomalous effects, such as incomplete spin relaxation due to emergent time-scale separation and ultrafast damping of quantum nutation. A novel geometrical spin torque is responsible for an anomalous precessional motion with a strongly enhanced effective Larmor frequency.

[1] M. Sayad, R. Rausch and M. Potthoff, Europhys. Lett. **116**, 17001 (2016)
[2] M. Sayad, R. Rausch and M. Potthoff, Phys. Rev. Lett. **117**, 127201 (2016)
[3] C. Stahl and M. Potthoff, Phys. Rev. Lett. **119**, 227203 (2017)

Horia Cornean

Nonequilibrium Green's Functions from a mathematical point of view

We will revisit the celebrated Jauho–Meir–Wingreen formula for the time-dependent current through an interacting sample coupled to non-interacting leads, and show how it can be derived
in a non-perturbative way. Our method employs neither complex-time Keldysh contours, nor Langreth rules of 'analytic continuation.' It is only based on Duhamel type identities and the KMS property of the partitioned initial state. If time permits, we will also study the Dyson equation for the advanced/retarded interacting Green's function and construct its (irreducible) self-energy using the theory of Volterra operators. The talk is based on a recent joint work with V. Moldoveanu and Claude-Alain Pillet.

Daniel Karlsson

Progress in non-equilibrium Green's functions: coupled fermion-boson systems and the GKBA with initial correlations

We present recent results within the fundamentals of the non-equilibrium Green's function (NEGF) formalism. We focus on two separate aspects:
i) We briefly review the NEGF formalism for coupled fermion-boson systems. Traditionally, fermions and bosons are often treated differently in the literature. Instead, we stress the similarities of the equations of motion for the fermionic and bosonic Green's functions using the contour formalism. The decoupling of the non-interacting n-particle Green's function, i.e., Wick's theorem, is then almost identical in the fermionic and bosonic case, and a unified formalism is obtained. [1]
ii) We extend the Generalized Kadanoff-Baym Ansatz (GKBA) to allow for initially correlated states. Within the NEGF formalism, the GKBA has stood out as a computationally cheap method to investigate the dynamics of interacting quantum systems driven out of equilibrium. Current implementations, however, suffer from a drawback: real-time simulations require noncorrelated states as initial states. Consequently, initial correlations must be built up through an adiabatic switching of the interaction before turning on any external field, a procedure that can be numerically highly expensive. In this work, we extend the GKBA to allow for initially correlated states which makes it possible to efficiently separate the calculation of the initial state from the real-time simulation. [2]

[1] Daniel Karlsson and Robert van Leeuwen, *Non-Equilibrium Green's Functions for coupled fermion-boson systems*, to appear in Handbook of Materials Modeling. Volume 1 Methods: Theory and Modeling

[2] Daniel Karlsson, Robert van Leeuwen, Enrico Perfetto and Gianluca Stefanucci, *The Generalized Kadanoff-Baym Ansatz with Initial Correlations*, arXiv:1806.05639

Frank Jahnke

Excitons versus electron-hole plasma in monolayer transition metal dichalcogenide semiconductors

When electron-hole pairs are excited in a semiconductor, it is a priori not clear if subsequent relaxation leads to a gas of bound excitons or to an interacting plasma of unbound electrons and holes. Usually, the exciton phase is associated with low temperatures. In atomically thin transition metal dichalcogenide semiconductors, excitons are particularly important even at room temperature due to strong Coulomb interaction and a large exciton density of states.

Using a many-bodytheory based on the Green's functions technique, we show that the thermodynamic fission-fusion balance of excitons and electron-hole plasma can be efficiently tuned via the dielectric environment as well as charge carrier doping [1]. We observe entropy ionization of excitons at low excitation densities and a Mott transition to a fully ionized plasma at high densities between $3\text{x}10^{12}$ cm^{-2} and $1\text{x}10^{13}$ cm^{-2} depending on experimental parameters. Below the Mott transition, excitons become dominant with maximal fractions of excitons between 70 % and more than 99.9 %. Moreover, we find that excitonic screening, although two orders of magnitude less efficient than free-carrier screening at comparable excitation densities, plays an important role in the description of the exciton-plasma balance.

We propose the observation of these effects by studying exciton satellites in photoemission and tunneling spectroscopy, which are sensitive to the

single-particle spectral functions, thus containing information about the degree of exciton fission and the extent of exciton wave functions in reciprocal space. Photoemission spectroscopy presents a direct solid-state counterpart of high-energy collider experiments on the induced fission of composite particles.

[1] A. Steinhoff, M. Florian, M. Rösner, G. Schönhoff, T. O. Wehling and F. Jahnke, Nature Communications **8**, 1166 (2017).

Heiko Appel

Time-dependent density-functional theory for QED: an ab initio framework for modeling light-matter interactions

This talk provides an overview of the recent work in our group on density functional theory (DFT) for quantum electrodynamics (QED) [1-8]. The first part of the presentation introduces the foun- dations of time-dependent DFT for QED. Compared to traditional density functional theory, our approach allows to explicitly include the quantized photon field in a density functional description. While this becomes important for small photon numbers and large field fluctuations, the limit of large photon numbers and small electromagnetic field fluctuations is well described by a Hartree level of approximation. For a real-time evolution, this corresponds to coupled Ehrenfest-Maxwell- Kohn-Sham equations. We illustrate this limit for molecules exposed to strong magnetic fields, as well as molecules exposed to strong femtosecond laser fields.

To describe larger electromagnetic field fluctuations, we introduce as further example how trajec- tory methods that have originally been developed for coupled electron-nuclear dynamics can be extended to describe light-matter coupling beyond the Maxwell-Schro¨dinger limit. In

particular, we illustrate how to assess correlation functions of the quantized photon field as well as sponta- neous emission processes with such trajectory approaches and discuss the significance for photo- stimulated processes.

In the remaining part of the presentation, we are focussing on atomic and molecular systems em- bedded in optical cavities or nanoplasmonic environments, where the matter-photon interaction is considered from the weak- to the strong-coupling limit and for individual photon modes as well as for the multimode case. We introduce a recently proposed cavity Born-Oppenheimer approxima- tion and identify fundamental changes in Born-Oppenheimer surfaces, spectroscopic quantities, and conical intersections.

[1] M. Ruggenthaler et. al. Phys. Rev. A **90**, 012508 (2014).
[2] C. Pellegrini et.al., Phys. Rev. Lett. **115**, 093001 (2015).
[3] J.Flicket.al.PNAS **112**, 15285(2015).
[4] J. Flick et. al., J. Chem. Theory Comput. **13**, 1616 (2017).
[5] J. Flick et. al. PNAS **114**, 3026 (2017).
[6] J. Flick et. al. ACS Photonics **5**, 992 (2018).
[7] J. Flick et. al. arXiv:1803.02519
[8] M. Ruggenthaler et. al. Nature Reviews Chemistry **2**, 0118 (2018).

Michael Bonitz

Femtosecond correlated electron dynamics in finite graphene clusters

We give an overview on our recent developments in Nonequilibrium Green functions (NEGF) simulations for finite strongly correlated lattice systems. This includes implementation of novel selfenergy approximations, computational optimizations and GPU computing that have allowed us to gain several orders of magnitude in performance. The accuracy of NEGF simulations has been benchmarked against DMRG results [1]. We then

present recent applications. The first is the time-dependent response of correlated fermionic atoms following a confinement quench where our results are in excellent agreement with cold atom experiments [2]. The second application is doublon formation in graphene nanoribbons (GNR) due to the impact of energetic ions [3, 4]. Finally we present NEGF results for the optical excitation of GNR by a UV laser pulse and investigate carrier multiplication [5].

[1] N. Schlünzen et al., Phys. Rev. B **95**, 165139 (2017)
[2] N. Schlünzen et al., Phys. Rev. B **93**, 035107 (2016)
[3] K. Balzer et al., Phys. Rev. B **94**, 145118 (2016)
[4] K. Balzer et al., submitted for publication, arXiv: 1801.05783
[5] J. P. Joost et al., to be published

Fabian Heidrich-Meisner

Transport properties of the Fermi-Hubbard model

The Fermi-Hubbard model can readily be realized with ultracold atomic gases. Recent experiments investigated static correlations in 1d and 2d systems. Here, we focus on transport in the 1d case. Due to its integrability, some transport coefficients diverge even at finite finite temperature. For instance, the 1d Fermi-Hubbard model is always a ballistic energy conductor. I will discuss recent numerical advances in computing the finite temperature transport coefficients of this model, both in ballistic and potentially diffusive regimes. Furthermore, I will present a concrete proposal of how to measure diffusion constants and Drude weights in optical lattice experiments using quantum-gas microscopes. In a second part, I will discuss aspects of the nonequilibrium transport in the same model, recently studied in a joint theory-experiment collaboration.

[1] Jin et al. PRB **92**, 205103 (2015)
[2] Karrasch, Kennes, Heidrich-Meisner, PRL **117**, 116401 (2016**)**

[3] Karrasch, Prosen, Heidrich-Meisner, PRB **95**, 060406 (2017)
[4] Scherg et al.arXiv:1805.10990

Andela Kalvová

Beyond the Generalized Kadanoff-Baym Ansatz

The numerical study of the non-equilibrium electron transport can often be made much easier if the complete Kadanoff-Baym equations are simplified using the Generalized Kadanoff-Baym Ansatz (GKBA). For the class of molecular bridges, the Ansatz is empirically known to safely work for the case of a weak tunneling and flat tunneling functions. It fails, if either of the conditions is not satisfied. The case in point is a molecular bridge formed by an Anderson type local center treated in the mean field approximation and linked by tunneling junctions to two ferromagnetic electrodes whose tunneling spectral functions simulate nickel and have a complex sd structure. The transient magnetic currents under a constant galvanic bias between the electrodes are invoked by sudden switching on of both junctions. We consider three tasks. 1. To establish quantitative criteria for the validity of the Ansatz; 2. To develop a practically tractable correction to the Ansatz working beyond its validity range 3. To obtain the Generalized Master Equations for the one-electron distribution following from both the Ansatz and its corrected form avoiding thus the Kadanoff-Baym equations. All three points are resolved by treating first the stationary (non-equilibrium) limit and transferring the results to finite times. In particular, the corrections to the Ansatz are obtained as a stationary approximation to the vertex part of the exact reconstruction equations whose free term is the standard GKBA.

Michael Galperin

Green function methods for optoelectronics and molecular dynamics

We discuss implementation of Green function methods to simulation of transport and optical response (fluxes, noise and noise spectrum) of molecular junctions comparing non-equilibrium Green's function (NEGF), pseudo-particle NEGF (PP-NEGF) and Hubbard NEGF approaches. We also present general first principles derivation of expression for current-induced forces. The expression is applicable in non-equilibrium molecular systems with arbitrary intra-molecular interactions and for any electron-nuclei coupling. We show connection of the expression with previous studies, and discuss effective ways to evaluate the friction tensor.

Urs Aeberhard

NEGF theory of photovoltaics at the nanoscale – progress and challenges

Nanostructures are ubiquitous in modern high-efficiency solar cell architectures. They assume diverse functionalities, from tunable absorbers to passivation layers and selective contacts. As a common feature, they introduce device regions where the material properties and the photovoltaic processes are no longer adequately described by the semiclassical bulk physics that forms the basis of conventional solar cell device simulation [1-2]. In this talk, we review the progress and actual challenges in an alternative framework for theory and simulation of nanostructure photovoltaics, which is based on the non-equilibrium Green's function formalism (NEGF) in a steady-state formulation that encompasses both quantum transport and quantum optics [3-5] . Starting from the NEGF description of the elementary processes in nanostructure photovoltaics [6-8], the focus is set on the role of self-consistency in the evaluation of scattering self-energies and on the implication of both

localization and non-locality for the evaluation of physical quantities [9]. Finally, some remaining challenges and directions of future research are discussed.

[1] P. Würfel, “Physics of solar cells : from basic principles to advanced concepts”, Wiley-VCH (2009)
[2] J. Nelson, “The physics of solar cells”, Imperial College Press (2003)
[3] U. Aeberhard, “Theory and simulation of quantum photovoltaic devices based on the non-equilibrium Green's function formalism”, J. Comput. Electron. **10**, 394 (2011)
[4] U. Aeberhard, “Quantum-kinetic perspective on photovoltaic device operation in nanostructure-based solar cells” J. Mater. Res. **33**, 373 (2018)
[5] U. Aeberhard, “Photovoltaics at the mesoscale: insights from quantum-kinetic simulation”, J. Phys. D (2018)
[6] U. Aeberhard, “Quantum-kinetic theory of photocurrent generation via direct and phonon-mediated optical transitions”, Phys. Rev. B **84**, 035454 (2011)
[7] U. Aeberhard, “Non-equilibrium Green's function theory of excitonic effects in the photocurrent response of semiconductor nanostructures”, Phys. Rev. B **86**, 115317 (2012)
[8] U. Aeberhard, “Quantum-kinetic theory of steady-state photocurrent generation in thin films: Coherent versus incoherent coupling”, Phys. Rev. B **89**, 115303 (2014)
[9] U. Aeberhard, “The non-equilibrium Green's function picture of inelastic processes in nanostructure photovoltaics”, J. Comput. Electron. **15**, 1219 (2016)

Tue Gunst

New approaches for first-principles modeling of inelastic transport in devices with thousands of atoms

In the race towards high-performance nanometer-scaled devices the electronics industry now faces a major challenge from phonon-assisted tunneling. Despite the rapid size-reduction in experiments, system sizes still fall outside what is feasible for existing device models including electron-phonon coupling from first-principles. Therefore, the role of phonon-assisted tunneling in sub-10-nanometer gate-length devices has not been accurately quantified so far.

I present transport simulations based on density functional theory combined with nonequilibrium Green's functions. I compare two methods which both enable large-scale first-principles device simulations including electron-phonon coupling (EPC). The first method is based on first order perturbation theory in the EPC self-energy while the second includes phonon-assisted tunneling through a single "special thermal displacement" of the atomic coordinates at almost the same cost as elastic transport calculations[1]. I apply the methods to ultrascaled silicon devices and demonstrate the importance of phonon-assisted band-to-band and source-to-drain tunneling. In a diode the phonons lead to a rectification ratio suppression in good agreement with experiments, while in an ultrathin body transistor the phonons increase off currents by four orders of magnitude, in agreement with our state-of-the-art perturbation theory calculations. In addition, electron-phonon coupling of nanostructured devices in operation conditions can change significantly from its bulk value[2]. This makes the methods an appealing design tool for next-generation devices and nanomaterials.

[1] T. Gunst *et al.*, Phys. Rev. B **96**, 161404(R) (2017).
[2] T. Gunst *et al.*, Phys. Rev. Lett. **118**, 046601 (2017).

Antti-Pekka Jauho

Quantum transport in double quantum dot

Double quantum dots display a host of interesting quantum transport phenomena. Here, I discuss two recent examples [1,2], where the theoretical analysis employs the Nonequilibrium Green's function techniques. In a parallel configuration, where the two dots are separately contacted and interact through Coulomb interaction (no dot-to-dot tunneling takes place) the system may display the phenomenon of Coulomb drag: a current driven through one dot induces a response in the other dot, which may –depending on the detailed circumstances – induce a current or a voltage drop across the second dot. This system has attracted significant recent interest, both in experiment and theory [3,4]. Here, we adapt the Meir-Wingreen formalism to address the Coulomb drag system in a parameter regime that hitherto has escaped attention. Namely, we extend the theory from the Coulomb blockade regime (which is typically treated with a Master Equation approach and assumes a weak dot-to-lead coupling) to larger tunnel amplitudes. We do this by applying a perturbation theory in the Coulomb interaction with appropriate self-energy functionals embedded in the nonequilbrium formalism. We show that the drag is governed by the power spectrum of the fluctuations in the occupancy of the drive system, and delineate carefully the conditions that are necessary for the occurrence of Coulomb drag. In a serial configuration, with superconducting leads, the double dot system can be used as a Cooper pair pump: applying time-dependent gate voltages to the two dots with the same amplitude but a relative phase difference allows one to engineer the supercurrent flowing through the system. We develop a theory for this situation employing the time-dependent Meir-Wingreen formula in the Floquet formulation. Our theory leads to an intuitive picture of the pumping mechanism.

[1] M. A. Sierra, D. Sánchez, K. Kaasbjerg, and A. P. Jauho: "Coulomb drag in interacting quantum dot systems", in preparation

[2] C. Ortega Tabener, A. P. Jauho, and J. Paaske: "Non-adiabatic Cooper pair pumping through a double quantum dot", in preparation
[3] K. Kaasbjerg and A. P. Jauho, Phys. Rev. Lett. **116**, 196801 (2016)
[4] A. J. Keller et al., Phys. Rev. Lett. **117**, 066602 (2016)

Pawel Danielewicz

Quantum Dynamics of Nuclear Slabs: Mean Field and Short-Range Correlations

Computational difficulties aside, nonequilibrium Green's functions appear ideally suited for investigating the dynamics of central nuclear reactions. Many particles actively participate in those reactions. At the two energy extremes for the collisions, the limiting cases of the Green's function approach have been successful: the time-dependent Hartree-Fock theory at low energy and Boltzmann equation at high. The strategy for computational adaptation of the Green's function to central reactions is discussed. The strategy involves, in particular, incremental progression from one to three dimensions to develop and assess approximations, discarding of far-away function elements, use of effective interactions and preparation of initial states for the reactions through adiabatic switching. At this stage we concentrate on inclusion of correlations in one dimension, where relatively few approximations are needed, and we carry out reference calculations that can benchmark approximations needed for more dimensions. We switch on short-range interactions generating the correlations adiabatically in the Kadanoff-Baym equations to arrive at correlated ground states for uniform matter. As the energy of the correlated matter does not quite match the expectations for nuclear matter we add mean field to arrive at the match in energy. From there on, we move to finite systems. In switching on the correlations we observe emergence of extended tails in momentum distributions and evolution of single particle occupations away from 1 and 0. Subsequently, we

differentiate neutrons and protons in the calculations and introduce energy associated with neutron-proton imbalance. We study oscillations of the slabs where neutrons and protons move either in phase or out of phase with each other.

Aurel Bulgac

Time-Dependent Density Functional Theory for Fermionic Superfluids: From Cold Atoms, to Nuclei and Neutron Star Crust

In cold atoms and the crust of neutron stars the pairing gap can reach values comparable with the Fermi energy. While in nuclei the neutron gap is smaller, it is still of the order of a few percent of the Fermi energy. The pairing mechanism in these systems is due to short range attractive interactions between fermions and the size of the Cooper pair is either comparable to the inter-particle separation or it can be as big as a nucleus, which is still relatively small in size. Such a strong pairing gap is the result of the superposition of a very large number of particle-particle configurations, which contribute to the formation of the Copper pairs, unlike electronic systems. These systems have been shown to be the host of a large number of remarkable phenomena, in which the large magnitude of the pairing gap plays an essential role: quantum shock waves, quantum turbulence, Anderson-Higgs mode, vortex rings, domain walls, soliton vortices, unexpected dynamics of fragmented condensates and the role of pairing correlations in collisions on heavy-ions, Larkin-Ovchinnikov phase as an example of a Fermi supersolid, and the role pairing correlations control the dynamics of fissioning nuclei, selfbound superfluid fermion droplets of extremely low densities.

Posters: title and abstract in alphabetical order

Zeno-hindered Auger decay, photon fields and magnetic ordering: Challenges for NEGF?

Emil Viñas Boström

Steady and fast-paced conceptual and computational progress in NEGF has brought several areas of nonequilibrium physics within the reach of a NEGF description, as e.g. cold atoms physics, spectroscopy, quantum transport, Kondo physics, and thermalization.
There is also a number of less explored areas of application which can benefit from NEGF, such as interacting continua, the treatment of electron-photon interactions, and the dynamics of magnetic systems. Here, we provide three case studies aimed to explore implications and potential challenges to be met by NEGF research in these areas. Specifically,
i) We describe how external fields can hinder the the Auger process in time, by inducing the so-called quantum Zeno effect. This allows the lifetime of the Auger decay to be extended to several times its unperturbed value, enough to be measurable in prototypical systems such as the Li atom.
ii) We then consider the fluorescence spectrum of a two-level atom interacting with a quantized photon field. In contrast to the predictions of perturbation theory, we show that the system admits second harmonic generation, a finding supported by an analysis of the parity of the coupled electron-photon states. We also consider how the spectrum changes if atomic motion is included in the description, both in the Ehrenfest approximation and in a full quantum mechanical manner.
iii) Finally, we look at the dynamical competition between Kondo and RKKY ordering in a periodic Anderson model forming a one-dimensional ring penetrated by a magnetic flux. We construct a non-equilibrium Doniach-like phase diagram of the system and show how an optimally

controlled flux can drive the system between Kondo and RKKY phases with high efficiency and fidelity.
All our results are obtained with exact diagonalization, with the obvious but valuable byproduct of accurate theoretical benchmarks for more general NEGF approaches.

Dynamics of photo-doped charge transfer insulators

Denis Golež

Strong correlations between spin, charge and orbital degrees of freedom play an important role in materials and a recent development of ultrafast spectroscopies enabled to disentangle these relevant degrees of freedom by their temporal evolution. I will present recent development of theoretical tools based on dynamical mean field theory (DMFT) that enable us to describe the non-equilibrium dynamics in strongly correlated materials and show how the theoretical development is approaching a realistic description of solids, which is crucial to provide a proper feedback to the experimental community.

We study the dynamics of charge-transfer insulators after photo-excitation using the three-band Emery model and a non-equilibrium extension of GW +EDMFT. We identify two origins of the renormalization of the charge-transfer gap after photo-doping: the excitonic effect due to the Coulomb interaction between the d and p orbitals and the enhanced screening. The time-resolved photo-emission spectrum and the optical conductivity show clear signatures of the renormalized charge-transfer gap and p band position in agreement with experiments. Our formalism enables the realistic description of nonequilibrium phenomena in a large class of charge-transfer insulators, and provides a tool to explore the optical manipulation of interaction and correlation effects

Contour calculus for many-body correlators

Markku Hyrkäs

We generalize Langreth rules, used to convert contour integral equations into integral equations on the real axis, for equations involving many-body correlators and for general initial states out of equilibrium.

Calculating X-ray absorption spectra under non-equilibrium steady-state conditions

Alberto Marmodoro

Recent theoretical developments [1–4] have proposed a practical route to extend the Korringa, Kohn and Rostoker (KKR) / Green function method to non- equilibrium steady-state regimes. The underlying multi- ple scattering technique has historically been well estab- lished, particularly in connection to the study of spec- troscopy experiments, such as in the case of x-ray pho- toemission absorption (XAS) and magnetic dichroism (XMCD) [5]. In combination to general sum rules [6], this kind of measurements can cast insight on fine de- tails of magnetic properties of a material [7], including nano-engineered samples such as multi-layers [8] or finite clusters [9].

Beside the static, geometrically -governed effects ap- pearing in these scenarios, new experiments [10, 11] have started to probe how the driving of a steady-state cur- rent through application of a finite, externally controlled electric field can further modify electronic structure prop- erties of a material.

We report on current work within the Munich SPRKKR package [12] to allow the parameter -free, ab initio investigation of this kind of phenomenology in com- bination with density functional theory.

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High-harmonic generation in Mott insulators

Yuta Murakami

Strong light-matter couplings lead to intriguing non-perturbative phenomena such as high-harmonic generation (HHG). HHG was originally studied for gas systems and is used as laser sources. Recently, HHG was observed in semiconductors, and HHG in solid states are now attracting interests. In this poster, we discuss possibility and mechanism of HHG in Mott insulators [1]. We study the Hubbard model driven by the AC field using the dynamical mean-field theory implemented by the Floquet Green's function and a strong coupling impurity solver. We show that qualitative behavior of the HHG spectrum is different between weak and strong field regimes, which originates from qualitative difference in doublon/holon dynamics under the driving. We also discuss the similarity and the difference of the HHG intensity in the Mott insulator compared to that of semiconductors and disordered systems.

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Nonequilibrium DMFT study of superconductivity under parametric phonon driving

Yuta Murakami

The resonant excitation of mid-infrared phonon modes provides a new way of controlling material properties. An interesting result of such experiments is a light-induced superconducting (SC)-like behavior[1], for which several possible mechanisms have been theoretically proposed. However, in driven systems, the competition from heating and nonthermal energy distributions must be taken into account in order to assess the ultimate effect of such nonequilibrium states on SC. In the present work, we focus on the scenario that the parametric excitation of Raman-active phonons through nonlinear couplings to the mid-infrared phonons enhances the phonon-mediated attractive interaction and hence SC [2]. We systematically investigate the effects of the parametric phonon driving on SC in the Holstein model by analyzing the transient dynamics and the non-equilibrium steady states (NESS) using the non-equilibrium dynamical mean-field theory (DMFT) [3]. In the strong electron-phonon coupling regime, even though the attractive interaction can indeed be enhanced by the driving, the SC gap and the superfluid density are always suppressed in the NESS, in particular, at the parametric resonance. Even in the transient dynamics towards the NESS, the net effect is a suppression of SC. In the weakly-coupled regime, we study how the SC fluctuations evolve under the driving. Generically these correlations decay faster in the presence of driving and the decay becomes particularly fast around the parametric resonance. Our systematic analysis shows that, in a wide parameter range, the heating of the system is the dominant effect and that the parametric phonon driving has a negative effect on SC.

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Non-Markovian pure decoherence in a dielectric excited by a few-cycle laser pulse

Stanislav Yu Kruchinin

The analysis of recent experimental results on high harmonic generation in solids exposed to visible/near-infrared few-cycle laser pulses [1–3] has required the assumption of very short decoherence times ($T_2 \sim 2 - 4$ fs) to reach an agreement with the state-of-the-art quantum-mechanical models. In the Markov approximation, this assumption leads to a completely wrong dependence of the residual carrier population on the field amplitude due to unphysical spectral broadening and the opening of an artificial single-photon excitation channel. The similar problem has been previously noticed in the semiclassical description of electron dynamics with the Boltzmann equations [4–6].

Here, I present a theoretical approach to pure decoherence of charge carriers excited by a few-cycle laser pulse in dielectrics and semiconductors, which resolves this controversy allowing for both temporally fast decoherence and physically meaningful dependence of carrier excitation on the external field amplitude. Here, the equations of motion are generalized into the non-Markovian regime by replacing the constant dephasing rate with the time-dependent ones. This approach has been tested on two different baths: the Ornstein–Uhlenbeck model and the environment of harmonic oscillators (spin-boson model). It is shown that the excitation-induced dephasing with the environment of harmonic oscillators [7, 8] significantly improves the problem with opening of an unphysical single-photon excitation channel peculiar to the simulations with a constant (Markovian) decoherence rate and allows for temporally high dephasing during the main peak of the laser field, where the high

harmonics are primarily emitted, and the greater part of charge carriers is generated.

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Significance of vertex-corrections on the interface between non-equilibrium greens functions and time-dependent density-functional theory for cavity quantum electrodynamics

Christian Schäfer

With the consistent improvement of experimental and theoretical techniques for quantum electrodynamics on chemical scales, this novel connection presents in the mean-while a fascinating and realistic knob to modify chemical reactions using quantized transversal light interacting to matter. While the incorporation of the additional photonic contribution is straight forward in many-body perturbation theory, novel efficient

reformulations such as quantum electrodynamic density functional theory suffer from the typical lack of sufficient functionals. This settings, makes the investigation at the interface between them a pivotal domain of interest, communicating between accuracy and efficiency and unraveling the impact of quantum nature. Within this work, we present different levels of approximations of the time- dependent optimized effective potential (TDOEP) for the interaction between matter and light. We do so, by setting the TDOEP in context to simplifications along the line of the time-dependent Krieger-Li-Iafrate approximation (TDKLI), classical Maxwell descriptions but also extension along the line of a consistent GW approximation. By discussion of computational feasibility and intrinsic physical drawbacks, we tackle the question which route is promising, paving the way for further developments. By comparison to exact results, we address the significance of vertex-corrections, emerging for resonant light-matter interaction. We will observe, that a sufficiently accurate photon-propagator can indeed capture the correct physical behavior for all reasonable light-matter couplings, implying that the vertex is indeed not of essential importance.

Dynamical equilibration and thermalization of Hall response of topological systems

Michael Schüler

How topological properties such as the notion of protected edge states and the bulk and surface bulk response can be understood in a genuine nonequilibrium setup is a vivid field research. In particular, performing a quench across distinct topological phases, the topological invariant such as the Chern number stays constant. Nevertheless, several properties provide a signature of a dynamical topological phase transitions, such as circular dichroism in photoabsorption [1] which is even observed in the transient

regime [2] or the dynamical build up of a Hall response [3]. Although – strictly speaking – the topological character on a many-body level is conserved in such a scenario, the implications for the single-particle observables or the Hall response in an interacting system are nontrivial and relate to fundamental questions. In this work, we address exactly this point. Based on the paradigmatic two-band Bernevig-Hughes-Zhang (BHZ) model with electron-electron interactions, we investigate the generic post-quench dynamics of the single-particle density matrix and the nonequilibrium Hall response. To account for the pronounced nonequilibrium features in the highly-excited system, we combine a multi-scale treatment in reciprocal space with the generalized Kadanoff-Baym ansatz (GKBA) for an accurate description. Our findings show that the system acquires a finite Hall conductance as a clear signature of a topological phase transition.

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libCNTR: High-performance simulation toolbox for nonequilibrium strongly correlated systems

Michael Schüler

Nonequilibrium dynamics holds great promises of allowing for disentangling various degrees of freedom, thus providing microscopic insights in correlated systems. Simulating interacting electronic and/or bosonic systems in realistic scenarios remains a challenge. The nonequilibrium Green's function method in its time-dependent

formulation provides on of the most flexible approaches to include various types of interaction effects. The drawback of the method is its substantial numerical effort scaling at least cubically with the simulation time, as well as the inherent complexity of the underlying algorithms. The latter applies, in particular, to higher-order schemes required to study long-time behavior. We present the open-source software suite libCNTR for treating Green's functions on the Kadanoff-Baym (KB) contour. libCNTR provides a number of tools for constructing Feynman diagram and solving efficiently the corresponding equations of motion. The specialty of the software is the high-order implementation: dividing the KB contour into N points, the total integrated error of the equilibrium treatment and the two-time propagation scales with as $O(N^{-5})$. Efficient distributed-memory parallelization over reciprocal space allow large-scale calculations on extended systems. Furthermore, the software will be fully compatible with a follow-up software package for nonequilibrium dynamical mean-field theory in the strong-coupling limit.

Adiabatic Preparation of a Correlated Symmetry-Broken Initial State with the Generalized Kadanoff-Baym Ansatz

Riku Tuovinen

A fast time-propagation method of the nonequilibrium Green's function by using the generalized Kadanoff-Baym Ansatz (GKBA) is considered. We study a system with a symmetry-broken initial state, an excitonic insulator. The adiabatic preparation of this correlated initial state by the GKBA is assessed by comparing to a solution of the Dyson equation on the imaginary time-contour.