Schedule

	Sunday	Monday (5 Sept)	Tuesday (6 Sept)	Wednesday (7 Sept)	Thursday (8 Sept)
		Opening (8.50 am)			
9.00		Schollwöck	Krönke	Manthe	Programming tutorial @Institute of Physics
		Titvinidze	Schmelcher	Burghardt	
		Jeckelmann	Geißler	Mondelo-Martell	
		Coffee break	Coffee break	Coffee break	Coffee break
11.00		Steinigeweg	Scrinzi	Weber	Programming tutorial (cont'd)
		Mishchenko		Grossmann	
		Eissing	Fennel	Welsch	
		Alvermann	Bauke	Lee	
12.30		Lunch	Lunch	Lunch	Lunch
14.00		Breuer	Wimberger	Tanimura	Programming tutorial (cont'd)
		Mančal	Holthaus	Strunz	
		Intravaia	Leclerc	Kramer	
			Coffee break	Coffee break	Coffee break
16.00			Pehlke	Duan	Programming tutorial (cont'd)
		University Tour (16.30)	Ruggenthaler	Thorwart	
17.00	Registration		Joubert-Doriol	Perez-Leija	
			Ishibashi	Hübner	
				Closing remarks	
18.00		Poster session	Public evening lecture		
19.00					

Registration: Sunday, 17.00-19.00 in the Alfried-Krupp-Wissenschaftskolleg, or during the workshop
Welcome: Monday, from 20.00 (after Poster Session)

Public evening lecture: Tuesday, 18.00 in the Alfried-Krupp-Wissenschaftskolleg

Workshop start: Monday, 5 Sept, 8.50 Workshop end: Wednesday, 7 Sept, 18.00
Tutorial day: Thursday, 8 Sept, 9.00—17.30, Institute of Physics

Monday Sessions

Morning Sessions

08:50–09:00 **Welcome Address**
Holger Fehske, Christian Suhm

Chair: Holger Fehske

09:00–09:45 **Non-Equilibrium Physics in Strongly Correlated Systems with DMRG**
Ulrich Schollwöck

09:45–10:05 **Auxiliary master equation approach for correlated quantum impurities out of equilibrium**
Irakli Titvinidze

10:05–10:25 **Matrix-product-state method with local basis optimization for electron-phonon systems out of equilibrium**
Eric Jeckelmann

Coffee break ☕

Chair: Eric Jeckelmann

11:00–11:30 **Quantum typicality as numerical approach to real-time relaxation**
Robin Steinigeweg

11:30–11:50 **ARPES and optical conductivity of the Hubbard model with electron-boson coupling: equilibrium and pump-probe phenomena**
Andrey Mishchenko

11:50–12:10 **Renormalization in periodically driven quantum dots**
Katharina Eissing

12:10–12:30 **Quantum simulations with Faber-Chebyshev polynomials**
Andreas Alvermann

Lunch 🍴

Afternoon Sessions

Chair: Martin Holthaus

14:00–14:45 **Non-Markovian Quantum Dynamics of Open Systems**
Heinz-Peter Breuer

14:45–15:15 **Whither Master Equations?**
Tomáš Mančal

15:15–15:35 **Non-Markovianity in non-equilibrium atom-surface interactions**
Francesco Intravaia

Guided University Tour

16:30–17:45 **Tour through the University main and historic buildings**
We start at 16:35 at the Rubenow Statue in the Domstraße

Poster Session & Welcome

18:00–20:00 **Poster presentations**

20:00– **Welcome**
Food and Drinks in the Krupp-Kolleg

Tuesday Sessions

Morning Sessions

Chair: Uwe Manthe

09:00–09:45	**Solving the time-dependent many-body Schrödinger equation with Multi-Configuration Time-Dependent Hartree methods** *Sven Krönke*
09:45–10:15	**Applications of the Multi-Layer Multi-Configuration Time-Dependent Hartree Method for Bosons to the Nonequilibrium Quantum Dynamics of Ultracold Systems** *Peter Schmelcher*
10:15–10:35	**Tackling the multitude of bosonic quantum phases in Rydberg-dressed optical lattice systems** *Andreas Geißler*

Coffee break

Chair: Dieter Bauer

11:00–11:45	**Cutting the scale: the tSurff method for strong-field induced breakup** *Armin Scrinzi*
11:45–12:15	**Electron dynamics in temporally, spectrally, and spatially controlled intense fields** *Thomas Fennel*
12:15–12:35	**Numerical approaches to quantum field theory** *Heiko Bauke*

Lunch

Afternoon Sessions

Chair: Yoshitaka Tanimura

14:00–14:45	**Numerical simulation of lattice models for interacting bosons** *Sandro Wimberger*
14:45–15:15	**Floquet Condensates** *Martin Holthaus*
15:15–15:35	**Vibrational filtration using Zero-Width Resonances : an adiabatic Floquet approach** *Arnaud Leclerc*

Coffee break

Chair: Irene Burghardt

16:00–16:30	**TDDFT-Molecular Dynamics Simulation of Electronically Nonadiabatic Processes at Surfaces** *Eckhard Pehlke*
16:30–16:50	**Extensions of density-functional theories to QED: QED-Chemistry** *Michael Ruggenthaler*
16:50–17:10	**Geometric phase effects in multidimensional quantum dynamics** *Loïc Joubert-Doriol*
17:10–17:30	**Synchronization of mechanical oscillators in the quantum regime** *Kenta Ishibashi*

Public Evening Lecture

Chair: Holger Fehske

18:00-19:00	**Quantensimulation: quanten–analog versus klassisch–digital** *Ulrich Schollwöck*

Wednesday Sessions

Morning Sessions

Chair: Peter Schmelcher

09:00–09:45 **Polyatomic reaction dynamics and the multi-configurational time-dependent Hartree approach**
Uwe Manthe

09:45–10:15 **High-dimensional quantum dynamics of functional organic polymer materials: Coherence and correlations at the nanoscale**
Irene Burghardt

10:15–10:35 **Diabatization of the H_2@SWCNT system: time scale separation of confined and free coordinates**
Manel Mondelo-Martell

Coffee break

Chair: Sandro Wimberger

11:00–11:20 **Tuning of ICD-emitter properties in charged semiconductor double quantum dot systems**
Fabian Weber

11:20–11:50 **Semiclassical hybrid dynamics with molecular applications**
Frank Grossmann

11:50–12:10 **Non-equilibrium quantum correlation functions from Ring-Polymer Molecular Dynamics**
Ralph Welsch

12:10–12:30 **Quantum dynamics of a vibronically coupled linear chain using a surrogate Hamiltonian approach**
Myeong H. Lee

Lunch

Afternoon Sessions

Chair: Oliver Kühn

14:00–14:45 **Reduced Hierarchy Equations of Motion Approach to a Quantum Dissipative System**
Yoshitaka Tanimura

14:45–15:15 **Hierarchy of Stochastic Pure States for Open Quantum System Dynamics**
Walter Strunz

15:15–15:35 **Efficient energy transfer and computation in photosynthetic systems**
Tobias Kramer

Coffee break

Chair: Armin Scrinzi

16:00–16:20 **Signatures of primary charge separation in the reaction center of Photosystem II revealed by 2D electronic spectroscopy**
Hong-Guang Duan

16:20–16:50 **Quantum dynamical iterative path-integral simulations**
Michael Thorwart

16:50–17:10 **Noise-driven quantum interactions**
Armando Perez-Leija

17:10–17:30 **Theory of ultrafast spin dynamics and nano-spintronic logic functionalities on two-, three-, and four-center magnetic molecules**
Wolfgang Hübner

Closing & Farewell

17:30–17:45 **Closing remarks**
Dieter Bauer, Holger Fehske, Oliver Kühn

TALKS

Mon 09:00–09:45

Non-Equilibrium Physics in Strongly Correlated Systems with DMRG

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ABSTRACT

In this tutorium, I will give an introduction to the simulation of the time-evolution of strongly correlated quantum systems at $T = 0$ and $T > 0$ in one spatial dimension and illustrate it with examples from ultracold atomic systems. I will also discuss the calculation of spectral functions with time-dependent DMRG and illustrate this by examples from quantum magnetism. Time permitting, I will hint at the usefulness of DMRG as an impurity solver for time-dependent dynamical mean-field theory, which is applicable in the opposite limit of high dimensions.

Auxiliary master equation approach for correlated quantum impurities out of equilibrium

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ABSTRACT

The auxiliary master equation approach [1,2] allows for an accurate and efficient treatment of correlated impurities out of equilibrium. The method is based upon a mapping onto an auxiliary open quantum system in which the impurity is coupled to bath orbitals as well as to a Markovian environment. The intervening auxiliary orbitals allow for a treatment of non-Markovian dynamics at the impurity. The time dependence of this auxiliary system is controlled by a Lindblad master equation whose parameters are used to optimize the mapping. The auxiliary system exponentially approaches the original impurity problem upon increasing the number of parameters, i.e. of bath orbitals. Green's functions are evaluated via (non-hermitian) Lanczos exact diagonalisation [2] or by matrix-product states (MPS) [3]. In particular, our MPS implementation produces highly accurate spectral functions for nonequilibrium correlated impurity problems in the Kondo regime. Specifically, we can treat large values of the interaction and low temperatures T, well below the Kondo scale T_K. For $T = T_K/4$ and $T = T_K/10$ we find a splitting of the Kondo resonance into a two-peak structure at bias voltages just above T_K. The approach turns out to be an efficient impurity solver in equilibrium as well: a benchmark of our results for $T = T_K/4$ reveals a remarkably close agreement to numerical renormalization group. Applications to nonequilibrium dynamical mean-field-theory [4] as well as an implementation within Floquet theory for periodic driving [5] will be discussed as well.

References

[1] E. Arrigoni et al., Phys. Rev. Lett. 110, 086403 (2013)

[2] A. Dorda et al., Phys. Rev. B 89 165105 (2014)

[3] A. Dorda et al., PRB 92, 125145 (2015)

[4] I. Titvinidze et al., PRB 92, 245125 (2015)

[5] M. Sorantin et al., in preparation.

Mon 10:05–10:25

Matrix-product-state method with local basis optimization for electron-phonon systems out of equilibrium

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ABSTRACT

We present a method for simulating the time evolution of quasi-one-dimensional correlated systems with bosonic degrees of freedom using matrix product states [1]. Our goal is the accurate description of systems with large bosonic fluctuations for long periods of time. For this purpose we combine the time-evolving block decimation (TEBD) algorithm with a local basis optimization approach. We discuss the performance of this approach in comparison to TEBD with a bare boson basis, exact diagonalizations, and diagonalizations in a limited functional space. We show that this approach can reduce the computational cost by orders of magnitude when boson fluctuations are large and thus that it allows one to investigate problems that are out of reach of other approaches. First, we test our method on the non-equilibrium dynamics of a Holstein polaron [2] and show that it allows us to study the regime of strong electron-phonon coupling. Second, the method is applied to the scattering of an electronic Gaussian wave packet travelling through a small structure with electron-phonon coupling [1]. Our study reveals a rich physics including transient self-trapping and dissipation.

References

[1] C. Brockt, F. Dorfner, L. Vidmar, F. Heidrich-Meisner, and E. Jeckelmann, Phys. Rev. B 92, 241106(R) (2015)

[2] F. Dorfner, L. Vidmar, C. Brockt, E. Jeckelmann, F. Heidrich-Meisner, Phys. Rev. B 91, 104302 (2015)

Mon 11:00–11:30

Quantum typicality as numerical approach to real-time relaxation

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ABSTRACT

The concept of typicality states that a single pure state can have the same properties as the full statistical ensemble. This concept is not restricted to specific states and applies to the overwhelming majority of all possible states, drawn at random from a high-dimensional Hilbert space. In the cleanest realization, even a single eigenstate of the Hamiltonian may feature the properties of the full equilibrium density matrix, assumed in the well-known eigenstate thermalization hypothesis. The notion of property is manifold in this context and also refers to the expectation values of observables. Remarkably, typicality is not only a static concept and includes the dynamics of expectation values. Recently, it has become clear that typicality even provides the basis for powerful numerical approaches to the dynamics and thermalization of many-particle systems at nonzero temperatures. These approaches are in the center of my talk.

I demonstrate that typicality allows for significant progress in the study of real-time relaxation. To this end, I review numerical work [1-3] on current-current correlation functions of spin-1/2 systems. A comprehensive comparison with state-of-the-art methods unveils that typicality is satisfied in finite systems over a wide range of temperature and is fulfilled in both, integrable and non-integrable systems. Moreover, without the restriction to small systems or short times, typicality sheds light on several long-standing questions. As important examples, I discuss the relaxation process in integrable chains [1], non-integrable ladders [2], and many-body localized phases [3].

References

[1] R. Steinigeweg, J. Gemmer, and W. Brenig, PRL 112, 120601 (2014).

[2] R. Steinigeweg, J. Herbrych, X. Zotos, and W. Brenig, PRL 116, 017202 (2016).

[3] R. Steinigeweg, J. Herbrych, F. Pollmann, and W. Brenig, arXiv:1512.08519 (2016).

Mon 11:30–11:50

ARPES and optical conductivity of the Hubbard model with electron-boson coupling: equilibrium and pump-probe phenomena

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ABSTRACT

Angle resolved photoemission spectra (ARPES) [1,2] and optical conductivity (OC) [3,4] of the 2D Hubbard model with strong coupling to bosonic excitations are in good agreement with experiments on underdoped cuprates. Comparison with experiments shows that the effective strength of the electron-boson interaction is suppressed with doping. Analysis of the doping dependence of manifestations of the electron-boson interaction with novel many-polaron diagrammatic Monte Carlo method [5] shows that it is occurs because of suppression of the vertex corrections according to Migdal's theorem. General analysis shows that the energy scales of electron-electron and electron-boson interactions are very similar and it is difficult to prove or disprove its relevance in the spectroscopic experiments with the systems in equilibrium. Hence, a pump-probe technique is an exclusive route to disentangle different interactions and evaluate its importance. Time dynamics of the physical properties and OC of the 2D Hubbard-Holstein model is studied theoretically and experimentally when undoped system is put out of equilibrium by an ultrashort powerful light pulse [6,7]. It is shown that the ultra-fast dynamics of the underdoped cuprates can be explained only in case when strong electron-boson coupling is assumed.

References

[1] A.S. Mishchenko and N. Nagaosa, Phys. Rev. Lett. 93, 036402 (2004).

[2] A.S. Mishchenko et al, EPL 95, 57007 (2011).

[3] A.S. Mishchenko et al, Phys. Rev. Lett. 100, 166401 (2008).

[4] G. De Filippis et al, Phys. Rev. B 80, 195104 (2009).

[5] A. S. Mishchenko, N. Nagaosa and N. Prokof'ev, Phys. Rev. Lett. 113, 166402 (2014).

[6] G. De Filippis et al, Phys. Rev. Lett. 109, 176402 (2012).

[7] F. Novelli et al, Nature Comm. 5, 5112 (2014).

Mon 11:50–12:10

Renormalization in periodically driven quantum dots

KATHARINA EISSING

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ABSTRACT

A newly developed flexible renormalization-group-based approach for periodic driving is applied to the interacting resonant level model. We aim at the steady state of setups with one or more of the dot or lead parameters varied periodically in time, which is reached after all transients have died out. The interacting resonant level model is characterized by power-law scaling of observables in the relevant energy scales with interaction dependent exponents in equilibrium and if driven by a time constant bias voltage. The functional renormalization group has proven to be a versatile tool to investigate correlated, low-dimensional systems in and out of equilibrium e.g. by capturing these power laws correctly. Hence, we take explicitly advantage of the periodicity by applying the Floquet theorem and setting up the RG procedure in the Floquet basis. This allows to study the role of the driving frequency Ω as an infrared cutoff of the underlying renormalization group flow. I will discuss how the correlation between lead and dot electrons enhance or suppress the amplitude of the driving depending on the sign of the interaction. It is shown analytically that the magnitude of this effect follows a power law in the driving frequency which in turn manifests itself in the pumping power of the resulting non-Markovian, single parameter quantum pump.

References

Katharina Eissing, Volker Meden, Dante Kennes, PRL 116, 026801 (2016)

Mon 12:10–12:30

Quantum simulations with Faber-Chebyshev polynomials

ANDREAS ALVERMANN

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ABSTRACT

Polynomials and polynomial approximations are an essential part of general numerical and specific quantum physics algorithms. The talk addresses two complementary aspects of the role of polynomials in quantum simulations.

First, how static and dynamic properties of quantum systems are computed with eigensolvers, spectral function estimators, and time-propagation techniques based on Chebyshev and Faber polynomials.

Second, how we use polynomials to construct the matrix representation of Hamilton operators for systems with continuous degrees of freedom. With open quantum systems as the motivating example, we will show how polynomial techniques allow for the computation of phase transitions and non-equilibrium dynamics in the presence of strong system-environment correlations.

Mon 14:00–14:45

Non-Markovian Quantum Dynamics of Open Systems

HEINZ-PETER BREUER

Theoretical Condensed Matter Physics and Quantum Statistics
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ABSTRACT

The dynamics of open quantum systems plays a key role in many applications of quantum mechanics, examples ranging from fundamental problems, such as the environment-induced decay of quantum coherence and relaxation in many-body systems, to applications in condensed matter theory, quantum transport, quantum chemistry and quantum information. In close analogy to a classical Markovian stochastic process, the interaction of an open quantum system with a noisy environment is often modelled phenomenologically by means of a dynamical semigroup with a corresponding time-independent generator in Lindblad form, which describes a memoryless dynamics of the open system typically leading to an irreversible loss of characteristic quantum features. However, in many applications open systems exhibit pronounced memory effects and a revival of genuine quantum properties such as quantum coherence, correlations and entanglement. We will discuss recent theoretical results on the rich non-Markovian quantum dynamics of open systems, paying particular attention to the rigorous mathematical definition, to the physical interpretation and classification, as well as to the quantification of quantum memory effects.

Mon 14:45–15:15

Whither Master Equations?

TOMÁŠ MANČAL

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ABSTRACT

In recent years, we have seen a steady progress in exact simulation techniques for propagation of reduced density matrix of increasingly complex systems. Approximate techniques such as master equations are now often seen as less trustworthy, and prone to losing physicality. In this contribution, we will look in detail on the nature of approximations involved in the standard master equation approaches, such as Redfield and Modified Redfield equations, Förster energy transfer rates etc. Our discussion will go beyond the usual statements about the weak and strong interaction limits, and we will propose an alternative perspective on the reasons for failures of these master equations in certain parameter regimes. We will show several cases where exact results can be obtained by (seemingly) second order perturbation theory. Insights from these examples and from our detailed analysis of master equation failures will be used to answer the questions whether there is a reasonable hope for exact master equations for practical cases in energy transfer and spectroscopy. We will shortly introduce a new computational tool we are developing, which allows an integration and straightforward comparison of different theoretical approaches to molecular energy transfer problems and spectroscopy.

Mon 15:15–15:35

Non-Markovianity in non-equilibrium atom-surface interactions

FRANCESCO INTRAVAIA

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ABSTRACT

We discuss the failure of the Markov approximation in the description of quantum friction. Like ordinary friction, this effect describes a force acting on an object moving near another one. Unlike the classical case, however, quantum friction is mediated by the interaction with the electromagnetic field at zero temperature. Using general theoretical arguments, we show that the Markov approximation can lead to erroneous predictions on such a phenomenon with regard to both strength and functional dependencies on system parameters. In particular, we show that the long-time power-law tails of temporal correlations, and the corresponding low-frequency behavior, of two-time dipole correlations, neglected in the Markovian limit, affect the prediction of the force. Our findings highlight the importance of non-Markovian effects in dispersion interactions.

References

F. Intravaia, R. O. Behunin, C. Henkel, K. Busch, and D. A. R. Dalvit, eprint: arXiv:1603.05165

Tue 09:00–09:45

Solving the time-dependent many-body Schrödinger equation with Multi-Configuration Time-Dependent Hartree methods

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ABSTRACT

Simulating correlated quantum dynamics of many degrees of freedom is a challenge since many eigenstates typically contribute to the system's evolution, since correlations between the degrees of freedom may dynamically emerge and decay and since the complexity of the problem in general scales exponentially with the number of degrees of freedom. This lecture-like tutorial is devoted to a certain class of wavefunction propagation methods, the Multi-Configuration Time-Dependent Hartree (MCTDH) family, which faces these problems by expanding the system's state w.r.t. a time-dependent, dynamically optimized many-body basis. The idea of a co-moving basis can be applied to various situations, covering e.g. nuclei dynamics in molecules, system-bath problems and the many-body dynamics of bosonic or fermionic ultra-cold atoms and mixtures thereof.

First, the general idea of this class of methods and the derivation of the corresponding equations of motion is explained by an example. Thereafter, an in-situ simulation will illustrate how one typically prepares the initial state and judges the convergence of the subsequent propagation. Finally, we will review several methods of the MCTDH family, which are tailored to various classes of systems, and focus on the Multi-Layer MCTDH method for bosons, which allows for simulating the correlated quantum dynamics of ultra-cold bosonic mixtures.

Tue 09:45–10:15

Applications of the Multi-Layer Multi-Configuration Time-Dependent Hartree Method for Bosons to the Nonequilibrium Quantum Dynamics of Ultracold Systems

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ABSTRACT

The multi-layer multi-configuration time-dependent Hartree method for bosons represents a powerful ab initio method for the investigation of the non-equilibrium quantum dynamics of single and multi-species bosonic systems in traps and optical lattices. After briefly introducing the method we discuss a number of different applications ranging from the correlated quantum dynamics in optical lattices to beyond mean-field behaviour of solitons and collisionally coupled correlated species. Firstly we demonstrate in a ‘bottom-up approach’ the correlated many-particle effects in the collective breathing dynamics for few- to many-boson systems in a harmonic trap. Many-body processes in black and grey matter-wave solitons are explored thereby demonstrating that quantum fluctuations limit the lifetime of the soliton contrast, which increases with increasing soliton velocity. For atomic ensembles in optical lattices we explore the interaction quench induced multimode dynamics leading to the emergence of density-wave tunneling, breathing and cradle-like processes. An avoided-crossing in the respective frequency spectrum provides to a beating dynamics for selective modes. A particular far from equilibrium system is then studied at hand of the correlated quantum dynamics of a single atom collisionally coupled to a finite bosonic reservoir. In the last part of the presentation we provide some selective aspects of our recent investigations on atom-ion hybrid systems using the same methodology. First the ground state properties of ultracold trapped bosons with an immersed ionic impurity are discussed. Subsequently the capture dynamics of ultracold atoms in the presence of the impurity ion is explored.

Tackling the multitude of bosonic quantum phases in Rydberg-dressed optical lattice systems

ANDREAS GEISSLER[1], MATHIEU BARBIER, IVANA VASIC, YONGQIANG LI, AND WALTER HOFSTETTER

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ABSTRACT

Our recent results [1] have shown the rich diversity of quantum phases which are induced by the strong correlations inherent to Rydberg-dressed bosonic atoms trapped in optical lattices. While experimental feasibility of the dressing itself has recently been demonstrated for the first time for a small number of particles [2] as well as in a many-body system [3], a better understanding of the crystallisation dynamics induced by Rydberg-dressing is still required. We have analysed Rydberg-dressed lattice systems for various lattice geometries by applying the real-space extension of bosonic dynamical mean-field theory (RB-DMFT). These results serve as a benchmark for Gutzwiller-type mean-field calculations, where dissipative dynamics can be simulated within the Lindblad master equation approach. We take into account both the spontaneous decay of Rydberg states and effective dephasing due to black-body radiation. We can thus observe dissipative crystallisation dynamics and the stability of crystalline and supersolid order. We additionally investigate quasiparticle excitations of the many-body system, which we determine from linearised Gutzwiller equations.

References

[1] A. Geißler et al., arXiv:1509.06292.

[2] Y.-Y. Jau et al., Nat. Phys., **12**, 71-74 (2016).

[3] J. Zeiher et al., arXiv:1602.06313.

Tue 11:00–11:45

Cutting the scale: the tSurff method for strong-field induced breakup

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ABSTRACT

Length- and time-scales of typical laser pulses widely exceed the corresponding scales of atoms, molecules, or a few layers of a solid surface. As a result, any brute-force numerical solution of the corresponding time-dependent Schrödinger equation quickly reaches the limits of computer resources. The time-dependent surface flux (tSurff) method allows to disentangle the spatial scales and to reconstruct breakup spectra from atomic scale simulations. In my presentation I will introduce the concept and discuss the main technical ingredients. The power of the method will be demonstrated on several challenging problems of strong field physics, such as ionization in arbitrary polarization fields, molecular ionization in presence of multi-electron dynamics, and molecular break up.

Tue 11:45–12:15

Electron dynamics in temporally, spectrally, and spatially controlled intense fields

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ABSTRACT

Current laser technology allows the generation of well-controlled ultrashort and intense light fields in a broad spectral range from the infrared to the x-ray domain. Such fields enable to drive, manipulate, and probe attosecond electron dynamics in atoms, molecules, and nanostructures with unprecedented resolution. Understanding the physics starts from solving the full time-dependent Schrödinger equation which is feasible for one or maybe few particles. However, quasi-classical or mean-field approaches are typically indispensable to include (i) collective effects and (ii) electromagnetic wave propagation or (iii) to reveal a clear physical picture of the predominant mechanism. In this talk I will discuss typical features of strong-field ionization and their specific signatures due to quantum and classical dynamical behavior. These will be illustrated based on results for atomic two- color ionization from mixed NIR-XUV fields [1], the carrier-envelope phase dependent electron emission from dielectric nanostructures [2,3], and the atomic ionization dynamics in phase-controlled bichromatic laser fields [4], leading to the so-called phase-of-the-phase

References

[1] P. Radcliffe, M. Arbeiter, W. B. Li, S. Düsterer, H. Redlin, P. Hayden, P. Hough, V. Richardson, J. T. Costello, T. Fennel, M. Meyer, “Atomic photoionization in combined intense XUV free-electron infrared laser fields”, New J. Phys. 14, 043008 (2012)

[2] S. Zherebtsov, T. Fennel, J. Plenge, E. Antonsson, I. Znakovskaya, A. Wirth, O. Herrwerth, F. Süßmann, C. Peltz, I. Ahmad, S. Trushin, V. Pervak, S. Karsch, M.J.J. Vrakking, B. Langer, C. Graf, M.I. Stockman, F. Krausz, E. Rühl, M.F. Kling, “Controlled near-field enhanced electron acceleration from dielectric nanospheres with intense few-cycle laser fields”, Nat. Phys. 7, 656 (2011)

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[4] S. Skruszewicz, J. Tiggesbäumker, K.-H. Meiwes-Broer, M. Arbeiter, T. Fennel, D. Bauer, „Two-Color Strong-Field Photoelectron Spectroscopy and the Phase of the Phase“, Phys. Rev. Lett. 115, 043001 (2015)

Tue 12:15–12:35

Numerical approaches to quantum field theory

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ABSTRACT

Relativistic quantum dynamics of strong field processes becomes accessible via emerging strong-field laser facilities. This regime requires a description via the Dirac equation and quantum field theory. Analytical solutions of the time-dependent Dirac equation or even of the full quantum field theory are not feasible for most systems of interest. Analytical approximations or numerical approaches have to be applied.

In a very strong electromagnetic environment, a possible back-reaction of the electron dynamics to the electromagnetic environment is negligible. In this case, the electron dynamics is described by the quantum field theory of the Dirac electron in an external field. The electronic quantum field state can be determined by solving the time-dependent Dirac equation [1,2] for all basis vectors of the corresponding Hilbert space [3]. The proposed quantum field theoretical treatment the full electronic quantum field state that results after the action of a strong electromagnetic field.

The proposed method is applied to study nonperturbative multiphoton electron-positron pair creation in ultrastrong electromagnetic fields with elliptic polarization [4]. Our numerical approach allows to take into account the time-spatial variations of the standing electromagnetic field. The calculated quantum field state contains all information about the spectrum of created pairs. Thus, the method provides the probabilities of detecting specific multi-pair states (characterized by momentum and spin) and it shows how these probabilities evolve as a function of the time of interaction with the electromagnetic environment.

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Tue 14:00–14:45

Numerical simulation of lattice models for interacting bosons

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ABSTRACT

After giving a motivation in which contexts lattice models for neutral ultracold atoms are relevant for experiments in the laboratory, we focus on exact ab-initio techniques for the numerical treatment of such models for bosons. Our basic ingredient is Floquet-Bloch theory exploiting the temporal and spatial periodicity of the system. We also present a direct integration method and check how it compares with the spectral method of exact diagonalization. Applications of exact techniques are presented, e.g. for a multi-band Wannier-Stark problem. If time allows we discuss as well approximate numerical techniques beyond mean-field and their respective applications such as in open chains of quantum dots for bosons. Our presentation is based on the following two reviews: Comp. Phys. Comm. 186, pp. 19-30 (2015) and Eur. Phys. J ST 224, pp. 2127-2171 (2015).

Tue 14:45–15:15

Floquet Condensates

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ABSTRACT

Although the concept of Bose-Einstein condensation (BEC) usually is applied to states of (or at least close to) thermal equilibrium, the fundamental Penrose-Onsager criterion for BEC of interacting particles[a] does not require that the system under consideration necessarily is in thermal equilibrium, nor even in a steady state. Might there be interesting physics of nonequilibrium condensates?

This question is particularly pertinent in situations where an interacting Bose gas is driven by some (possibly strong) time-periodic external force, such as ultracold atoms in time-periodically modulated traps: In such cases the Floquet theorem suggests the existence of a particular basis of time-periodic states which carry *constant* occupations, despite the driving force. Could such a Floquet state be macroscopically occupied, *i.e.*, are there "Floquet condensates"?

In this contribution these questions will be taken up in a hands-on manner. After reviewing some landmark experiments which clearly show that a macroscopic matter wave can be strongly driven without losing its coherence, I will introduce a (deceptively) simple model system which can be treated numerically without too much effort. This model indicates that the physics of Floquet condensates may be surprisingly rich, providing ample opportunity to boldly go where no man has gone before.

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Tue 15:15–15:35

Vibrational filtration using Zero-Width Resonances : an adiabatic Floquet approach

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ABSTRACT

We present an adiabatic strategy for the quantum control of diatomic molecules, subject to photodissociation under the influence of a short laser pulse. In the context of molecular cooling, our control objective is the filtration of one given vibrational state within a vibrational wavepacket. The general idea is to play with the contrast between different lifetimes of Floquet eigenstates issuing from different field-free vibrational states, in order to isolate one given state in the wavepacket, all the other states decaying as fast as possible. Motivated by a semi-classical analysis, we have looked for zero-width resonance (ZWR) paths, corresponding to quasi-infinite lifetimes, leading to a strong photodissociation quenching. We have explored the morphology of Floquet resonance eigenvalues in the laser parameter plane (wavelength and intensity) to design optimal laser pulses, along which a single state can be adiabatically transported without decay. Since the Floquet eigenstates imply both bound and continuum states, the practical realization of an adiabatic approximation is not straightforward and should be tested to avoid uncertain conclusions: The final step consists in a full wave-packet propagation taking into account all the possible non-adiabatic effects which could jeopardize the filtration process. Using this method leads to 95% of survival population in the selected state while neighbouring states are decaying fast [1]. Obtaining such results requires many repetitive partial diagonalizations of non-hermitian Hamiltonians. We have used the wave-operator formalism which is well-suited for such calculations. The full time-dependent calculations have been performed using a recently developed algorithm [2] based on the time-dependent wave-operator formalism and using Fourier basis sets over time.

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Tue 16:00–16:30

TDDFT-Molecular Dynamics Simulation of Electronically Nonadiabatic Processes at Surfaces

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ABSTRACT

During the scattering, or the sticking reaction, of atoms and molecules on metal surfaces electronically nonadiabatic processes can occur. Such electronically nonadiabatic effects have been observed directly in experiment [1-3]. The objective of the work presented in this talk is to simulate energy transfer processes at surfaces ab initio. To this purpose, the simultaneous integration of the time-dependent Kohn-Sham equations from TDDFT for the electronic degrees of freedom, and the Ehrenfest equations for the nuclei, has been implemented in the fhi96md density functional total energy program [4] by the authors of [5]. An overview over electronic energy dissipation mechanisms active in case of H-atoms interacting with an Al(111)-surface as investigated by M. Lindenblatt et al. [6,7], and the vibrational – electron hole pair coupling of highly vibrationally excited HCl molecules in front of an Al(111) surface [8] will be given. For HCl/Al(111) the electronic energy transfer can be reproduced within a simple tight-binding model. This finding may pave the way for future inclusion of electronic nonadiabatic effects in standard Born-Oppenheimer molecular dynamics simulations.

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Tue 16:30–16:50

Extensions of density-functional theories to QED: QED-Chemistry

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ABSTRACT

In this talk we will review the recent advances within ground-state and time-dependent density-functional theory to include the interaction with photons from first principles. This so-called quantum-electrodynamical density-functional theory (QEDFT) treats light and matter on equal quantized footing and bridges quantum chemistry and quantum optics. QEDFT provides a numerically feasible scheme to investigate the influence of the quantum nature of light on the properties of complex systems, and how we can manipulate such systems with the help of photons. We highlight possible applications and show for a prototype system of photons interacting with electrons how the effective potential of the corresponding Maxwell-Kohn-Sham scheme describes the local forces induced by the photons exactly. We compare these exact results with an optimized-effective-potential approach for the coupled matter-photon system and present a coupled Maxwell-Kohn-Sham calculation for realistic systems.

Tue 16:50–17:10

Geometric phase effects in multidimensional quantum dynamics

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ABSTRACT

In the adiabatic representation, nonadiabatic couplings between electrons and nuclei induce radiationless electronic transitions in photo-induced processes. Conical intersections (CIs) are nuclear manifolds for which these couplings become infinite. Therefore, CIs play an important role in radiationless processes. A specific topology of the electronic wavefunction at a CI causes appearance of the geometric phase (GP) [1], which induces an interference pattern in the nuclear wavepacket. Ignoring this interference can significantly change the dynamics.

Therefore, the presence of the GP must be accounted for in molecular dynamics simulations. To understand GP effects, we simulate molecular dynamics of model Hamiltonians with and without GP. Resulting dynamics show that removing the GP for an internal conversion process at a CI reduces the electronic transition probability [2]. Furthermore, we observed that the GP also affects low energy dynamics between two wells by blocking the population transfer [3]. We investigate this latter effect on an electron transfer in the photolyase protein (PL) involved in a DNA repair process. We developed a technique to explore GP effects in the diabatic representation and show that in the PL, the GP slows down the electron transfer by creating destructive interference.

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Tue 17:10–17:30

Synchronization of mechanical oscillators in the quantum regime

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ABSTRACT

We study the synchronization of coupled mechanical oscillators in the quantum regime. While synchronization phenomena are ubiquitous in nature and have been well studied in the classical theory, its quantum theory has just started to draw attention due to the recent development of optomechanical control techniques of nonlinear mechanical oscillators in the low-excitation regime. Regardless of the recent pioneering studies, quantum synchronization has not been completely comprehended. For example, there is no common understanding of the connection between entanglement and synchronization.

In this presentation, we present the characterization of the phase dynamics of two oscillators by using the mutual information, reduced Wigner function, and power spectrum. The phase entrainment is found to survive in the quantum regime, and its phase diagram agrees with that of the classical theory, provided that the phase boundaries are blurred due to quantum fluctuations. We also investigate the amplitude dynamics, and propose a method to control the individual oscillator within the multiple coupled oscillators. As a peculiar feature in quantum systems, we discuss the entanglement between two mechanical oscillators associated with the phase or amplitude synchronization.

Wed 09:00–09:45

Polyatomic reaction dynamics and the multi-configurational time-dependent Hartree approach

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ABSTRACT

The accurate description of the quantum dynamics of polyatomic systems is a challenging subject. The present talk focuses on accurate numerical methods for high-dimensional quantum dynamics simulations, theoretical concepts for the simulation and analysis of polyatomic reaction processes, and discusses prototypical applications.

The scaling of the numerical effort with dimensionality is a key problem of quantum dynamics calculations studying polyatomic systems. The multi-configurational time-dependent Hartree (MCTDH) approach and it multi-layer extension have been established as efficient schemes for the accurate treatment of high-dimensional systems. In the present talk, the central ideas of the approach are introduced and discussed.

The quantum transition state approach provides an intuitive framework for the accurate description of reactions proceeding via a potential barrier. It is typically used to study reaction rates. In recent work, the theory has extended to the calculation of fully state-resolved reaction probabilities and cross-sections. In this talk, the theoretical concept is introduced and accurate results obtained for the $H + CH_4 \rightarrow H_2 + CH_3$ reaction are presented. The ro-vibrational control of the chemical reactivity is discussed and the correlation between the quantum states of the reactants and products is analyzed.

High-dimensional quantum dynamics of functional organic polymer materials: Coherence and correlations at the nanoscale

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ABSTRACT

We present high-dimensional quantum dynamical studies of ultrafast photoinduced exciton migration and dissociation in functional organic materials, in view of understanding the intricate interplay of electronic delocalization, coherent nonadiabatic dynamics, and trapping phenomena. Our approach combines first-principles parametrized lattice Hamiltonians, based on TDDFT and/or high-level electronic structure calculations, with accurate quantum dynamics simulations using the Multi-Configuration Time-Dependent Hartree (MCTDH) method [1] and its multi-layer and Gaussian-based [2] variants, as well as non-Markovian reduced dynamics techniques [3]. In addition, hierarchical effective mode representations [3,4] are employed to obtain reduced-dimensional models. This talk will specifically address (i) exciton dissociation and free carrier generation in donor-acceptor materials [5,6], (ii) exciton multiplication in acene materials [7], and (iii) the elementary mechanism of exciton migration as well as charge-transfer exciton formation in polythiophene type materials [8]. A perspective is given on the role of temporal and spatial coherence, along with a consistent description of the transition to a classical statistical regime.

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Diabatization of the H_2@SWCNT system: time scale separation of confined and free coordinates

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ABSTRACT

The MCTDH approach has been used to successfully simulate the quantum dynamics of a variety of molecular systems with dozens [1,2] or even more [3] degrees of freedom. Recently, we have used this method to study the quantum confinement effects of H_2 and D_2 confined in single-walled carbon nanotubes using a general Potential Energy Surface and a 6D Hamiltonian [4]. In spite of the advantages of MCTDH, the calculation of the potential energy matrix elements remain as the bottleneck of the computation, as well as the main source of numerical error and instability.

In the present work, we find a workaround to the potential energy matrix problem found in this system by applying a diabatization strategy. Full dimensional (6D) calculations on this system reveal a low coupling between the translational degree of freedom of H_2 along the nanotube axis and the remaining five confined coordinates. The diabatization of the confined degrees of freedom would thus allow to reduce the dimensionality of the system from six to two, and furthermore circumvent the need to adapt the potential energy surface in the 6D calculations. The approach is tested by comparing diffusion rate constants of H_2 along the nanotube computed with both the full dimensional and diabatized models. It is seen that both the computational cost and the numerical stability of the calculation are greatly improved.

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Wed 11:00–11:20

Tuning of ICD-emitter properties in charged semiconductor double quantum dot systems

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ABSTRACT

In previous studies on electron dynamics [1], the interatomic Coulombic decay (ICD) process was predicted to take place in charged double quantum dot (DQD) systems. ICD represents an ultrafast energy transfer process [2], that is triggered by photoexcitation of a conduction band (CB) electron in a photon-absorbing QD. Deexcitation leads to the release of a CB electron in a neighbouring electron-emitting QD. As ICD depends on the electronic properties [3], which in turn are defined by the QD geometry and material [4], the process can be optimized to yield high decay rates. So far, the optimization of the system focused on the photon-absorber QD [5] while keeping the properties of the electron-emitter QD fixed. The present work focuses on maximizing the ICD rate in a DQD model system by optimization of the emitter QD through electron dynamics with the multiconfiguration time-dependent Hartree method [6]. Additionally, the picture of a virtual photon transfer [7] is used to investigate the dependency on the photoionization cross section of the emitted electron during the process. Using both the optimization of absorber and emitter, we conclude that a nanowire of longitudinally arranged cigar-shaped QDs is suitable for experimental studies. From the virtual photon transfer process study we point out, that the emitter QD geometry may be optimized idependently from the absorber QD for a given absorber geometry.

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Wed 11:20–11:50

Semiclassical hybrid dynamics with molecular applications

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ABSTRACT

The semiclassical initial value formalism to solve the time-dependent Schroedinger equation will be reviewed. Special focus will be laid on the multi-trajectory Herman-Kluk method [1] and Heller's thawed Gaussians [2]. The connection of the two methods by a Gaussian integration will be stressed. This conncetion lays the foundation for the semiclassical hybrid formalism that combines both approaches [3].

I then present results for the quenching of quantum interference in Iodine molecules in solution, described by a Caldeira-Leggett Hamiltonian [4]. Finally, I discuss the combination of the hybrid approach with time-averaging methods [5] to quickly calculate vibrational spectra.

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Wed 11:50–12:10

Non-equilibrium quantum correlation functions from Ring-Polymer Molecular Dynamics

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ABSTRACT

During the last decade the Ring-Polymer Molecular Dynamics (RPMD) approach has proven to be an efficient method to approximately simulate quantum mechanical effects in chemical systems. Based on the path integral formulation, RPMD incorporates quantum effects using classical trajectories in an extended ring-polymer phase space. It exhibits appealing features including preservation of detailed balance, being exact in several limiting cases and it can be run efficiently exploiting techniques known from molecular dynamics. However, the current formalism has been limited to the calculation of correlation functions associated with the equilibrium Boltzmann distribution. In this poster the extension of the RPMD approach to simulate processes with non-equilibrium initial conditions is discussed. Furthermore, the prospects of the approach to study photoexcited processes like excited-state proton transfer is investigated.

Wed 12:10–12:30

Quantum dynamics of a vibronically coupled linear chain using a surrogate Hamiltonian approach

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ABSTRACT

Vibronic coupling between the electronic and vibrational degrees of freedom has been reported to play an important role in charge and exciton transport in organic photovoltaic materials, molecular aggregates and light-harvesting complexes. Explicitly accounting for effective vibrational modes rather than treating them as a thermal environment has been shown to be crucial to describe the effect of vibronic coupling. We present a methodology to study dissipative quantum dynamics of vibronically coupled systems based on a surrogate Hamiltonian approach, which is in principle not limited by Markov approximation or weak system-bath interaction, using a vibronic basis. We apply vibronic surrogate Hamiltonian method to a linear chain system and discuss how different types of relaxation process influence population dynamics of dissipative vibronic systems.

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Wed 14:00–14:45

Reduced Hierarchy Equations of Motion Approach to a Quantum Dissipative System

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ABSTRACT

In this course of lectures, I will teach the essence of quantum dissipative dynamics using the PowerPoint lecture note available at the above WWW site. I will first introduce a system-bath model to explain how fluctuation and dissipation arises from the environment, and the concept will be illustrated with some examples from chemistry and biology. Then the quantum Fokker-Planck equations will be derived from this in hierarchy form based on the Feynman-Vernon influence functional formalism. The derived hierarchy equations can deal with strong system-bath coupling and non-Markovian noise. The reduced hierarchy equations of motion for system with discrete energy levels will also be derived using a coherent state representation for the path integrals. Various applications of the hierarchy equation of motion approach, including linear and nonlinear spectroscopies, electron transfer, photo dissociation, chemical reactions, quantum tunneling, and quantum information problems will also be discussed. The distributed source code, "nonMarkovian2009" and "tanimuranFP15", will be demonstrated and examined.

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Wed 14:45–15:15

Hierarchy of Stochastic Pure States for Open Quantum System Dynamics

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ABSTRACT

A hierarchy of stochastic evolution equations for pure states (quantum trajectories) for open quantum system dynamics with non-Markovian structured environments based on non-Markovian quantum state diffusion (NMQSD) is presented. This hierarchy of pure states (HOPS) provides the exact reduced density operator as an ensemble average over normalized states. We stress the meaning of the stochastic approach in terms of the unitary dynamics of system plus environment and present applications.

Wed 15:15–15:35

Efficient energy transfer and computation in photosynthetic systems

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ABSTRACT

The accurate modeling of time-resolved spectra of larger photosynthetic complexes provides important insights into the choreography of the energy transport in molecular networks. We show results from the first many-node, many-core HEOM implementation enabling us to identify the physical mechanisms at work.The scalable algorithms are developed in collaboration with the Intel Center for parallel computing at ZIB and exploit parallel features of upcoming processors and GPUs and run from small hand-held devices up to top 100 supercomputers. A ready-to-run cloud version is available at nanoHUB.org

Wed 16:00–16:20

Signatures of primary charge separation in the reaction center of Photosystem II revealed by 2D electronic spectroscopy

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ABSTRACT

PSII RC is a unique biological system that converts sunlight energy into chemical energy by water splitting. The structure of the PSII RC [1] shows two symmetric branches containing 8 pigments each. It provides a very important model for the study of natural charge separation and charge transfer and has been extensively studied experimentally and theoretically. The charge separation processes in the PSII RC have been extensively investigated in the past by means of time-resolved spectroscopies; however, the 2D electronic spectroscopy was applied only recently (at 77 K [2] and at room temperature [3]). These studies were focused on the energy transfer processes and coherent oscillations arising in the series of 2D-spectra measured at different waiting times. We performed an extended 2D electronic spectroscopy study of the PSII RC at ambient temperature combined with theoretical modelling. This approach allowed identification of the primary charge separation process in the RC and the corresponding timescale, as well as resolving energy transfer pathways. An excellent agreement was achieved between the measured and simulated 2D spectra (Fig. 1) and 2DDAS without involving any long-lived oscillations in the model to enhance charge separation [3] in the PSII RC.

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Wed 16:20–16:50

Quantum dynamical iterative path-integral simulations

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ABSTRACT

In this talk, I present the technique of iterative real-time path integral simulations which can be applied to a large variety of quantum statistical problems in which the coupling to a bath or reservoir is important. After explaining the basic concept, I will show several applications where bosonic baths are involved. The concept can also be used for fermionic baths in quantum transport set-ups. Importantly enough, the technique is numerically exact, meaning that there is a systematic way to ensure numerical convergence. The applications range from the nonlinear 2D optical spectroscopy of biomolecular excitons in presence of a strong vibronic coupling to molecular quantum transport under strong nonequilibrium conditions.

Wed 16:50–17:10

Noise-driven quantum interactions

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ABSTRACT

In quantum physics, noise represents a prominent adversary since it precludes the generation, control, and preservation of fundamental properties such as entanglement and quantum correlations. In other words, the presence of noise destroys quantum coherence giving rise to classical effects through a process known as decoherence. Notably, under certain condi- tions, the existence of noise may drive quantum and classical systems to undergo intriguing nontrivial effects. In this regard, for instance, chemical reactions in condensed molecular systems take place via a process termed noise-activated barrier crossing. As such, this phenomenon—originally introduced in the context of Brownian particles escaping from metastable energy-states—is ubiquitous in many scientific areas, e.g. in chemical kinetics, electron fluxes in semiconductors, and excitonic transport in photosynthetic systems. In the quantum realm, noise-activated barrier crossing effects can be thought of as the stochastic enabling of initially inhibited quantum states. That is, the generation of noise-driven quantum interactions between entities, which are otherwise uncoupled. Along these lines, we demonstrate, both theoretically and experimentally, the existence of genuine noise-driven quantum interactions in dynamically disordered photonic networks. Using fourth-order field correlation measurements, we observe that on average the spatial wave function of indistinguishable photon pairs delocalizes populating sites that in the absence of noise were uncoupled. Even more interestingly, we demonstrate that the system always tends to the same steady state, irrespectively of the initial biphoton state. Remarkably, the measured correlation matrix corresponding to such steady state exhibits the higher probabilities along the main diagonal thereby demonstrating that the steady state of the photons corresponds to a generalized maximally entangled NOON-like state.

Wed 17:10–17:30

Theory of ultrafast spin dynamics and nano-spintronic logic functionalities on two-, three-, and four-center magnetic molecules

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ABSTRACT

We present an ab initio theory of ultrafast laser-induced magnetic switching and spin transfer dynamics in magnetic two-, three, and four-center molecules. The theory is based on coupled-cluster wavefunction methods and a genetic algorithm to target specific states by suitably tailored femtosecond pulses. Two magnetic centers: Local spin-flip and transfer are discussed in Fe- Co- and Ni-dimers, possibly with nonmagnetic bridges, or in fullerene cages. Ligand stabilized complexes are used for joint dynamics on two centers in different chemical environment and to combine highly correlated electron physics with recent experimental possibilities for synthesis and femtosecond spectroscopy of excited states [1,2]. Three magnetic centers: A minimal model molecule is constructed from three Ni atoms in different local symmetry and bridged by two Na atoms [3]. This molecule serves as a prototype for AND and OR functionalities based on laser induced local spin dynamics in a magnetic field as gating partner. A Co-trimer [4] is demonstrated as a cyclic spin-SHIFT register. Four-center magnetic molecules, e. g. Ni_4, may exhibit additional functionalities. Spin bifurcation and its reverse (spin association/ merging), as well as the which path interference are demonstrated. We construct two elements for nanologic applications: a 4-bit cyclic SHIFT register [5] and a pure-spin OR gate [6], without dynamically active external magnetic field.

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PROGRAMMING TUTORIAL Thu, 9:00–17:30

Node-Level Performance Engineering

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ABSTRACT

The advent of multi- and manycore chips has led to a further opening of the gap between peak and application performance for many scientific codes. This trend is accelerating as we move from petascale to exascale. Paradoxically, bad node-level performance helps to "efficiently" scale to massive parallelism, but at the price of increased overall time to solution. If the user cares about time to solution on any scale, optimal performance on the node level is often the key factor. We convey the architectural features of current processor chips, multiprocessor nodes, and accelerators, as far as they are relevant for the practitioner. Peculiarities like SIMD vectorization, shared vs. separate caches, bandwidth bottlenecks, and ccNUMA characteristics are introduced, and the influence of system topology and affinity on the performance of typical parallel programming constructs is demonstrated. Performance engineering and performance patterns [1] are suggested as powerful tools that help the user understand the bottlenecks at hand and to assess the impact of possible code optimizations. A cornerstone of these concepts is the roofline model [2,3] , which is described in detail, including useful case studies, limits of its applicability, and possible refinements (ECM model [4,5]).

References

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POSTERS

Phase-of-the-phase spectroscopy in the multiphoton regime

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ABSTRACT

Recently, phase-of-the-phase (PoP) spectroscopy has been introduced and applied to the tunneling regime of strong-field ionization [1]. Briefly, the momentum-resolved photoelectron yield as a function of the relative phase between the strong ω and weak 2ω component of a colinearly polarized ω–2ω two-color pulse is measured and Fourier-transformed. This tells us how much and with which phase lag the yield changes with varying relative phase.

The canonical two-club structure of PoP spectra in the tunneling and rescattering regime were analyzed in Ref. [1]. On our poster, we present the corresponding results for the multiphoton regime. We find that the alternating PoP along the above-threshold ionization rings generates a characteristic checkerboard pattern in the PoP spectra, which corresponds to the „carpet“ structure in ordinary photoelectron spectra found previously [2]. The pattern is most clearly visible when the strong-field approximation (SFA) is applied. Within SFA, even an analytical formula can be derived. However, results obtained via the ab initio solution of the time-dependent Schrödinger equation show a more complex behavior of the momentum-resolved PoP due to long-range Coulomb effects on the outgoing electrons.

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Simulating the Dynamics of Superconducting Qudits with Iterative Time Ordering for Open Quantum Systems

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ABSTRACT

In order to obtain information about the dynamics of any quantum system, its equation of motion needs to be solved. An explicit time dependence of the Hamiltonian, for example due to an external driving field, introduces an additional challenge. The most commonly used propagation approaches usually rely on dividing the overall propagation time into small steps, where the time dependence of the Hamiltonian is approximately constant. This inevitably introduces inaccuracies due to neglected time ordering. In contrast, the iterative time ordering (ITO) approach allows to fully account for any explicit time dependence of the Hamiltonian. It was originally constructed for numerically exact propagation in Hilbert space. Here, we generalize it to Liouville space and optimize the implementation of the algorithm. We apply iterative time ordering to a superconducting phase qudit under Pythagorean control and show that highly accurate simulations can be carried out efficiently even if decoherence is explicitly accounted for.

An Analytic Mapping of Stacked Oligomer Potential Energy Surfaces to an Effective HJ-Aggregate Model: Vibronic Hamiltonian and Application to Exciton Migration

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ABSTRACT

As highlighted by recent experiments, exciton migration in functional organic materials can be guided by quantum coherence, despite the presence of a high-dimensional phonon environment. Hence, a conventional Förster theory treatment is not appropriate, and an explicit quantum dynamical approach is required. This, in turn, raises the question of constructing suitably parametrized potential energy surfaces (PESs), typically from oligomer ab initio electronic structure calculations [1,2].

As an extension to our recent work [3], we introduce here an analytic mapping procedure which allows for the direct parametrization of an HJ-aggregate model [4] as a function of interchain and intrachain modes [5]. The mapping procedure is based on the analytic solution of an inverse eigenvalue problem for an effective Frenkel Hamiltonian featuring interchain and intrachain next-neighbour couplings, which can be understood as a generalized multi-dimensional Hückel problem.

Based upon this parametrization, quantum dynamical simulations of exciton dynamics and spectral properties have been carried out for an asymmetric parallel stack composed of oligothiophene 10mer and 20mer units. Quantum dynamical calculations have been performed using the multi-layer Multiconfiguration Time-Dependent Hartree (MCTDH) method [6].

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Time-dependent renormalized natural orbital theory for intense laser-atom interaction

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ABSTRACT

Natural orbital theory (NOT) is a computationally useful approach to the few and many-body quantum problem. While natural orbitals (NOs) are known and applied since many years in electronic structure applications [1], their potential for time-dependent problems is being investigated only since recently [2, 3, 4, 5, 6]. We give a short introduction to the time-dependent renormalized NOT (TDRNOT) and test it on cases where time-dependent density (TDDFT) functional theory fails (doubly excited states, Fano profiles in absorption spectra, and nonsequential double ionization). Exact numerical solutions of the time-dependent Schrödinger equation for a laser-driven one-dimensional He model atom serve as benchmarks and show that TDRNOT is a promising approach indeed.

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Calculation of electronic spectra including vibrational pre-excitation and IVR

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ABSTRACT

The recently developed mixed IR/UV VIPER laser pulse sequence [1] has been developed to achieve selective photo-uncaging, i.e. the selective removal of a photolabile protecting group in a chemical or biological system. Theoretical calculations accompanying the experiments serve to determine the influence of vibrational pre-excitation on the UV/VIS absorption spectrum of the photocage. In this framework, the vibrational fine-structure of a single electronic transition was calculated. To this end, the high-dimensional vibrational wavepacket was propagated on the excited state potential energy surface (PES) within the harmonic approximation including Dushinsky rotation effects, employing the multilayer variant of the Multiconfigurational Time-Dependent Hartree (ML-MCTDH) method [2]. The resulting autocorrelation function was subsequently Fourier transformed to obtain the spectrum. The initial condition was prepared as a direct product of eigenfunctions of the ground state normal modes, with all modes in their respective ground states, except for one mode that was placed in its first excited state.

The effects of intramolecular vibrational energy redistribution (IVR) were analyzed by first propagating the vibrational wavepacket on the ground state PES, using the initial condition mentioned above. Anharmonic effects were introduced through the use of numerical third derivatives of the normal modes. After propagating for a chosen time interval, the resulting wavepacket was used as an initial condition for the calculation of the electronic spectrum.

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Unstable dynamic localization in tilted shaken optical lattices

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ABSTRACT

The existence of quasienergy bands in periodically driven lattices under the influence of an additional static force has first been predicted by J.Zak ["Finite Translations in Time and Energy", Phys. Rev. Lett 71, 2624 (1993)]. Among other things, it was suggested that within a single-band approximation such bands may take the form of cosine bands ”modulated with the Bessel function of the shaking amplitude”. Considering the experimentally accessible example of ultracold atoms in deep shaken cosine lattices, we show by numerical calculations of the full quasienergy spectrum that the single-band approximation is not reliable, and the quasienergy bands in fact are broken by a multitude of multiphoton resonances. This means that particles prepared in such bands tend to heat up on short time scales, and dynamic localization will be hard to observe.

Gaussian-based-multiconfiguration time-dependent Hartree: A two-layer approach

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ABSTRACT

Multiconfigurational Gaussian wavepacket (GWP) methods have proven versatile tools for on-the-fly dynamics in conjunction with electronic structure calculations, as well as the explicit representation of quantum mechanical system-bath problems. In this context, the variational Gaussian-based multiconfiguration time-dependent Hartree (G-MCTDH) approach [1] and its variational multiconfigurational Gaussians (vMCG) [2] variant were originally derived as semiclassical variants of the MCTDH method [3]. These methods mostly use Frozen Gaussian (FG) basis sets that are far less flexible than the single-particle function (SPF) representation of standard MCTDH, and may require a significantly larger number of GWPs to reach convergence.

To remedy the lack of flexibility of FG basis sets, we recently introduced a novel two-layer (2L-G-MCTDH) approach [4]: Here, the first layer is composed of flexible SPFs, while the second layer is composed of low-dimensional FGs. The numerical scaling properties are significantly improved as compared with the conventional G-MCTDH (vMCG) scheme. We report on the first implementation of the method in an in-house code, along with applications to (i) a model of site-to-site vibrational energy flow [5] and (ii) a multidimensional charge transfer model [6]. A perspective is given on the future combination with existing multi-layer MCTDH schemes; indeed, such a combination is straightforward since the first layer of the 2L-G-MCTDH approach can be chosen to be orthogonal.

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Ising tricriticality in the extended Hubbard model with bond dimerization

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ABSTRACT

The tricritical Ising universality class emerges at the end point of a line of continuous Ising phase transitions, above which the transition is first order. In 1+1 dimensions, it is described by the second minimal model of conformal field theory (CFT) with central charge c=7/10. Recently, there has been a revival of interest in the tricritical Ising model due to its emergent space-time supersymmetry. Combining numerical (infinite density-matrix renormalization group) and field theoretical (bosonization and CFT) techniques, there authors demonstrate here that Ising tricriticality occurs in a paradigmatic extended one dimensional Hubbard model with explicit bond dimerization. The low-energy sector can be described by a triple sine-Gordon model, which, in the transition region, reduces to the tricritical Ising CFT. The theoretically predicted decay of various two-point correlation functions is found to be in excellent agreement with the density-matrix renormalization group data.

References

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Nonequilibrium Kondo effect in a magnetic field: Auxiliary master equation approach

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ABSTRACT

We solve the single-impurity Anderson model in a magnetic field and out of equilibrium with an auxiliary master equation approach [1,2,3]. Treating the master equation within the framework of matrix product states allows us to generate highly accurate results, especially for the spectral functions. In equilibrium we find a remarkable agreement to spectral functions achieved with NRG, cf. [3]. The application of a bias voltage V and a magnetic field B both individually result in a splitting of the Kondo resonance around the Kondo temperature. With our method we can resolve a four-peak structure in the spectral function for nonzero B and V, due to both effects. This four-peak structure manifests itself in the differential conductance, which is very well accessible by experiments. We finally compare our results to recent experiments [4,5,6] and draw conclusions about the underlying spectral functions.

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Performing quantum dynamics simulations on nitrobenzene with the DD-vMCG method

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ABSTRACT

Nitrobenzene (nb) is the smallest nitroaromatic compound and yet is characterized by a challenging and rich photophysics and photochemistry. Experimentally, the broad absorption spectra recorded in both solution and gas-phase together with the lack of fluorescent and phosphorescent emissions have hampered the determination of the system photodynamics [1,2]. Theoretically, the electronic structure of the molecule has been proven to be significantly sensitive to the computational strategy employed, and to require third–order methods in order to obtain a sufficiently accurate description of its excited states [3]. The molecule is characterized by a high value of the triplet quantum yield (> 0.80), it is able to efficiently release the absorbed energy in a non-radiative way, and can undergo three different photodissociation processes leading respectively to NO_2, NO or O [2,4]. In our laboratory we are currently studying the system photoresponse performing direct quantum dynamics simulations using the direct-dynamic variational multi-configurational Gaussian (DD-vMCG) method implemented in the Quantics package [5,6], interfacing it with the Molcas program for electronic structure calculations [7]. The Molcas software has been selected due to the proven robustness of the implemented CASSCF and CASPT2 methods, which are used in the project in order to obtain Hessians, gradients and couplings (CASSCF) and energies (CASPT2), respectively. Due to the importance of the triplet manifold in nb, the dynamics include both singlet and triplet states, coupling them through the computation of the corresponding spin-orbit couplings. The results obtained up to date are here presented.

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Quantum description of spiropyran ring-opening

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ABSTRACT

The purpose of this work is to understand and modelize the photochromic isomerisation of spiropyran molecules. We mainly focus on the mechanism of light induced ring opening which is the first step of the reaction. This mechanism is characterized by a femtosecond time scale explained by a non-radiative decay at a conical intersection.[1]

We aim to describe this mechanism by the construction of coupled electronic potential energy surfaces, in high dimensionality (around 13 dimensions). Moreover, we take into account dynamical electronic correlation at a post-CASSCF ab-initio level (XMCQDPT2 [2]). The construction of this model is particularly challenging due to the large amplitude motion, that necessitate the use of curvilinear coordinates.

Once these surfaces are constructed, we will use the Multi Configuration Time Dependent Hartree (MCTDH) approach to perform non-adiabatic quantum dynamics. We will then simulate the experiments currently carried out in the group of Dijon(FRANCE), studying the ring-opening process by femtosecond spectroscopy coupled with LASER pulse control.

Experiments already showed the ultrafast time scale of the process and proved the possibility to change the yields, specifically using quantum interferences created by LASER coherent light.

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Caldeira-Leggett Model Description of Condensed Phase Vibrational Spectroscopy

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ABSTRACT

Formulating a rigorous system-bath partitioning approach remains an open issue. In this context the famous Caldeira-Leggett (CL) model that enables a simple modeling of system-bath interactions via so-called spectral density functions has enjoyed popularity. Thus, parametrizing these spectral densities, e.g. from explicit molecular dynamics data, is important for using the model. We discuss that this task is naturally achieved in frequency domain and propose a Fourier-based method that outperforms existing time-domain methods [1]. Moreover, the widely used rigid bond method turns out to be inappropriate in general and leads to a systematic overestimation of relaxation times, unless the system under study is indeed of the CL form. Although the CL model is a useful theoretical tool, its validity for describing anharmonic dynamics of real systems is often taken for granted. It is shown that such a use does not pass a self-consistency check for a broad class of solute-solvent systems, unless the system part of the potential is effectively harmonic, due to the "invertibility problem" [2]. In contrast, dynamics in solid environments or on surfaces can be well described by the CL model, even with anharmonic system forces [3]. It is thus important to address the validity of a CL description for the particular system under study. Such a check can be performed by comparing the spectra resulting from the corresponding generalized Langevin dynamics with their counterparts from explicit classical molecular dynamics.

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Adiabatic preparation of Floquet condensates

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ABSTRACT

We argue that a Bose-Einstein condensate can be transformed into a Floquet condensate, that is, into a periodically time-dependent many-particle state possessing the coherence properties of a mesoscopically occupied single-particle Floquet state. Our reasoning is based on the observation that the denseness of the many-body system's quasienergy spectrum does not necessarily obstruct effectively adiabatic transport. Employing the idealized model of a driven bosonic Josephson junction, we demonstrate that only a small amount of Floquet entropy is generated when a driving force with judiciously chosen frequency and maximum amplitude is turned on smoothly.

References

Christoph Heinisch, Martin Holthaus, arxiv.org/abs/1605.08199

The $2U_p$ plateau is a Coulomb effect

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ABSTRACT

The strong-field approximation (SFA) is known to reproduce photoelectron spectra of atoms in strong laser fields quite well (see [1] for a review). Nevertheless when it comes to the plateau structure up to $2U_p$ visible for certain parameter ranges the SFA fails as it only shows decay with increasing slope. Using the SFA in first order one can generate a plateau up to $2U_p$, but only in dependence on an arbitrary screening parameter. This indicates the relevance of scattering for the description of this feature. The simple mans theory (SMT) reproduces the $2U_p$ cutoff as well, but with vanishing probability since the latter is proportional to the electric field at emission time, and those electrons reaching the cutoff energy are emitted when the electric field approaches zero. Although this seems quite contradictory, it is rarely discussed in the literature.

We show that by applying a Coulomb correction inspired by [2] to the trajectory-based SFA [3-5] we can reproduce the plateau and cutoff as seen in calculations with the time-dependent Schrödinger equation (TDSE). This approach uses fully complex trajectories and is therefore independent of the (often somewhat arbitrary) definition of a tunnel exit as this point looses its physical (and numerical) significance. We compare this method to the plain SFA, the TDSE and a different Coulomb-corrected SFA used in previous works. The generation of the $2U_p$ plateau and cutoff due to the complex nature of the method is demonstrated. The emerging problem of branch cuts in the complex Coulomb potential (as described in detail in [6]) is discussed.

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Dynamics of quantum correlations in the Mott-insulator regime of the Bose-Hubbard model

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ABSTRACT

We present a general formalism for the solution of non-stationary lattice problems which deals with the dynamical equations for the reduced density matrices of different number of lattice sites and allows to consider nonlocal quantum correlations at arbitrary distances. Truncating the infinite set of equations taking into account only correlations of a certain number of sites. e.g., two or three, located at arbitrary distances from each other, the resulting system of equations can be easily solved numerically. Using this method, we have studied propagation of correlations as well as creation of the particle-hole pairs in the Mott-insulator regime of the Bose-Hubbard model.

Correlation effects in core hole spectroscopy with XUV-initiated high harmonic generation

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ABSTRACT

The creation of XUV pulses of attosecond duration by the high harmonic generation (HHG) process can be used for probing of atoms and molecular systems by their own electrons, yielding information on their electronic and nuclear structure. IR-field induced tunneling ionization is one of the key steps in the HHG process but makes it difficult or impossible to probe inner-shell electrons. However, by applying an XUV pulse, tunneling ionization can be replaced by one-photon ionization from the inner shell. This allows for the direct probing of inner-shell electrons and hence broadens the applicability of HHG [1].

Here we present a numerical simulation of a proof-of-principle experiment that demonstrates XUV-initiated HHG from the ground state of helium. Our model shows that the intensity of the high harmonics is increased by several orders of magnitude after including the XUV pulse. The simulations employ the recently developed time-dependent generalized-active-space configuration-interaction (TD-GAS-CI) theory [2–4]. Changing the delay between the IR and the XUV pulse gives characteristic oscillations in the HHG spectrum. TD-GAS-CI simulations show that electron correlation is essential for the description of these processes.

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Representation of Coupled Potential Energy Surfaces Using Neural Networks

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ABSTRACT

Theoretical and experimental research in chemical reaction dynamics has undergone major advances in the recent past, moving from very detailed studies of atom plus diatom to truly polyatomic systems. One problem which was adressed in this context, is the construction of accurate *ab initio* PESs in high dimensionality. Successful schemes were the Shepard interpolation approach introduced by Collins and coworkers and the invariant polynomial approach of Bowman, Braams and coworkers. Recently, the construction of PESs for polyatomic reactions using neural networks provided very promising results. However, allmost all work was focused on the construction of uncoupled adiabatic PESs.

In chemical reactions like $H_2+X \rightarrow H+HX$ and $CH_4+X \rightarrow HX+CH_3$ $(X = F, Cl, O(^3P))$ conical intersections are present in the entrance channel or even close to the transition state. Their accurate treatment requires multi-sheeted PESs. The present work investigates the promises of neural network for the construction of coupled diabatic PES describing chemical reactions. The key idea is to use adiabatic energies for fitting a diabatic Ansatz and to represent all elements of the diabatic matrix using a single neural network. First results studying the $H_2 + Cl \rightarrow H + HCl$ reaction will be presented.

Supersymmetry and eigensurface topology of pendular states

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ABSTRACT

We make use of supersymmetric quantum mechanics (SUSY QM) to find three sets of conditions under which the problem of a planar quantum pendulum with potential becomes analytically solvable. The analytic forms of the pendulum's eigenfuntions make it possible to find analytic expressions for observables of interest, such as the expectation values of the angular momentum squared and of the orientation and alignment cosines as well as of the eigenenergy. Furthermore, we find that the topology of the intersections of the pendulum's eigenenergy surfaces can be characterized by a single integer index whose values correspond to the sets of conditions under which the analytic solutions to the quantum pendulum problem exist. In the next stage we undertook a mutually complementary analytic and computational study of the full-edged spherical (3D) quantum rotor subject to combined orienting and aligning interactions characterized, respectively, by dimensionless parameters η and ξ. By making use of supersymmetric quantum mechanics (SUSY QM), we found two sets of conditions under which the problem of a spherical quantum pendulum becomes analytically solvable. These conditions coincide with the loci of the intersections of the eigenenergy surfaces spanned by the η and ξ parameters. The integer topological index k is independent of the eigenstate and thus of the projection quantum number m. These findings have repercussions for rotational spectra and dynamics of molecules subject to combined permanent and induced dipole interactions.

Emission properties of periodically driven quantum systems

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ABSTRACT

The time-evolution of a laser-driven quantum system is generated by a Hamiltonian with a periodic time-dependence. In order to compute the emission characteristics of such a system, we have to account for this periodicity and also for the coupling to the environment. Therefore, the driven system has to be described by a (Markovian) master equation with time-dependent coefficients. To obtain a simpler differential equation with constant coefficients, we first have to compute the Floquet states and then choose these states as the basis for the solution of the master equation. We demonstrate this procedure for the example of few emitters strongly coupled to a cavity mode and driven by an external laser. As an evidence for the dynamical Stark effect, we observe shifted peaks in the emission spectra for different laser intensities. The emission of nonclassical light can be analyzed with the Glauber function, which determines the statistics of emitted photons. We find clearly distinguished parameter regimes of super- and sub-Poissonian light emission and explain the additional features appearing for finite laser intensity in terms of the quasienergy spectrum of the driven emitter-cavity system.

Electronic Structure and Quantum Dynamics of an Oligothiophene H-Aggregate

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ABSTRACT

We present combined electronic structure and multiconfigurational quantum dynamics simulations for an oligothiophene H-aggregate, to investigate the ultrafast dynamics following photoexcitation in the full normal mode representation. Based on TDDFT calculations, a vibronic coupling model is constructed that comprises both Frenkel excitonic and charge-separated configurations. In recent experiments on related regioregular P3HT materials, long-lived periodic oscillations were detected and assigned to vibrational coherences from excited electronic states[a] [b]. However, the vibrational and/or electronic nature of these oscillations is still subject to debate. Our simulations suggest that the observed periodic oscillations stem from a combination of excitonic coherences and pronounced vibrational contributions in the same frequency range. Furthermore, we provide evidence for the ultrafast formation of inter-chain charge-transfer excitons in the neat material, as also observed experimentally. All dynamical calculations are carried out with the Multilayer Multiconfigurational Time Dependent Hartree (ML-MCTDH) package[c]. For an H-aggregate trimer, the model Hamiltonian features a total of seven electronic states and 120 normal modes. Additionally, calculations for an H-aggregate pentamer have been carried out, employing 13 electronic states and 200 normal modes.

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Effective-mode approach to coupled exciton/charge transfer dynamics in DNA

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ABSTRACT

Many photoactive polymer and biopolymer systems exhibit coupled Frenkel and charge-transfer (CT) excitonic manifolds, whose combined dynamics is strongly influenced by electron-phonon coupling. Here, we investigate specifically alternating cytosine-guanine $(CG)_n$ sequences occurring in natural and artificial DNA, which have been shown to feature pronounced CT exciton participation [Huix-Rotllant et al., JPCL 6, 2247 (2015)]. The relevant states can be described in terms of local Frenkel type $\pi\pi^*$ excitations and guanine-to-cytosine CT states.

Building upon TDDFT calculations, a linear vibronic coupling model for $(CG)_n$ sequences with state-specific spectral densities was constructed. In order to develop reduced-dimensional models, we employ effective-mode chain representations [Martinazzo et al., JCP 134, 011101 (2011)] that are specifically adapted to the case of correlated spectral densities. Several effective-mode layers are shown to yield good convergence properties. The Multi-Layer Multiconfiguration Time-Dependent Hartree (ML-MCTDH) [G.A. Worth et al. The MCTDH Pacakge, Version 8.5 (2013), 2006. http://www.pci.uni-heidelberg.de/tc/usr/mctdh/] method is employed for sequences comprising up to five $(CG)_n$ repeat units.

Applications of Chebyshev expansion in open Quantum systems

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ABSTRACT

The application of external fields or fluctuations within a molecular system's environment give rise to time-dependent perturbations in charge or energy transport processes. The theoretical treatment of time-dependent transport phenomena in such systems remains challenging. Quantum master equations, hierarchical equations, and equations based on non-equilibrium Green's functions are some of the approaches that may be employed to calculate the dynamics of such systems. Some of these approaches assume an exponential type of the corresponding correlation functions, which results in certain restrictions in the parameter space such as on the complexity of the system-environment coupling and cause numerical inefficiency at low temperatures.

These issues may be overcome by the utilization of a decomposition of the correlation functions constructed of Chebyshev polynomials and Bessel functions. Here we present the quantum master equations in second-order perturbation theory developed using the Chebyshev expansion and applied to both fermionic and bosonic systems. The proposed set of differential equations are benchmarked against existing methods for simple systems. The improvements and shortcomings brought about by the Chebyshev approach are discussed.

Calculating vibrational spectra of molecules using tensor train decomposition

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ABSTRACT

In this work we propose a new algorithm for solving vibrational Schroedinger equation. The main difficulty to solve this type of problems is that the number of parameters grows exponentially with the dimension. To avoid exponential growth of storage we use tensor train (TT) decomposition to approximate a solution. Despite there exist a number of methods to approximate eigenvalues using tensor formats, we found that they become impractical when a lot of eigenvalues (>100) are needed.

To solve this problem we propose a manifold-preconditioning concept to well established iterative methods. This approach explicitly incorporates the knowledge that the solution can be well approximated in the TT-format, i.e. it lies on a certain low-dimensional non-linear manifold.

We calculate vibrational spectra of acetonitrile molecule (100 eigenstates) and compare our results with those of the H-RRBPM method by Carrington and Thomas [J. Phys. Chem. A, 119 (52), 2015]. We show that with the same amount of storage and computational time our method significantly outperforms accuracy of the H-RRBPM. Moreover, we show that our method provides more accurate results than the sparse grid approach and is more memory efficient.

Exact simulation of intense-laser-driven helium using TDRNOT

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ABSTRACT

Correlated phenomena of laser-atom interaction are inaccessible in TDDFT using any practicable approximation to the exchange-correlation functional. We study an alternate approach to the driven quantum few-body problem called *time-dependent renormalized-natural-orbital theory* (TDRNOT). Renormalized natural orbitals (RNOs) are eigenfunctions of the one-body reduced density matrix (1-RDM). By using those orbitals as the basic quantity, TDRNOT is supposed to overcome some limitations of TDDFT which is based on the *diagonal* of the 1-RDM—i.e., the single-particle density—alone.

Considering a 1D helium model atom, TDRNOT has already proven to describe doubly excited states including autoionization [1], Rabi oscillations [2], nonsequential double ionization [3], and Fano profiles in radiation absorption spectra [4] as well as the second plateau in high-harmonic generation from He caused by $\mathrm{He}^{+} \leftrightarrow \mathrm{He}^{2+}$ transitions [4]. The good performance of two-particle TDRNOT results from an exact, explicit, and adiabatic (i.e., memory-free) expression of the 2-RDM in terms of the RNOs [2].

This poster addresses TDRNOT for a possibly laser-driven helium atom in full dimensionality: Firstly, equations of motion for the RNOs expanded in spherical harmonics are shown. Moreover, we present the simplifications of the RNO structure if one restricts to the groundstate configuration with $L = M = 0$ or to linearly polarized laser fields with $M = 0$. Finally, the poster includes recent results of our efforts on reproducing the 1D results in full dimensionality.

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System-Bath Dynamics For Driven Environments

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ABSTRACT

"Commonly, nanosystems are characterized by their response to time-dependent external fields in the presence of inevitable environmental fluctuations. The direct impact of the external driving on the environment is generally neglected. While this approach is satisfactory for macroscopic systems, on the nanoscale, an interaction of external fields with the environment is often unavoidable on principle. We extend the standard linear response theory of quantum dissipative systems to strongly driven baths. Significant modifications are found for two paradigm examples. First, we evaluate the polarizability of a molecule immersed in a strongly polarizable medium that responds to terahertz radiation. We find an increase of the molecular polarizability by about 30%. Second, we determine the response of a semiconductor quantum dot in close proximity to a metallic nanoparticle. Both are placed in a polarizable medium and exposed to electromagnetic irradiation. We show that the response of the quantum dot is qualitatively modified by the driven nanoparticle, including the generation of an additional channel of stimulated emission."

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Unraveling the Quantum State Mixing of Excitonic and Vibronic Excitations in the Dynamics of Molecular Aggregates

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ABSTRACT

One possible source of coherent oscillations observed in two-dimensional electronic spectroscopy experiments is the quantum state mixing (QSM) of electronic and vibronic excitations in molecular aggregates. In the present contribution the signatures of Coulomb coupling induced QSM between excitonic and vibronic excitations in the dynamics of a model aggregate are discussed. To this end numerically exact dissipative exciton dynamics calculations applying the hierarchy equations of motion (HEOM) method are performed. A Fourier analysis of the coherent oscillations in the population dynamics after an initial excitation of the highest exciton state is supplemented by exciton-vibronic structure calculations employing direct diagonalization [1]. This allows us to unravel the origin of the oscillations and to obtain insights on the general influence of QMS on the dynamics as a function of Coulomb and vibronic coupling strengths [2].

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Correlated Exciton-Vibrational Dynamics in the FMO Complex

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ABSTRACT

Excitation energy transfer (EET) is one of the most fundamental processes in nature. A controversial discussion is focused on the role of coupled quantum dynamics of the excitonic and vibrational degrees of freedom. Especially, the question to which extend vibrational-assisted resonances facilitate an efficient exciton transfer has been in the focus of interest [1-3]. In the present contribution we study the exciton-vibrational dynamics by solving the time-dependent Schrödinger equation for a high-dimensional model of the Fenna-Matthews-Olson (FMO) complex by applying the multilayer multiconfiguration time-dependent Hartree formalism. This allows us, for the first time, to access information on the dynamics of electronic ground/excited state vibrational/vibronic excitations during the coherent excitation energy transfer in this complex. Different EET pathways are analyzed for the seven site and the eight site models of the FMO complex by means of the reduced one-exciton density matrix.The question of the effect of initial state preparation is addressed by comparing the case of an instantaneous Franck-Condon excitation at a single site with that of a laser field excitation [4,5].

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Quantum dynamics calculations of the metastable prereactive $F \cdot CH_4$ complex

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ABSTRACT

The $F + CH_4 \rightarrow HF + CH_3$ reaction is a prototypical example of an early barrier reaction. Prereactive complexes are assumed to play an important role in the dynamics of the reaction. The long-living resonance states resulting from the formation of a metastable $F \cdot CH_4$ complex are investigated in detail. Full-dimensional quantum dynamics calculations employing the multi-configurational time-dependent Hartree (MCTDH) approach and its multi-layer extension are presented. Block relaxation using a state-averaged MCTDH descripiton is used to obtain the quasi-bound energy levels. Results employing a single adiabatic potential energy surface to study the low-lying quasi-bound states of the $F \cdot CH_4$ complex for vanishing total (nuclear) angular momentum are presented. The computed dissociation energy of the $F \cdot CH_4$ complex with respect to the reactant asymptote is 170 cm^{-1}. About 60 resonance states with energies below the reactant asymptote are found. A detailed analysis of the computed wavefunction of the low-lying states shows an almost free relative rotation of F and CH_4 and an approximately separable F-CH_4 stretching vibration. The present results are compared with transition state spectroscopy experiments which study the photodetachment spectrum of the $F \cdot CH_4^-$ anion.

Time-resolved core-level photoemission studied by quantum kinetics of electron-boson models

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ABSTRACT

Recent experiments allow following the time-resolved photoelectron signal originating from plasmon satellites in correlated materials and so open the door for studying the build-up and decay dynamics of screening effects in real time. Motivated by these developments, we present the time-dependent Green's function formalsm - the Kadanoff-Baym equations - for the nonequilibrium time evolution of interacting fermions and bosons. In contrast to the fermionic case, the bosons are described by second-order differential equations. The solution of the bosonic Kadanoff-Baym equations - which is the central ingredient of this work - requires substantial modification of the usual two-times electronic propagation scheme. Our approach treats both degrees of freedom on equal footing and can be applied to a number of problems, such as the interaction of electrons with quantized photons, phonons, and other bosonic excitations. Here the formalism is applied to the photoemission from a deep core hole accompanied by plasmon excitation. We compute the time-resolved photoelectron spectra and discuss the effects of intrinsic and extrinsic electron energy losses and their interference.

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Towards "Mott Solar Cells"

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ABSTRACT

Recently, Mott-insulating heterostructures have been proposed as candidates for highly efficient solar cells [1]. Here, photoexcited doublons and holes act as charge carriers which can proliferate due to impact ionisation processes [2]. Previous works have investigated the doublon dynamics in such systems within time-dependent Dynamical Mean-Field Theory (DMFT) by looking at the time evolution after a photoexcitation [3].

In the present work we focus on the (quasi-) steady state of periodically driven quantum systems. Specifically, we implement an algorithm to deal with periodic steady states of strongly correlated systems, making use of the nonequilibrium Floquet Green's function formalism within the DMFT approximation.Our model consists of a correlated layer subject to a periodic driving via a homogeneous electric field and coupled to leads with different chemical potentials.

We present results obtained with a Floquet DMFT implementation using the Auxiliary Master Equation Approach (AMEA) [4] as an impurity solver. AMEA is based upon mapping the system to an open quantum system described by a Lindblad Master Equation. This allows the impurity to be affected by short-ranged non-Markovian dynamics. For comparison, we also carry out calculations on the same model within iterated perturbation theory [5].

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Nonlinear dynamics and quantum multistability of optomechanical systems

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ABSTRACT

We study the dynamics of two optomechanical systems, the cavity-cantilever and the membrane-in-the-middle setup, with a particular focus on the nonlinear classical dynamics and the transition into the quantum regime. We start with the analysis of the classical dynamics, where we identify fixed point bifurcations, Hopf bifurcations leading to self-induced oscillations, and period doubling bifurcations on the route to chaos. A typical signature of both systems is the multistability of solutions, which manifests itself through the coexistence of several stable orbits at different amplitudes. Then, we study how the classical multistability is realized in the quantum regime. There, new dynamical patterns appear because quantum trajectories can move between different classical orbits. We explain the resulting quantum dynamics from the phase space point of view, and provide a quantitative description in terms of autocorrelation functions. In this way we can identify clear dynamical signatures of the crossover from classical to quantum mechanics in experimentally accessible quantities. Finally, we discuss a possible interpretation of our results in the sense that quantum mechanics protects optomechanical systems against the chaotic dynamics realized in the classical limit.

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