43. EAS - Meeting "Extreme Atomic Systems"

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– list of abstracts –

David Bachmann, Albert-Ludwigs-University, Freiburg

Exploring new possibilities for free-space quantum communication

Free-space quantum communication is an active, application-oriented research area responding to the ever-growing demand of secure and flexible communication channels. We show that instantaneous spatial singular modes of light in a dynamically evolving, turbulent atmosphere offer significantly improved high-fidelity signal transmission as compared to standard encoding bases corrected by adaptive optics. Their enhanced stability in stronger turbulence is associated with suppressed intermodal crosstalk and a subdiffusive algebraic decay of the transmitted power with evolution time.

Baghdasar Baghdasaryan, Friedrich-Schiller-University, Jena

The Gouy Phase in Parametric Downconversion: Decoupling of Spatial and Spectral Degrees of Freedom

Spontaneous parametric down-conversion (SPDC) is a widely used source for studying and applying photonic entanglement. Years of focused research have led to a solid understanding of the process, but a cohesive description of the paraxial two-photon state has yet to be achieved. In particular, SPDC is usually described in the narrowband or thin-crystal approximations at degenerate frequencies. Taking the full spectral dependence into account is an important step towards describing realistic experimental settings and engineering quantum states with high purity and efficiency. We derived a general expression for the spatio-temporal two-photon state that applies universally across common experimental settings and correctly describes the non-separability of spatial- and spectral modes. In particular, we find that higher-order spatial modes generated in SPDC have different spectral properties. This fact must be taken into account in the development of pure spatial entanglement. We formulate a criterion, how to decouple the spatial from the spectral degree of freedom by taking into account the Gouy Phase of interacting beams. This work provides new insights into the role of the Gouy phase in nonlinear processes, and also into the preparation of engineered entangled states for multidimensional quantum information processing.

Dr. Andrei Benediktovitch, Deutsches Elektronen-Synchrotron (DESY)

X-ray Laser Oscillator at LCLS: first steps in modeling and experimental realization

X-ray Free Electron Lasers (XFEL) have opened a new page in the x-ray – matter interaction studies, however, the stochastic temporal profile of XFEL limits the applicable experimental techniques. To overcome these limitations, the X-ray Laser Oscillator (XLO) concept aims to produce repeatable and highly temporally-coherent pulses. In the XLO concept, XFEL radiation is focused on the atomic medium and initiates collective emission of characteristic x-ray radiation pulse. This radiation is looped with help of Bragg reflection from crystals arranged to form a cavity and is used as a seed for the emission process initiated by the next XFEL pulse.

There are numerous challenges in the modeling of the corresponding processes and in the technical realization that is underway at LCLS. In this talk, we will describe our progress in modeling – which is based on a stochastic partial differential approach to the collective emission of characteristic x-ray radiation initiated by XFEL pulse. Also, the current status of the XLO setup at LCLS will be described.

Dana Bloß, University of Kassel

X-ray-induced ionisation of solvated metal ions

To understanding radiation damage on a molecular level one way is to look at the microscopic response of biological systems to ionizing radiation. Of special interest in this context are low-energy electrons (LEEs), which usually have energies below 30 eV and are known to be responsible for DNA double-strand breaks as well as for the creation of reactive species in the water surrounding of the DNA [1].The main source of these LEEs upon X-ray irradiation are secondary processes induced by photo- and Auger electrons or the scattering of the photo- and Auger electrons themselves on their way through the environment. Recently, some of the secondary processes like energy-transfer processes as the interatomic Coulombic decay (ICD) or charge-transfer processes as the electron-transfer-mediated decay (ETMD) have attracted attention as a source to the efficient emission of LEEs close to the site of ionization [2]. ICD and ETMD have been studied intensely throughout the last decades in clusters and solutions, and their role in radiation biology and potential for medical applications is lively debated [1]. Stumpf et al. predicted that X-ray ionization of microsolvated metal ions, e.g., Mg2+, results in a multi-step cascade of ICD and ETMD processes [3]. We experimentally investigate the theoretically predicted LEE emission for Mg^{2+}, Ca^{2+} and Al^{3+} solutions at different synchrotron facilities with a combination of coincident electron detection and liquid microjets.

[1] E. Alizadeh et al., Annu. Rev. Phys. Chem. 66, 379 (2015)
[2] T. Jahnke et al., Chem. Rev. 120, 11295 (2020)
[3] V. Stumpf et al., Nat. Chem. 8, 237 (2016)

Eric Brunner, Albert-Ludwigs-University, Freiburg

Many-body interference at the onset of chaos

We unveil the signature of many-body interference across dynamical regimes of the Bose-Hubbard model. Increasing the particles' indistinguishability enhances the temporal fluctuations of few-body observables, with a dramatic amplification at the onset of quantum chaos. By resolving the exchange symmetries of partially distinguishable particles, we explain this amplification as the fingerprint of the initial state's coherences in the energy eigenbasis. In the domain of fully developed quantum chaos, ergodic delocalisation of the eigenstates suppresses this fingerprint.

Stasis Chuchurka, Deutsches Elektronen-Synchrotron (DESY)

Stochastic methodology for light-matter interaction in cavities based on positive P representation

Stochastic methodology for light-matter interaction in cavities based on positive P representation Without any assumptions and approximations, realistic quantum systems have an exponentially large number of degrees of freedom. In some cases, the system is permutationally symmetric, and the number of degrees of freedom is significantly reduced [1]. However, even this reduction is not sufficient for a large number of particles. Phase-space methods are capable of resolving this curse of dimensionality. A high-dimensional density matrix can be replaced by a quasi-probability distribution function [2]. In some cases, this distribution function satisfies the Fokker-Planck equation and may be sampled by stochastic trajectories, with quantumness inscribed at the correlation properties of stochastic processes. The price of this is the intrinsic instability of stochastic equations [3].
Previously, we successfully applied this methodology to the problem of supperradiance [4] in free space. This positive experience motivated us to test the formalism on quantum systems in cavities. Besides classical models of quantum optics (Dicke and Tavis-Cummings), we consider more practical problems such as non-Markovian dynamics in organic polaritons. In the talk, I will demonstrate our stochastic formalism on the example of these models and share some insights and problems that we encountered in our studies.

[1] V. Sukharnikov, S. Chuchurka, A. Benediktovitch and N. Rohringer, Second quantization of open quantum systems in Liouville space, arXiv preprint arXiv:2207.14234 (2022).
[2] P. D. Drummond and M. Hillery, The quantum theory of nonlinear optics (2014).
[3] P. Deuar and P. D. Drummond, Physical Review A 66, 033812 (2002).
[4] Gross, M., Haroche, S. (1982). Superradiance: An essay on the theory of collective spontaneous emission. Physics Reports, 93(5), 301–396.

Dr. Christoph Dittel, Albert-Ludwigs-University, Freiburg
Monogamy in the symmetry of many-body quantum states

We show that, for general many-body quantum states, the symmetry (i.e., the expectation value of the particles' exchange operator) with respect to the exchange of any two particles is monogamous. Using group representation theory, we first lift the hitherto known monogamy relation for the particular case of three entangled, distinguishable particles [M. Karczewski, et al., Phys. Rev. Lett. 121, 090403 (2018)] to the realm of general three-body quantum states. We then investigate the nontrivial extension to four particles, and identify an additional inequality through which monogamy in the symmetry of general four-body quantum states can exhaustively be understood. The tightness of our relations are verified numerically, and the challenges for future generalizations to larger particle numbers are discussed.

Helena Drüeke, University of Rostock
Interaction-Induced Directional Transport on Periodically Driven Coupled Chains

Almost all hopping Hamiltonians can nowadays be implemented on cold-atom or photonic-waveguide platforms. This also includes interacting and driven (Floquet) systems. We discuss a simple, driven system in which interaction induces directional transport. To that end, we consider two chains a and b of equal length. On each chain, one particle hops while hoppings to the other chain are forbidden. However, the interaction between the two particles is across the chains. As a condensed-matter analog, one may think of interlayer excitons in a layered material.
The periodic driving of the system consists of two phases. Only particle a moves (isotropically to the left and right) during the first phase. During the second one, particle b moves. Without interaction, both particles diffuse along their respective chains. Interaction leads to directional transport; the two particles perform a leapfrogging motion. The relative position of the two particles in the starting configuration determines whether they move to the left, move to the right or remain stationary. We investigate correlations between the particles by mapping the two 1D coordinates to x- and y-coordinates of a single 2D particle on a square grid.

Dr. Matthew Eiles, Max-Planck Institute for the Physics of Complex Systems, Dresden
A trip to the quantum zoo

I will discuss the formation and properties of an unusual class of diatomic molecules known as long-range Rydberg molecules. Although these molecules are, at the level of their constituent atoms, little more complicated than H_2, their highly excited electronic states reveal rich physics at nearly macroscopic dimensions. I will show how the binding mechanism of these molecules can be reformulated and interpreted using a "dressed ion pair" model, i.e. $H^+ + H^-$, but with the twist that the charge on the negative ion is much smaller than that of an electron. This model leads to a powerful framework for understanding the behavior of these molecules, and allows us to predict an infinitely growing collection of such quantum creatures.

Prof. Jörg Evers, Max-Planck Institute for Nuclear Physics, Heidelberg
Direct x-ray excitation of the ultra-narrow nuclear 12.4 keV resonance of ^{45}Sc

In this talk, I will present recent theoretical and experimental progress on the direct x-ray-excitation of the 12.4 keV nuclear resonance of ^{45}Sc, which has a natural lifetime of approximately 460ms, corresponding to $\Delta E/E \sim 10^{-19}$. As compared to ^{229m}Th, the scandium isotope ^{45}Sc has the advantage that it is readily commercially available, that it exhibits a high Lamb-Mössbauer factor close to 0.8 already at room temperature, and that its natural life time does not suffer from an enormously high internal conversion factor. After an introduction and overview to the field, first results from an experiment at the European XFEL will be presented, which delivers x-ray pulses with a time structure ideally matching the lifetime of ^{45}Sc.

Prof. Stephan Fritzsche, Helmholtz Institute, Jena
Lorentz-force shifts in strong-field ionization with mid-infrared laser fields

In the past, the ionization of atoms and molecules by strong, mid-infrared (IR) laser fields has attracted recurrent interest. Measurements with different IR pulses have demonstrated the crucial role of the magnetic field on the electron dynamics, classically known as the Lorentz force, that acts upon all particles with charge q in motion. In practice, however, the strong-field approximation (SFA) is typically based on the dipole approximation alone and neglects both the magnetic field and the spatial dependence of the driving electric field.
Here we show and discuss that several, if not most, observations from strong-field ionization experiments with mid-IR fields can be quantitatively explained within the framework of SFA, if the Lorentz force is taken into account by nondipole Volkov states in the formalism. Details of such a treatment are explained for the (peak) shifts of the polar-angle distribution of above-threshold ionization photoelectrons along the laser propagation, the steering of electron momenta by two not quite collinear laser beams, or the enhanced momentum transfer to photoelectrons in standing-light fields.

Miriam Gerharz, Max-Planck Institute for Nuclear Physics, Heidelberg
Dynamically controllable resonant x-ray optics and interferometry

In this project we introduce a concept for dynamically controllable resonant x-ray optics. Using piezo-control methods, we can displace a solid-state target much faster than the lifetime of its resonances. This creates a mechanically induced phase shift, which can be associated with a frequency-dependent effective refractive index $n(\omega)$ of the moving target. Hence, we can achieve polarization control by mechanically induced birefringence. We theoretically and experimentally demonstrate the approach with a x-ray polarization interferometer, in which the interference is controlled by the mechanically-induced refractive-index control. This setup can be used for temporal gating and provides a sensitive tool for a noise background analysis on sub-Ångstrom level.

Igor Gianardi, Max-Planck-Institut for the Physics of Complex Systems, Dresden
Manipulating materials at lattice scale with cavity quantum light

The possibility of manipulating quantum materials' proprieties through their coupling with the quantum vacuum fluctuations of a cavity electromagnetic field has recently been investigated both theoretically and experimentally.
While the most recent experiments fully corroborate this proposal, on the other hand a satisfactory theory shedding light on the underlying mechanism remains absent to date. A key theoretical advance is to go beyond phenomenological models based on the homogeneous single-mode approximation, which are incapable of making reliable predictions about the magnitude or even the sign of vacuum-induced effects. Consequently we carried out a study of these effects on a 2D quantum material, taking into account the full electromagnetic spectrum of the cavity field. As a paradigmatic example we considered an "extended Fermi-Hubbard model" on the honeycomb lattice coupled to an electromagnetic field inside a Fabry-Pérot cavity.
This model provides an optimal platform to explore the possibility of changing the semi-metallic phase of graphene into an insulating phase by tuning the electron interactions at lattice scale via the cavity.
We obtained that the cavity disfavours the antiferromagnetic and charge density wave instabilities leading to the insulating phases, favouring rather a ferromagnetic ordering, contrary to the results obtained so far in the literature. Furthermore our findings clearly show that the EM-mode-volume compression induced by the cavity per se is not sufficient to explain the substantial vacuum-induced corrections which have been observed in recent experiments. Rather, we hypothesise that the cavity's effectiveness in modifying material properties may rely on the pronounced dynamical (as opposed to static) nature of electron-electron interactions mediated by it.

Prof. Stefanie Gräfe, Friedrich Schiller University, Jena
Attosecond time-resolved electronic and vibrational dynamics in furan

Non-adiabatic dynamics and conical intersections play a central role in the chemistry of most polyatomic molecules, ranging from isomerization to heterocyclic ring opening and avoided photo-damage of DNA. Studying the underpinning correlated dynamics of electronic and nuclear wave packets is a major challenge in real-time and many times involves optically dark transient states. In close collaboration with the group of Prof. Jens Biegert (ICFO, Spain), we show that attosecond core-level spectroscopy reveals the pathway dynamics of neutral furan across its conical intersections and dark states. Our method measures electronic-nuclear correlations to detect the dephasing of electronic coherence due to nuclear motion and identifies the ring-opened isomer as the dominant product. These results demonstrate the efficacy of attosecond core level spectroscopy as a potent method to investigate the real-time dynamics of photochemical reaction pathways in complex molecular systems.

Prof. Frank Großmann, Technical University Dresden (TUD)
Non-Markovianity in open quantum systems: Vibrational relaxation at surfaces

We review the multi Davydov ansatz for the solution of the time-dependent Schroedinger equation of a multi-level system coupled to a (finite) bath of harmonic oscillator modes. After an introduction with a focus on spin-boson dynamics, we investigate the vibrational relaxation at surfaces, focussing on the D-Si-Si bending mode at a D:Si(100)-(2x1) surface. Here a hierarchical effective mode model [1] allows to effectively treat a bath of more than 2000 phonon modes.
The multi Davydov results are benchmarked against MCTDH calculations and the non-Markovianity of the dynamics is quantified by a comparison to a Lindblad-type Liouville-von-Neumann approach [2]. Among other markers of non-Markovianity, we observe blackflow of energy from the bath into the system.

[1] E. W. Fischer et al., J. Chem. Phys. 153 064704 (2020)
[2] E. W. Fischer et al., J. Chem. Phys. 156 214702 (2022)

Lennart Guth, Max-Planck Institute for Nuclear Physics, Heidelberg
XUV frequency comb for precision spectroscopy of trapped highly charged ions

Highly charged ions have a few tightly bound electrons, which allow to probe fundamental physics and develop new frequency standards. However, most transitions are in the extreme ultraviolet (XUV)[1]. To perform spectroscopy on these with unprecedented precision, we have built an XUV frequency comb by transferring the coherence and stability of a near-infrared (NIR) frequency comb to the XUV using high-harmonic generation [2]. To reach the required peak intensity, NIR frequency comb pulses (200 fs) are amplified to 80 W in a chirped pulse fiber amplifier and resonantly overlapped in a femtosecond en- hancement cavity ($P_{peak}25kW$,$\mathrm{I}_{peak}31014W/cm2$) [3]. High harmonics up to the 35th order are coupled out of the cavity and will be used for future direct XUV spectroscopy of highly charged ions, trapped and sympathetically cooled in a superconducting Paul trap [4].

[1] M.S. Safronova et al., Phys. Rev. Lett. 113, 030801 (2014).
[2] G. Porat et al., Nat. Photon, 12, 387 - 391 (2018).
[3] J. Nauta et al., Nucl. Instrum. Meth. B 408, 285 (2017).
[4] J. Stark et al., Rev. Sci. Instrum., 92, 083203 (2021).

Dr. Andreas Hans, University of Kassel

New insight into interatomic interactions in clusters through coincident electron and photon spectroscopy

The role of inter-particle charge and energy transfer mechanisms in radiation biology is currently lively debated due to emerging genotoxic products such as radicals and slow electrons. The coincident detection of reaction products enables unprecedented conclusions about such processes following light-matter interactions. While coincidence experiments in the past have largely concentrated on charged particles, I will present recent progress in the experimental realization of multielectron-photon coincidence spectroscopy. Applied to atomic clusters, this technique allows for identification and quantification of otherwise elusive processes. I will discuss the examples of core-level interatomic Coulombic decay and Auger decay in the vicinity of a positive charge.

Yu He, Max-Planck Institute for Nuclear Physics, Heidelberg

Resonant Perfect Absorption and Autoionization Dynamics Revealed by Attosecond Transient Absorption Spectroscopy

We introduce a general approach to manipulate and substantially enhance the resonant absorption property of a macroscopic medium. By emptying the population of the excited state after its excitation, the polarization decay of the target system is temporally reshaped and confined. The tunable temporal gate between excitation and termination allows us to tailor the tail of the excitation pulse developed during propagation, which thus interferes controllably with the original pulse. Numerical and analytical results on an ensemble of two-level systems demonstrate that the resonant absorption of light can be reduced or significantly enhanced by more than 5 orders of magnitude relative to that without laser manipulation, and resonant "perfect absorption" can be achieved at certain conditions. These results are further supported by large-scale calculations of the coupled time-dependent Schrödinger and Maxwell wave equations in helium. Experimentally, we report the transient-absorption measurement of sp2,n± autoionization states in helium gas. The spectral signature of sp2,4- shows up in the presence of a moderately intense visible pulse, which is otherwise suppressed due to its low dipole coupling to the ground state. The different temporal dynamics of these states are analyzed, and the roles of propagation effects in the line-shape manipulation are discussed.

Reference: Yu He et al., Resonant Perfect Absorption Yielded by Zero-Area Pulses, Phys. Rev. Lett. 129, 273201 (2022)

Emilia Heikura, University of Kassel

Distance dependency of photoelectron circular dichroism in chiral molecules

One of the most powerful methods to investigate molecular chirality in the gas phase is photoelectron circular dichroism (PECD). PECD arises from the asymmetry of the angular distribution of photoelectrons (probe electrons) scattered on the chiral backbone of the molecule even from randomly oriented molecules after interaction with circularly polarized ligh. One still unknown aspect of PECD is, how the distance between the probe electron emitter site and the center of a point-chiral molecule affects the magnitude of the PECD asymmetry. A velocity map imaging (VMI) electron spectrometer was used to measure these forward-backward asymmetries of emitted photoelectrons. Measurements were performed on sec-butyl trimethylsilylether and its derivatives which have been specifically synthesized for these experiments by the group of R. Pietschnig (Uni Kassel).

Dr. Markus Ilchen, Deutsches Elektronen-Synchrotron (DESY)
Observing electron dynamics in chiral systems from specific atomic sites

Polarization-controlled free-electron lasers (FELs) provide unique access to nonlinear and time-resolving investigations of transient and otherwise inaccessible states of matter. They have demonstrated rich potential for magnetization studies at a variety of facilities, but also open new avenues for gas-phase science. The emerging field of site-specific investigations of dynamics in (transient) chiral matter, i.e., matter that possesses a handedness, is an important example in this regard. Chirality has a profound relevance for a multitude of reasons. In fact, it influences our all-day-life and is discussed to shape the fabric of our existence to a great extent. Gas-phase studies of chiral molecules at FELs can unprecedentedly add new perspectives on their (re-)formation dynamics, their ultrafast response to light of different energies in general, their ionization dynamics and ionic functionality, and the relevance of a stereocenter-site for an effective choice of methodology.
The rapid evolution of technological advances at (X)FELs has leveraged the accessibility of this new topic, among others, by enabling narrow-bandwidth operation via seeding, multi-color schemes for FEL-pump FEL-probe methodology, isolated attosecond pulses, high-repetition rates, and finally, undulator-based polarization control. Only by combining these technological capabilities, we will ultimately enable a new depth of observing and steering ultrafast dynamics in chiral systems. However, already the metrology of such complex pulse properties, in particular at SASE-FELs, remains a challenge for itself, yet being a prerequisite for this young field of science.
This contribution will present the initial milestones of chirality science at FELs in the gas phase, the status of advanced metrology schemes for variably polarized, high-power (X)FEL pulses at the attosecond frontier, and the advent of related scientific prospects that are about to emerge at a variety of facilities worldwide. Exemplarily, the FLASH 2020+ agenda and the underlying exploitation strategy at DESY in Germany for this new field of interest will be sketched and discussed.

Dr. Daniil Kartashov, Friedrich-Schiller-University, Jena
High-order harmonic generation and confinement in artificial atoms

In this contribution we report on the results of experimental and theoretical investigation of high-order harmonic generation (HHG) in semiconductor quantum dots. Harmonic spectra were measured from layers of CdSe quantum dots in dependence on dot's size, laser wavelength, polarization and intensity. We observe drastically different dependence of harmonic yield on dot's diameter for harmonics with energies of quanta less than the bandgap (intraband harmonics, generated by the nonlinear Bloch current), and for harmonics with energies of quanta above the bandgap (Corkum interband recombination mechanism). When intraband harmonics demonstrate qualitatively the same behavior with reducing the dot's diameter, the yield of interband harmonics drops abruptly for dot's with the diameter less than 3 nm. Employing various theoretical approaches, including real-time time-dependent density functional, semi-empirical tight-binding and classical calculations, as well as quantum dynamical calculations for a single electron time-dependent Schrödinger equation, we suggest a mechanism that the drop-off for the above-bandgap harmonics for the smallest dots is caused by electron scattering off the dot's walls that works as an effective dephasing mechanism. Thus our results demonstrate the sensitivity of HHG to spatial confinement of the electron wavepacket motion in nanoscale solids.

Dr. Gregor Kastirke, Goethe University, Frankfurt
Charge-up and fragmentation dynamics of oxygen molecules detected by the COLTRIMS Reaction Microscope at European XFEL

The European X-ray free-electron laser (EuXFEL) has enabled the study of light-matter interaction under extreme conditions. Atoms and molecules which are irradiated by the XFEL are charged by a complex interplay of several subsequent photoionization events and electronic decay processes within a few femtoseconds. The fragmentation dynamics of oxygen molecules have been observed by measuring ions and electrons in coincidence. A newly designed COLTRIMS Reaction Microscope (REMI) was installed at the Small-Quantum-Systems-Instrument at SASE3-Beamline. Therefore, many of the components of the REMI end-station were specially adapted like a new target preparation system, improved spectrometer and the latest type of time and position sensitive multi-hit detectors.

Dr. Victor Kimberg, KTH Royal Institute of Technology, Stockholm
X-ray fluorescence imaging of nonlinear pulse propagation dynamics

It is a long-standing vision to extend concepts of non-linear optics into the X-ray regime, where short-lived local quasi-atomic core excitations give access to interactions on atomic length and time scales. Modern XFEL sources indeed allow investigation of nonlinear processes such as stimulated X-ray emission and Raman scattering, amplified X-ray emission, pulse compression, self-induced transparency, etc. The nonlinear effects for an optically thick medium with high aspect ratio are directed along the incident pump or seed beams and conventionally observed online the propagation direction showing an integral picture that does not allow to follow the propagation evolution point-by-point through the medium. Here we suggest a novel scheme where this evolution can be monitored by measuring the quasi-isotropic secondary decay of states, populated in the stimulated emission process, transversely to the propagation direction. In the proposed pump-probe framework the first X-ray pulse ionizes molecular deep core-orbital while the following seed pulse stimulates the emission of X-rays. Our theoretical simulations were performed for the sulfur hexafluoride gas medium as a study-case. We show, that in the presence of X-ray stimulated emission between sulfur 2p-1s orbitals, this transition becomes saturated following the nonlinear propagation dynamics, which can be observed as a gradual suppression of spontaneous X-ray fluorescence along the medium. The present theoretical modeling is inspired by the one-dimensional imaging soft X-ray spectrometer, being developed at the SQS end station of the European XFEL, with the capability to spatialy separate emission from microscopic target segments of the nonlinear medium. Our results in turn give a robust feasibility assessment for the future experimental observation.

Nikolai Klimkin, Max Born Institute, Berlin
Coherent control of correlated systems using CEP-stable pulses

Coherent control of quantum systems with phase-stable pulses offers enticing new opportunities for lightwave electronics. Here we extend this approach to correlated systems, demonstrating that a single few-cycle control pulse can create a sizable population asymmetry between the two degenerate polar ground states of the Ising model. This opens a route for femtosecond-scale data processing and storage, allowing one to control the final ground state of a correlated system in an all-optical way.

Nick Lackmann, Max-Planck Institute for Nuclear Physics, Heidelberg
Development of and extreme-ultraviolet beamline for quantum logic spectroscopy of highly charged ions

Atomic clocks based on highly charged ions are prosperous candidates for quantum sensors with unprecedented precision, sensitive for physics beyond the Standard Model [1,2]. To drive the clock transitions, an extreme-ultraviolet frequency comb was constructed based on cavity-enhanced high-harmonic generation of the driving 100MHz near-infrared frequency comb [3]. Harmonics up to 42 eV are generated in a gas jet and are subsequently guided through a beamline towards a superconducting Paul trap for direct XUV-comb spectroscopy [4, 5].

[1] M. G. Kozlov et al., Rev. Mod. Phys. 90, 045005 (2018)
[2] Safronova et al., Phys. Rev. Lett. 113, 030801 (2014)
[3] J. Nauta et al., Opt. Express 29, 2624 - 2636 (2021)
[4] P. Micke et al., Nature 578, 60 - 65 (2020)
[5] J. Stark et al., Rev. Sci. Instr. 92, 083203 (2021)

Dr. Dominik Lentrodt, Albert-Ludwigs-University, Freiburg
X-ray cavity QED with Mössbauer nuclei

In the last decade, Mössbauer nuclei have emerged as a new platform to implement quantum optics with hard x-ray photons. However, current experiments still operate in the linear optics regime and non-linear quantum effects have not yet been observed. In this talk, we will discuss how concepts from cavity QED may enable progress in this direction. First, we provide a theoretical explanation for the so-called collective Lamb shift, which was already observed experimentally in the linear regime a decade ago. We show that the observed shift is a result of multi-mode contributions in the cavity setup. Second, we utilize the developed theoretical tools to make predictions on whether ensemble inversion is already possible at current x-ray facilities. Surprisingly, we find that even for tightly focused pulses, specifically designed cavities can act as intensity boosters providing an enhancement of the nuclear excitation.

Dr. Aliaksandr Leonau, Center for Free-Electron Laser Science (CFEL), DESY
Operator Method of Non-Perturbative Description of Quantum Systems and Its Applications

Investigation of theoretical models in quantum optics and quantum mechanics, which do not contain a small physical parameter, require the non-perturbative approaches to solving the Schrödinger equation. At the same time, approximate analytical results for such systems could be important for checking the validity of the numerical simulation as well as describing the behavior of some physical properties in the limiting cases. In the present talk, we address the operator method [1], which is based on the specific transformation of the perturbation series. This method is promising in providing high accuracy of the zeroth-order analytical approximation and convergence of the successive corrections. As an example of the effectiveness of this method, we apply it to solving the stationary Schrödinger equation for one of the most fundamental models of quantum optics – the quantum Rabi model [2,3]. Another example is related to the analytical model of a many-electron atom (ion) [4,5].
[1] I.D. Feranchuk, A. Ivanov, V.H. Le and A.P. Ulyanenkov, Non-Perturbative Description of Quantum Systems, Cham: Springer, 2015.
[2] I.D. Feranchuk, A.V. Leonov and O.D. Skoromnik, J. Phys. A: Math. Theor. 49, 454001 (2016).
[3] I.D. Feranchuk, N.Q. San, A.U. Leonau and O.D. Skoromnik, Phys. Rev. A. 102, 043702 (2020).
[4] O.D. Skoromnik, I.D. Feranchuk, A.U. Leonau and C.H. Keitel, J. Phys. B: At. Mol. Opt. Phys. 50, 245007 (2017).
[5] K.D. Dzikowski, O.D. Skoromnik, I.D. Feranchuk, N.S. Oreshkina and C.H. Keitel, J. Phys. B: At. Mol. Opt. Phys. 54, 115002 (2021).

Dr. Hao Liang, Max-Planck Institute for the Physics of Complex Systems, Dresden
Time Delays in Resonant Two-Photon Ionization

A decade ago, a theoretical investigation reveal that there is a time delay proportional to pulse duration during the resonant two-photon ionization process. In the present talk, I will extent this concept to two extreme cases, beyond the perturbative region. For the first, if the coupling between initial and resonant states is strong enough to trigger Rabi oscillation, the simple linear law would break down, and Rabi oscillation plays the role. Then we will focus on resonant RABBITT case. A moderate infrared pulse could strongly couple the resonant and continue states. Both pulse envelope, ac-stark shift and decay rate alter the resonant time delay. A RABBITT experiment with large range delay scanning is performed and comparing with the simulation based on time-dependent Schr¨odinger equation and two-level model. In the end, we reconstruct the resonant ionization wavepacket in time domain with both amplitude and phase information.

Dr. Kang Lin, Goethe University, Frankfurt
Nondipole effects in strong-field ionization

Ultrafast lasers are a powerful and versatile tool towards the control of electronic motion, which is one of today's grand challenges of atomic and molecular physics. On this route however, the vast majority of the works so far has neglected the influence of magnetic component of the light field and of its spatial dependence. Including these important effects beyond the so-called electric dipole approximation for understanding light-matter interactions is one of the current frontiers of physics with short laser pulses.
Most recently we found that the energies of photoelectrons emitted against the light propagation direction are shifted toward higher values, while those electrons that are emitted along the light propagation direction are shifted to lower values due to the nondipole contribution (Science Advances 8, eabn7386 (2022)). More interestingly, the nondipole contribution to the envelope of the electron energy spectrum shows an opposite tendency (Phys. Rev. Lett. 128, 023201 (2021)). However, this only holds for low-energy electrons below the classical cutoff. The nondipole shift for high-energy electrons beyond the classical cutoff is found to be vastly different. We show that large-angle rescattering of the electrons strongly alters the partitioning of the photon momentum between electron and ion (Phys. Rev. Lett. 128, 113201 (2022)). As a result, the sensitivity of the nondipole shift can serve as a powerful tool to investigate electron correlation. We also solved a long-standing controversy in double ionization of xenon atoms by utilizing the nondipole effect and reveal that the double ionization proceeds via recollision-induced doubly excited states (Phys. Rev. Lett. 128, 023201 (2022)).

Frieder Lindel, Albert-Ludwigs University, Freiburg
Quantized fields for optimal control in the strong coupling regime

The control of quantum systems lies at the core of many quantum technologies. In the field of coherent control, classical fields coherently drive the quantum system from a given initial state into a target state. Exploiting the quantum nature of the field to improve these control protocols has so far been mostly limited to the weak coupling regime. Here we will discuss how the quantum statistics of a bosonic field can be optimally tailored in order to drive a weakly or (ultra-)strongly coupled quantum system, such as an atom or a molecule in a cavity, towards a desired target state. This extends optimal control theory to control and target systems that are both quantized and strongly coupled.

Arkajyoti Maity, Max-Planck Institute for the Physics of Complex Systems, Dresden
Valley polarization in pristine graphene with linearly polarised laser pulses

Information processing using preferential excitation of one of the two energy degenerate valleys in inversion symmetry broken graphene-like systems has been achieved by circularly polarized pulses. These pulses couple differentially to the valleys which have opposite orbital angular momentum, depending on their polarization[1]. Recent studies have, however, shown that linearly polarised light pulses can generate appreciable valley polarization, even in pristine graphene, without breaking inversion symmetry at the Hamiltonian level[2]. In our presentation, we will shed some light on the general mechanisms of this process of valley polarization with ultrashort laser pulses. We also show results for the terahertz regime, in which graphene shows strong non-linear behavior, and discuss the role of electronic decoherences for such longer pulses.

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[2]Hamed Koochaki Kelardeh, Ulf Saalmann, and Jan M. Rost. Ultrashortlaser-driven dynamics of massless dirac electrons generating valley polarization in graphene. Phys.Rev.Research, 4:L022014, Apr 2022

Kai Müller, Technical University Dresden (TUD)
Atomic Dynamics in Strongly Coupled Multimode Cavities Conditioned on Continuous Measurement

An important challenge in non-Markovian open quantum systems is to understand what information we gain from continuous measurement of an output field. For example, atoms in multimode cavity QED systems provide an exciting platform to study many-body phenomena in regimes where the atoms are strongly coupled amongst themselves and with the cavity, but the strong coupling makes it complicated to infer the conditioned state of the atoms from the output light. In this work we address this problem, describing the reduced atomic state via a conditioned hierarchy of equations of motion, which provides an exact conditioned reduced description under monitoring (and continuous feedback). We utilize this formalism to study how different monitoring for modes of a multimode cavity affects our information gain for an atomic state, and to improve spin squeezing via measurement and feedback in a strong coupling regime. This work opens opportunities to understand continuous monitoring of non-Markovian open quantum systems, both on a practical and fundamental level.

Dr. Saikat Nandi, Institut Lumière Matière, CNRS
Coherent light-matter interaction at XUV wavelength using a seeded FEL

Rabi oscillations, a prominent feature of coherent light-matter interaction, arise when a two-level system interacts periodically with external radiation. Despite being a cornerstone in quantum physics, they are usually studied in the long-wavelength region, from a few microns to a few tenths of a micron. Here, we show that intense femtosecond extreme-ultraviolet (XUV) pulses from FERMI seeded free-electron laser (FEL) [1] can drive Rabi oscillations between the two levels: $1s^2$ and 1s4p in helium atoms. The resulting photoelectron spectra show a symmetric doublet structure, identified to be an Autler-Townes (AT) splitting. The experimentally measured period of Rabi oscillations (52 fs) falls within 20% of the FEL-pulse duration, signifying a near single-cycle build-up of the AT-doublet. By tuning the photon energy of the XUV pulse across the resonance, we could observe an avoided crossing [2] in the corresponding photon energy diagram that appeared to be blue-shifted by 11 meV from the resonant transition. In addition, the photoelectron spectra become highly asymmetric as a function of the photon energy as one moves away from the transition [3]. To understand the origin of the blue shift as well as the asymmetry, we developed an analytical model by going beyond the usual strong-field approximation. With this model, we could show that a unique mechanism in the form of a giant Coulomb-induced wave from the ground state allows the non-resonant two-photon process to compete with the resonant one-photon process at the high intensities provided by the XUV-FEL beam [3]. Our results pave the way to study coherent dynamics in matter from a few- to sub-nanometer wavelength region.

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[2] C. Cohen-Tannoudji, Amazing Light, edited by R. Y. Chiao, Chapter 11 (Springer) (1996)
[3] S. Nandi et al., Nature 608, 488 (2022)

Dr. Francisco Navarrete, University of Rostock
Long-range order effects on the HHG spectra from solids

The mechanism of high-harmonic generation (HHG) in solids has been theoretically studied over the last two decades, and experimentally verified a decade ago [1]. While many conclusions have been drawn for this process in periodic crystals, it has also been predicted a strong dependence of the HHG spectrum on the topology of the sample [2]. Recently, by the study of a Fibonacci chain, it has been demonstrated that quasicrystals might constitute excellent materials for HHG due to their higher yield [3], when compared with crystals of the same composition. In this contribution, we will exhibit the crystal momentum resolved [4] numerically obtained spectra [5] for three different lattices: a pristine, a Fibonacci quasicrystal, and a random chain, which allows us to explore long-range order effects. We will describe the yield enhancement as well as the parity of the HHG spectrum in each material.

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[2] C. Jürß and D. Bauer, Phys. Rev. B 99, 195428 (2019)
[3] J.Q. Liu and X.B. Bian, Phys. Rev. Lett 127 213901 (2021)
[4] F. Navarrete et. al., Phys. Rev. A 100 033405
[5] F. Navarrete and D. Bauer (2022) (in preparation)

Dr. Chritian Ott, Max-Planck Institut for Nuclear Physics, Heidelberg

Differential measurement of electron ejection after two-photon two-electron excitation of helium

We report about a fundamental process of nonlinear light-matter interaction in AMO physics: the angular-differential measurement of two-photon single ionization of helium involving the excitation of a correlated two-electron excited state. For this we employ a new experimental methodology, combining the Reaction Microscope at Beamline FL26 at the FLASH Free-Electron-Laser (FEL) with the shot-by-shot detection of each FEL spectral intensity distribution. In a similar way as spectral-domain ghost imaging methods, we apply this new method here to a fundamental nonlinear two-photon-two-electron resonant excitation. As a result, the spectral resolution is significantly improved, allowing us to isolate the resonant transition. Our benchmark results are compared to state-of-the-art multichannel quantum defect theory, which are in good agreement. One possible explanation for remaining deviations is the coherent interaction between fundamental (omega) and second harmonic (two-omega) radiation, which creates an exciting prospect for the future exploration of novel coherent-control schemes with SASE FEL pulses.

Dr. Anton Peshkov, Physikalisch-Technische Bundesanstalt (PTB)

Interaction of twisted light with a trapped atom

Twisted light modes with orbital angular momentum (OAM) show great promise for applications in atomic clocks since excitation of a trapped atom in their low-intensity center can result in significant suppression of an undesirable light shift. In such experiments, however, an accurate description of induced Rabi oscillations is complicated by the transverse atomic motion within the strongly inhomogeneous optical field of twisted light. Here, we present a theoretical model to describe the time evolution of a single atom in a twisted Laguerre-Gaussian beam, taking into account vibrational states of the atom's center-of-mass motion in a harmonic potential created by a trap. Calculations have been performed for the $4s_{1/2} \rightarrow 3d_{5/2}$ electric quadrupole (E2) transition in Ca^+ ion. An analysis based on the density matrix formalism and the Liouville-von Neumann equation shows that the atom may undergo unconventional anharmonic Rabi oscillations that are attributed to the strong coupling between vibrational levels. This effect is accompanied by the angular momentum transfer from twisted light to the atomic center-of-mass motion and becomes most pronounced when the Rabi frequency is comparable to the trapping one.

Prof. Thomas Pfeifer, Max-Planck-Gesellschaft

Laser control of electrons: From pairs in atoms to crowds in molecules

Intense laser pulses can strongly modify the internal electronic structure of matter. Acting on very short time scales of few femtoseconds down to attoseconds, they outrun other processes such as electronic or internuclear relaxation that pull the system back to it's ground state. This ultrashort window of coherence can thus be used to induce, characterize, and modify novel, transient states of matter.

Here, we discuss a selection of our experimental results on the modification of matter on short time scales. Experimentally, in these activities we employ the combination of high-frequency light with intense infrared (IR) laser fields. This allows to observe electronic dynamics in atoms and molecules of increasing complexity after extreme-ultraviolet (XUV) excitation in a state-selective manner.

For two interacting electrons in helium atom, we uncovered that ultrashort laser induced quantum-level shifts right after pulsed excitation lead to phase shifts of coherently excited quantum states. The latter leave their imprint in the spectral line shape of an absorption spectrum, recorded after passage of light through an absorbing medium.

Moving on to multiple electrons in recent work, we focus on another fundamental quantum-mechanical effect, namely exchange interaction. It arises due to the fact that only one electron (due to its Fermionic nature) can occupy a certain quantum state, thus modifying the energy of multi-electron states depending on their spin configuration. Among many different realizations and applications in various fields of physics and chemistry, electronic exchange interaction plays a major role in molecules and their bonding dynamics. In our experiments, employing a few-cycle infrared(IR)-tunable laser system near 1.5 micron wavelength, we found that the relative line strength of a core-level resonance absorption doublet in the sulfur hexafluoride (SF6) molecule is modified by its interaction with the intense laser fields.

It turns out that this effect of line-strength variation can be understood within a model Hamiltonian by the mixing of two effective states exhibiting different magnitudes of exchange interaction. The laser intensity can thus be used as a control knob to tune the effective exchange interaction. A state-of-the-art simulation of the group of Maurits Haverkort (Heidelberg University) confirms the results of our measurement and model description of the general physical mechanism.

These results of laser steering the fundamental quantum dynamics of two, or even multiple, electrons—even for the complex case of poly-atomic molecules—open exciting future perspectives. They lay the foundation for controlling molecular dynamics and chemical reactions directly on the electronic level.

Prof. Maria Novella Piancastelli, Sorbonne University, LCPMR

Isotopic effects in multiphoton water and semi-deuterated water core ionization

The ultrafast structural dynamics of water following inner-shell ionization are a crucial issue in high-energy radiation chemistry in liquid media. We have exposed isolated water molecules to a short x-ray pulse from a free-electron laser, the EuXFEL in Hamburg, Germany, and detected momenta of all produced ions in coincidence. We have studied the series H2O-HDO-D2O. By combining experimental results and theoretical modelling, we can image dissociation dynamics of individual molecules in unprecedented detail. We reveal and compare significant molecular structural dynamics in H2O2+ and HDO2+, such as bond-angle opening, leading to two-body or three-body fragmentation on a timescale of a few femtoseconds. Furthermore, by exploiting isotopic effects, a deep insight into the fragmentation processes in HDO is achieved.

Prof. Thomas Pohl, Aarhus University Department of Physics and Astronomy Denmark
Crystallisation by heating

In this talk, we will consider the behaviour of atomic quantum gases with dipolar interactions. In particular, I will discuss theoretical and experimental evidence that such a system can form a supersolid state upon raising its temperature.

Shreyas Ramakrishna, Helmholtz Institute, Jena
Atomic magnetometer based on twisted light-atom interaction

We analyze the photoexcitation of atoms by azimuthally polarized Bessel beams in the framework of density matrix theory and based on the Liouville–von Neumann equation. In particular, we study the dependence of the population of excited atomic states on the oscillation of the external magnetic field for the case of the ($5s\ ^2S_{1/2}$ (F=1) $\rightarrow$ $5p\ ^2P_{3/2}$ (F=0)) electric dipole transition in ^{87}Rb atoms. We demonstrate that the population of the excited atomic state is very sensitive to the time period of the oscillating magnetic field, the position of atoms in the beam cross-section and the projection of orbital angular momentum of the incoming light beam. Furthermore, we study the dynamics of the excited atomic state population due to the effect of the time period of the oscillating magnetic field before and after the system has reached a steady state. The results of our calculation can have an impact on metrological experiments based on vortex light beams.

Dr. Laura Rego, Imperial College London
Application of structured ultrashort laser pulses to the study of chirality

Distinguishing between the left- and right-handed versions of a chiral molecule (enantiomers) is vital, but also inherently difficult. Traditional optical methods using elliptically/circularly polarized light rely on linear effects which arise beyond the electric-dipole approximation, posing major limitations for ultrafast spectroscopy. Here we show how chiral discrimination can benefit from the use of ultrashort laser pulses with structured polarization. In particular, we show how to turn an ultrashort elliptical pulse into an efficient chiro-optical tool: by tilting its polarization plane towards its propagation direction [1]. This forward tilt is achieved by focusing the beam tightly, creating structured light. Our field realizes an efficient interferometer that separates the nonlinear optical response of opposite enantiomers in space. This work provides a simple way of spatially structuring the polarization of light to image molecular chirality, with extreme enantio-efficiency and on ultrafast timescales.

[1] L. Rego, O. Smirnova, and D. Ayuso. Tilting light's polarization plane to spatially separate the ultrafast nonlinear response of chiral molecules. Under review (2022).

Jan Richter, Physikalisch-Technische Bundesanstalt (PTB)
Parity violation in highly charged ions - a proposal for the Gamma Factory

Atomic parity nonconservation phenomena arising due to the weak interaction of atomic electrons with nuclei have been in the focus of experimental and theoretical research for several decades. In this study, the focus lies on the influence of the mixing of opposite-parity ionic levels on the photoexcitation rates in highly charged ions. This mixture arises due to an external electric field and the weak interaction between electrons and the nucleus. In order to reinvestigate this "Stark-plus-weak-interaction" mixing, detailed calculations are performed in hydrogen- and lithium-like ions. The photoexcitation in such highly charged ions can be realized at the Gamma Factory at CERN. The basic idea of the Gamma Factory is to make use of the Doppler effect in a head-to-head collision between a laser beam and a beam of relativistic ions. Due to the relativistic motion of the ions, the photon frequency is boosted in the ion rest frame. This enables photoexcitation in ions with very high transition energies.

Prof. Nina Rohringer, DESY-CFEL

Controlling x-ray matter interaction by collision-induced changes of electronic populations

Irradiation of solids with focused soft x-ray FEL pulses transforms constituent atoms into an electronically highly excited, transient state that on an ultrashort time scale transforms to an equilibrium state of warm dense matter. In these regimes, new bound-bound electron transitions open – thus dramatically modifying the absorption as well as scattering properties of the solids. I present a combined experimental and theoretical study on characterising the transient state of warm dense copper by x-ray absorption and scattering, based on two experiments at the European XFEL's SCS instrument.

In the experiment, the XFEL pulse with a photon energy around 933 eV (Cu L-edge) is focused on a thin Cu foil, transforming Cu quasi instantaneously into an electronically excited state: L-shell photoionization and subsequent Auger-decay creates an out-of-equilibrium population of continuum electronic states. Electron-electron and electron-ion/atom collisions then drive the system to an equilibrium state of warm dense matter on a 10-100 fs timescale. We probe this transient state by spectrally resolving the transmitted x-ray pulse as a function of fluence, thereby revealing the properties of the atomic and ionic cores. For XFEL pulse intensities above $10^{13} W/cm^2$ W/cm^2 we observe a strong pre L-edge peak appears in the absorption spectrum which is due to 2p3/2 $\rightarrow$ 3d transitions. These absorption channels open due to holes in the Cu 3d shell that are produced by electron-impact ionization. With increasing intensity the observe absorption peak shifts towards lower energies, pointing to the effect of ionization potential lowering in the out-of equilibrium plasma. At intensities above $10^{-}15 W/cm^2$, the pre-edge peak broadens, drops in intensity, and the spectral absorption profile becomes smooth without a pronounced L–absorption edge. These features are consistent with the appearance of multiple higher ionic charge states in different electronic configurations, as our modeling based on the kinetic Boltzmann approach demonstrates.

In a recent second experiment, the x-ray scattering properties of the transient state have been studied, by spectrally resolving the diffracted x-ray radiation: We investigated the diffracted spectral intensity of the 5-th order superlattice peak of a [B_4C(2nm)/Cu(2nm)/SiC(2nm)]15 multilayer sample. As expected from the fundamental relation between scattering and absorption, the opening of absorption channels on 2p3/2 $\rightarrow$ 3d transitions likewise results in additional resonant elastic scattering contributions. In line with our first experiment, we observed a strong enhancement of the diffracted intensity in the pre-edge spectral region. This demonstration paves the way towards control of atomic scattering properties.

Daniele Ronchetti, Max Planck School of Photonics / DESY

Enhancing soft x-ray diffraction by collision-induced manipulation of electronic populations

The advent of X-ray Free Electron Lasers (XFELs) opened the way for ways of exploring and controlling non-linear and collective emission phenomena. Exploiting the extreme intensity of XFEL and building upon well-established techniques - such as x-ray lasing and x-ray superfluorescence - we aim at controlling the scattering properties of individual atoms. The process starts with irradiation by an XFEL pump pulse that creates core transient resonances in a target atom via photoionization. A second probe pulse, whose energy is resonant with one of the opened inner-shell transitions, can scatter, experiencing an enhanced atomic scattering factor.

In this talk, I will present the results of a recent experiment – performed at the SCS beamline of EuXFEL - in which we measured with spectral resolution the enhancement of the scattering response of copper atoms subject to XFEL pulses. We demonstrated that the enhancement strongly depends on the intensity of the pump pulse and can grow up to one order of magnitude. Our findings encourage the application of the effect in innovative crystallographic methods where 3d metals may act as heavy scatterers.

Emanuele Rossi, DESY-CFEL

Theoretical study of the preparation of coherent wave-packets launched by attosecond XFEL pulses

The electronic motion at its natural, attosecond time-scale can be studied by means of attosecond light pulses [1]. Attosecond electron dynamics influence longer-time scale photochemical processes such as photosynthesis, photocatalysis and light harvesting [2]. The control of the characteristics of the pulses makes it possible to manipulate the dynamics of the electrons after their photoexcitation, thus allowing the modulation of the photochemical reactivity [3]. The introduction of attosecond pulses at X-Ray Free Electron Lasers (XFEL) allows the study of ultrafast electron dynamics with elemental and spatial resolution by means of non-linear X-Ray techniques. A candidate nonlinear technique is Stimulated Resonant Inelastic X-Ray Scattering (SRIXS), which can launch a coherent, vibronic wave-packet (WP) of valence excited states on a specific element of a molecule [4]. In my contribution I'll present a model for the preparation of a SRIXS WP based on highly accurate, coupled cluster calculations. The properties of the launched WP and their dependence on the characteristics of the XFEL pulse will be discussed, together with an outlook for further theoretical and experimental developments.

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[2] arXiv:2207.05892
[3] Saalfrank P. et al, Advances in Quantum Chemistry, 2020, 81, 15-50
[4] Yong H. et al, J. Phys. Chem. Lett. 2021, 12, 40, 9800-9806

Prof. Krzysztof Sacha, Jagiellonian University, Kraków

Do absolutely stable discrete time crystals exist?

Discrete time crystals are closed periodically driven many-body systems that spontaneously evolve with the period longer than the driving period. They are supposed not to heat up despite the fact that generic periodically driven many-body systems are expected to evolve to infinite temperature structureless state. We will discuss the absolute stability of discrete time crystals and provide solution for this problem.

Sharath Sasikumar, European XFEL

FEL studies of complex metal nanoparticles and their structural dynamics

The study of isolated nanoparticles produced in the gas phase is of fundamental importance for understanding how properties of matter evolve from atomic and molecular features to those typical of bulk materials. Our work uses a Pulsed Microplasma Cluster Source (PMCS) for controlled gas phase synthesis of transition metal nanoparticle aggregates. With it we experimentally address in the time domain the coalescence process in complex aggregates, which is relevant for the understanding and control of gas-phase synthesis methods when size, stoichiometry, structure and morphology are of concern. Our approach is based on ion-electron spectrometry upon laser ionization by which we characterize the size and structure of the isolated nanoparticles; on a second instance, sub-ps pump-probe electron spectroscopy and scattering experiments at FELs will allow us to study the quenching of non-equilibrium nanoparticles and compare their observed structural relaxation to existing models for coalescence dynamics.

Riaan Philipp Schmidt, Physikalisch-Technische Bundesanstalt (PTB)
A Study of Twisted Light with the Hanle Effect

We analyze the depolarization of resonance fluorescence from atoms exposed to twisted (Bessel) radiation. Special attention has been paid to the dependence on the external magnetic field strength, known as the Hanle effect, which is investigated within the framework of the density matrix theory based on the Liouville–von Neumann equation. While the derived expressions can be employed to study the Hanle effect for any atomic system, detailed calculations of the P_1 Stokes parameter of the emitted radiation have been performed for the $5s\,^2S_{1/2} - 5p\,^2P_{3/2}$ transition in rubidium. Our results indicate how the fluorescence depolarization may be affected by the spatial structure and polarization of the incident light field, as well as by the applied magnetic field. This study contributes to a better understanding of the potential of twisted light in atomic spectroscopy.

Björn Senfftleben, European XFEL
Ultrafast electron dynamics in helium nanodroplets reaching into the attosecond domain

We analyze the depolarization of resonance fluorescence from atoms exposed to twisted (Bessel) radiation. Special attention has been paid to the dependence on the external magnetic field strength, known as the Hanle effect, which is investigated within the framework of the density matrix theory based on the Liouville–von Neumann equation. While the derived expressions can be employed to study the Hanle effect for any atomic system, detailed calculations of the P_1 Stokes parameter of the emitted radiation have been performed for the $5s\,^2S_{1/2} - 5p\,^2P_{3/2}$ transition in rubidium. Our results indicate how the fluorescence depolarization may be affected by the spatial structure and polarization of the incident light field, as well as by the applied magnetic field. This study contributes to a better understanding of the potential of twisted light in atomic spectroscopy.

Romain Soguel, HI Jena + FSU Jena
Valence-hole excitation in closed-shell system: QED treatment

An ab initio QED approach to treat a valence-hole excitation in closed-shell systems is developed in the framework of the two-time Green's-function method. The derivation considers a redefinition of the vacuum state and its excitation as a valence-hole pair. The proper two-time Green's function, whose spectral representation confirms the poles at valence-hole excitation energies, is proposed. An contour integral formula which connects the energy corrections and the Green's function is also presented. First-order corrections to the valence-hole excitation energy involving self-energy, vacuum polarization, and one-photon-exchange terms are explicitly derived in the redefined vacuum picture. Reduction to the usual vacuum electron propagators is shown, which agrees in the Breit approximation with the many-body perturbation theory expressions for the valence-hole excitation energy.

Jonas Sommerfeldt, Physikalisch-Technische Bundesanstalt (PTB), Braunschweig
Coulomb Corrections to Delbrück Scattering

Delbrück scattering is the process in which a photon is elastically scattered by the Coulomb field of a nucleus via the production of a virtual electron-positron pair. It is one of the few non-linear QED processes that can be observed experimentally [1] and, hence, testing the respective theoretical predictions serves as an important test for the standard model. Despite the strong motivation for the theoretical analysis of Delbrück scattering, most of the previous studies have been limited to some approximation regarding the coupling between the virtual electron-positron pair and the nucleus leading to large disagreements between theory and experiment for certain parameter regimes [2]. In this contribution, therefore, we present an efficient approach to calculate amplitudes for Delbrück scattering that accounts for the interaction with nucleus to all orders including the Coulomb corrections [3]. We apply our method to photon energies below and above the pair production threshold.

[1] M. Schumacher, Radiat. Phys. Chem. 56 (1999) 101-111
[2] P. Rullhusen et al., Z Physik A 293 (1979) 287–292
[3] J. Sommerfeldt et al., Phys. Rev. A 105 (2022) 02280

Sophia Strnat, Physikalisch Technische Bundesanstalt (PTB)
Elastic scattering of hard x-rays by atomic targets

The elastic scattering of x-rays by atoms is one of the fundamental processes in the interaction of radiation with matter. Over the past few years, increasing attention has been paid to the study of the properties of scattered photons. In our contribution, we study the polarization properties of elastically scattered hard x-rays by highly charged ions and neutral atoms. In particular, we explore how the interference between Rayleigh and Delbrück scattering may affect the cross section and the polarization properties of the outgoing radiation.

Vladislav Sukharnikov, DESY-CFEL
X-ray lasing in neon within the stochastic formalism

X-ray lasing experiments demonstrated the possibility of generating bright x-ray pulses with a narrow spectral bandwidth. In a typical experimental setup, a sample is pumped by an incoming XFEL pulse tuned above the ionization edge. Emergent inner-shell excited atoms predominantly decay through the non-radiative Auger effect. However, when the atomic density and population inversion are high enough, the atoms become synchronized and emit coherently by means of superfluorescence.

Before the buildup of macroscopic polarization, the quantum effects play a crucial role. The full quantum-mechanical treatment is not feasible since the number of equations scales exponentially with the number of particles. There are phenomenological ways to extend semi-classical equations to simulate the spontaneous emission which triggers the amplification. In contrast, we build our theory from the first principles and arrive at the set of stochastic differential equations with stochasticity restoring all the quantum effects. We apply our technique to simulate x-ray lasing in neon. In the talk, we show how the simulations can predict spectral features by comparing the numerical and experimental results.

Dr. Sebastian Ulbricht, Physikalisch-Technische Bundesanstalt (PTB)
Signatures of gravity in the transmission spectrum of an optical cavity

Modern technical devices, such as atomic clocks or gravitational wave detectors, can rely on high-finesse Fabry-Pérot cavities for laser frequency stabilization. In most of all applications, however, such a cavity-laser setup is located on our planet's surface. As we know from the theory of general relativity, any kind of energy density is subject to gravity. Here, also the energy density of a propagating light wave in a cavity-laser setup poses no exception.
In this talk, we investigate how gravity affects the transmission spectrum of an Earth-based Fabry-Pérot cavity near the frequency resonance and propose a measurement scheme that allows for practical analysis of the cavity internal light bending effect and technical applications.

Lucas Weitzel Dutra Soulo, Albert-Ludwigs University, Freiburg
Effective mode theory for open quantum systems

In the frequently employed theoretical approaches for open resonator QED systems, one usually considers a quantum emitter - such as an atom - in a cavity which interacts with a few discrete electromagnetic cavity modes. Losses and leakage from the cavity are modelled via a (weak) interaction with the environment. Models that employ this treatment, as is the case for the seminal Jaynes-Cummings model and its generalizations, have been tremendously successful in describing experiments. However, these models are intrinsically phenomenological and it is not known if they hold in all situations, as the underlying approximations are still unclear. For instance, in the case of strongly leaking systems, such as plasmonic cavities, this approach is not valid anymore and various theoretical assumptions need to be reassessed. We hence try to answer the following question: Is it possible to construct from first principles a few-mode description for leaky cavities in the spirit of the Jaynes-Cummings model? This will extend our theoretical understanding of more general systems or, in the case of a negative answer, lead to a no-go theorem.

Prof. Matthias Wollenhaupt, Carl von Ossietzky University, Oldenburg
Molecular electron vortices

Since their theoretical proposal [1] and their first experimental demonstration [2], free electron vortices have attracted significant attention. So far, most of the theoretical and all of the experimental investigations were performed on atoms. Here, we present the first experimental demonstration of free electron vortices by multiphoton ionization (MPI) of molecules. Specifically, we study the creation of molecular vortices on C60 fullerenes using counter rotating circularly polarized femtosecond laser pulse sequences generated from a white-light supercontinuum. Since the discovery of the C60 molecule it has served as a benchmark system to study photo-induced dynamics in complex systems. Due to its high symmetry, the C60 molecule, is an ideal system to bridge the gap between atoms and more complex systems such as polyatomic molecules and clusters. It has been shown that C60 exhibits distinct atom-like electronic orbitals, termed superatomic molecular orbitals (SAMOS) [3], which play an important role in the MPI of fullerenes. By tomographic reconstruction of the three-dimensional photoelectron momentum distribution, we show that ionization from a SAMO with the polarization-tailored laser field creates a six-armed free electron vortex.

[1] J. M. Ngoko Djiokap et. al, Phys. Rev. Lett., 115(11), 2015
[2] D. Pengel et. al, Phys. Rev. Lett., 118(5), 2017
[3] M. Feng et. al, Science, 320(5874), 2008

Dr. Zhongwen Wu, Helmholtz Institute, Jena
Angular distribution of Auger electrons following electron-impact excitation of Be-like ions

Electron-impact excitation of atoms or ions is one of fundamental processes in astrophysical and laboratory plasmas. Chen and Reed [Phys. Rev. A **50**, 2279 (1994)] studied the relativistic effect on the angular distribution of Auger electrons following electron-impact excitation of Be-like ions. However, as a main part of the relativistic effect, the contribution of the Breit interaction to the angular distribution was not considered. In the present contribution [Phys. Rev. A **105**, 032809 (2022)], we revisited the angular distribution of the Auger electrons emitted from the nonradiative decay $1s2s^22p_{1/2}\,J=1 \rightarrow 1s^22s\,J=1/2$ of Be-like Mg^{8+}, Fe^{22+}, Mo^{38+}, Nd^{56+}, Au^{75+}, and U^{88+} ions following the excitation $1s \rightarrow 2p$ of these ions using the multiconfigurational Dirac-Fock method and the relativistic distorted-wave theory. Special attention was paid to the effect of the Breit interaction on the angular distribution of the Auger electrons. It was found that for low-Z ions the Breit interaction hardly contributes to the angular distribution, especially at low impact energies, whereas for medium- and high-Z ions the contribution of the Breit interaction is of great essence, which becomes more and more pronounced with increasing impact energy.