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2.1 HIGHEST Precision

Ion Traps

Ions can be stored in traps by the superposition of electric and magnetic fields in an extreme vacuum. Penning traps allow storage of a single ion that performs a characteristic oscillating motion in the trap. The ion's mass and further properties like magnetic moments of the bound electron in highly charged ions can be deduced from the motional frequencies if the charge state and the magnetic field strength are known, even in the case of exotic particles that live only for a few milliseconds. Penning-trap mass spectrometers are operated at MPIK and at radioactive beam facilities like GSI and CERN.

In an electron-beam ion trap (EBIT), highly charged ions are produced by impact of energetic electrons, then spatially confined, and electronically heated up to temperatures of millions of degrees. Both, stationary and mobile EBITs are used to prepare and study atomic matter under extreme conditions. One of the highlights of the latest EBIT developments at MPIK is the Tip-EBIT at the experiment PENTATRAP where laser desorption and subsequent ionization is applied. It extends the range of available HCIs to rare isotopes, which are synthesized in only sub-nanogram quantities. A suite of accurate spectroscopic instrumentation attached to the EBITs collects precise data. A new cryogenic ion trap (Cryogenic Paul Trap Experiment: CryPTEx) has been built at MPIK in cooperation with the university of Aarhus, in which ion crystals can be produced by means of laser cooling, and highly charged ions cooled therein.



A newly developed miniature EBIT.

Nuclei – From the Building Blocks of Matter to the Formation of Elements

Penning-trap mass spectrometry at MPIK allowed recently to perform world-record measurements on the atomic masses of the electron and proton, i. e., the simplest nucleus. The proton mass was found to be smaller than the previously accepted value. This helped to understand observed discrepancies in the masses of light nuclei.

Looking at heavier elements, the chemical composition of our Universe shows some surprising peculiarities: The Sun mainly consists of hydrogen and helium; iron is much more



Chart of nuclides with the colour code showing the binding energy per nucleon: the most stable nuclides around iron are in dark blue.

abundant on Earth compared to heavy elements like gold. Nucleosynthesis follows reaction paths involving fusion and capture processes, some of them yet mostly unexplained. Since nuclear fusion stops at iron, heavier elements are generated via proton or neutron capture under extreme conditions like in supernova explosions of stars or in hot environments like accretion discs around Black Holes or neutron stars.

Based on Einstein's principle of mass-energy equivalence, high-precision mass measurements are used to determine nuclear binding energies which are crucial for reaction pathways in nucleosynthesis. In combination with theoretical models, the structure of nuclei even far from stability can be investigated. Mass measurements on these mostly short-lived exotic (e.g. neutron-rich) nuclei are used to explore the "terra incognita" on the chart of nuclides. This helps to figure out how many nuclides exist at all.

The atomic mass of the proton: 3 times more precise and 3σ smaller than expected

The electron, proton and neutron are the basic building blocks of the atomic structure of nature. The precise knowledge of their intrinsic properties, e. g., their rest mass, is essential for fundamental tests of quantum electrodynamics and the determination of fundamental constants like the Rydberg and the fine-structure constant. Until now, a combination of the mass measurements on light ions (proton, deuteron, helium-3 and the molecule HD) show a 5 σ discrepancy and thus question these measured values. In a new Penning-trap setup, the LIONTRAP (Light ION Trap) experiment, these masses will be determined in a new approach in atomic mass units and with unrivalled precision. Here, we compare the cyclotron frequency of a single trapped carbon ion to the one of a single trapped ion of interest. Alternately, both frequencies are measured in a non-destructive way in a newly designed seven-electrode cylindrical Penning trap.



In the first measurement campaign, the focus was on the proton mass. The strongly differing charge-to-mass ratios of a proton and a carbon nucleus require extraordinarily precise knowledge of the systematic shifts. Great effort has been invested to trap both ions subsequently at the same position by applying the same trapping potential and using two independent but finetuned cryogenic ultra-low noise axial detection systems. For the first time, a phase-sensitive measurement of the cyclotron frequency of the proton has been successfully implemented, which was made possible by the exceptionally good harmonicity of the electrostatic trap potential. With a relative uncertainty of 3×10^{-11} the new proton mass is a factor of 3 more precise than the previous literature value. Although our measured value is 3σ smaller than expected, this is not sufficient alone to solve the mentioned light ion mass puzzle, which is a strong motivation for further investigations on the other light ion masses.

References:

[1] F. Heiße et al., Phys. Rev. Lett. 119, 033001 (2017), DOI: 10.1103/PhysRevLett.119.033001

[2] F. Heiße et al., Phys. Rev. A 100, 022518 (2019), DOI: 10.1103/PhysRevA.100.022518

[3] S. Sturm et al., Eur. Phys. J. Special Topics 227, 1425{1491 (2018), DOI: 10.1140/epjst/e2018-800225-2

High-precision mass measurements of short-lived, neutron-rich copper and chromium isotopes

The precise mass determination of radioactive atoms is key to further understanding of the nucleus. Far from nuclear stability, the highly unbalanced proton-to-neutron ratio reveals new nuclear phenomena and allows for nuclear models to be tested.

Hence, in a recent study using the high-precision mass spectrometer ISOLTRAP at ISOLDE/CERN, the masses of neutron-rich copper isotopes ⁷⁵⁻⁷⁹Cu were measured [1]. With ⁷⁹Cu located only one proton above the flagship nucleus ⁷⁸Ni, these measurements offer a first accurate view on the doubly closed-shell nature of ⁷⁸Ni. Furthermore, the mass values allowed for refined simulations (see figure) of the crust composition of neutron stars, still containing bound atoms in the first hundreds of meters of their outer crust. The knowledge of their composition is important for the element abundance in the Universe as neutron-star and neutron-star-black-hole mergers are possible sites for the rapid neutron-capture process, being responsible for the creation of approximately half of the atomic nuclei heavier than iron.

Besides revealing a smooth development of nuclear groundstate collectivity towards N = 40, the neutron-rich chromium isotopes ⁵⁸⁻⁶³Cr measured in another experimental campaign [2] are also known to play an important role in the cooling and heating of the lower layers of the crust of accreted neutron stars, possibly impacting the associated astrophysical observables.



Simulated neutron-star-crust profile with its isotope content. S. Gorielli, private communication (2017).

References:

[1] A. Welker et al. Phys. Rev. Lett. 119, 192502 (2017), DOI: 10.1103/PhysRevLett.119.192502
 [2] M. Mougeot et al. Phys. Rev. Lett. 120, 232501 (2018), DOI: 10.1103/PhysRevLett.120.232501

Highly Charged Ions – Matter under Extreme Conditions

Highly charged ions (HCIs) are found in hot environments of more than one million degrees such as stellar atmospheres and cores, supernova remnants or accretion discs around neutron stars and black holes. In fact, most of the visible matter in the Universe is assumed to be highly ionized. Analysis of the observed light (visible, UV, or X-ray) from these ions needs support by theoretical structure calculations which are often not accurate enough to determine, e.g., the temperature of the hot environment. The controlled production of highly charged ions in an EBIT combined with high-precision spectroscopy provides direct

experimental information. One example is the investigation of the X-ray absorption of highly charged iron ions at the synchrotron PETRA III (DESY) which provided important new insight into the radiation transport within stars.

The cryogenic ion trap CryPTEx provides efficient cooling of trapped HCIs for high-precision laser spectroscopy. In collaboration with the PTB (Braunschweig), the MPIK contributes to the development of novel optical clocks using quantum logic spectroscopy and built a VUV/XUV frequency comb for precision spectroscopy. The ultimate goal will be to test the time dependence of natural constants. With PENTATRAP, for the first time very long-lived metastable electron configurations have recently been discovered in highly charged ions of heavy metals. This technique has the potential to become the method of choice for the search for metastable electron configurations suitable to HCI clocks. At ALPHATRAP, the ground-state *g*-factors of highly charged ions can be measured with fractional uncertainties at the 10^{-9} level. First results are in excellent agreement with state-of-the-art QED calculations.



Spectrum of iron ions which determine the radiation transport within the Sun.

First results of ALPHATRAP: the magnetic moment and fine structure of boronlike ⁴⁰Ar¹³⁺



Highly charged ions (HCI) are excellent systems for testing the validity of quantum electrodynamics (QED) in strong fields. Those ions allow for precise calculations and the extremely high electric field strength in heavy HCI provides a unique sensitivity to possible deviations from the Standard Model of physics. The recently developed ALPHATRAP experiment includes a room-temperature electron beam ion trap (EBIT) for medium-Z ions and a connection to the Heidelberg high-energy EBIT (HD-EBIT) for high-Z HCIs. The versatile double Penning-trap system can capture and trap HCIs of any charge state and mass. Ions are injected into the cryogenic trap system inside a superconducting magnet, where they can be stored for several months due to the extremely high-vacuum conditions of <10⁻¹⁷ mbar. Apart from the highly homogeneous trap that is used for high-precision spectroscopy, another one is used for spin-state detection via the continuous Stern-Gerlach effect (CSGE). Performing unambiguous spin-state detection was the basis for precise measurements of the ion's magnetic moment (q-factor). The first measurement was of the g-factor of boronlike ⁴⁰Ar¹³⁺ with 9-digits precision. We have also performed ab initio calculations of the g-factor, taking into account electron correlation and QED effects. By significantly improving the latter, we arrive to a relative theoretical uncertainty of 9×10^{-7} . The perfect agreement between theory and experiment paves the way toward an independent determination of the fine-structure constant. Additionally, we have shown that the CSGE can be used instead of fluorescence detection, to find and measure optical transitions. For this, laser spectroscopy of the fine-structure transition in the ground state of ⁴⁰Ar¹³⁺ was performed. These successful campaigns have demonstrated the potential of ALPHATRAP, which will allow to further explore the boundary of validity of strong-field QED.

References:

[1] I. Arapoglou et al., Phys. Rev. Lett. 122, 253001 (2019), DOI: 10.1103/PhysRevLett.122.253001
 [2] A. Egl et al., Phys. Rev. Lett. 123, 123001 (2019), DOI: 10.1103/PhysRevLett.123.123001



Towards an orbital-crossing clock to test fundamental constants

Optical frequencies in atoms and ions called 'clock transitions' are the most accurately measured quantities in science, and as stable as fundamental constants; 'varying' clocks would challenge the Standard Model of fundamental interactions. Since atomic properties limit the reach of ongoing searches, we look for maximally sensitive optical transitions. Rare 'orbital crossings' present in highly charged ions (HCI) magnify the effect of a 'varying' finestructure constant α , the best known one, by strongly shifting the initial and final states in the transition [1]. We found in the Pr⁹⁺ ion the second such crossing, and measured for the first time all spectroscopic levels of its potential clock transition, which has high sensitivity to new physics and the smallest predicted effects of external perturbations [2]. Our large-scale relativistic atomic structure calculations allowed an unambiguously identification of this transition. Its lifetime of about 8 years ensures ultimate clock stability. Since we also newly demonstrated how to sympathetically cool HCI for use in clocks [3], our experiment amply extends the frontiers of such searches for physics beyond the Standard Model.

References:

- [1] M. Kozlov et al., Rev. Mod. Physics (2018), DOI: 10.1103/RevModPhys.90.045005
- [2] H. Bekker et al., Nature Comm. (2019), DOI: 10.1038/s41467-019-13406-9
- [3] T. Leopold et al., Rev. Sci. Instrum. (2019), DOI: 10.1063/1.5100594



2.2 ATOMIC AND MOLECULAR DYNAMICS

Intense laser light/matter interaction projects the correlated electronic clockwork of atoms and molecules onto photons, electrons and ions, and allows to interfere with and control their dynamics.

Reaction Microscopes and Laser Systems

Reaction microscopes – "the bubble chambers of atomic and molecular physics" – have been developed and are continuously improved at MPIK. Ultra-short intense laser pulses or particle beams induce a breakup of simple molecules. The fragment ions and electrons are caught by means of electric and magnetic fields and recorded by large-area time- and position-sensitive detectors. Their complete momentum vectors, and thus the geometry and dynamics of the molecules before their break-up, can be determined from the reconstructed trajectories of the fragments ("kinematically complete experiments"). The instruments are deployed in-house and at external light sources such as free-electron lasers (FELs). For the cryogenic storage ring CSR, a specific reaction microscope was designed and is presently under construction. It will be a key instrument for the worldwide unique possibilities for the investigation of slow and cold ions in the CSR.

Scheme of a reaction microscope.

In the Institute's laser laboratories, phase-controlled laser pulses as short as 5 femtoseconds at intensities of up to about 10¹⁶ W/cm² are routinely available for experiments. Even shorter pulses of some attoseconds duration are generated by nonlinear optical techniques. The resulting coherent high-harmonic radiation in the extreme UV range can then be combined with broadband infrared/visible pulses from the main Ti:Sapphire laser. Isolated as well as double and triple attosecond pulses are produced and used to probe gaseous atomic and molecular samples by interferometric methods. For pump-probe measurements, the time delay between two pulses can be precisely adjusted on attosecond time scales. Combined with spectroscopy or imaging detectors, this allows for direct and time-resolved observation (and control) of nuclear and electronic quantum motions in chemical reactions.

Ultrashort Laser Pulses – the Microcosm in Extremely Slow Motion

How does a quantum system evolve in time and is it possible to visualize or even control its motion? Today, this old dream of physicists from the early days of quantum mechanics

has become a real and growing field of research. The time scales of processes elapsing in quantum systems are extremely short: During chemical reactions, the atoms are moving within 10 to 100 femtoseconds ($1 \text{ fs} = 10^{-15} \text{ s}$), while the electrons which mediate the chemical bond are even faster: here, attoseconds ($1 \text{ as} = 10^{-18} \text{ s}$) are the characteristic time scale.

A key tool for time-resolved experiments are ultrashort intense laser pulses which are used to steer the atomic or molecular dynamics with extremely high precision. An electron released from an atom by a strong laser field is driven back and forth and revisits its parent ion while probing its structure. The wave nature of the electron leads to interference effects like in a holographic image which can be analysed to resolve the time-dependent interaction with the residual electrons of the atom.



", Chirped mirror" setup for generating ultrashort laser pulses.



Interferences in the photo-electron momentum distribution measured with a reaction microscope.



The buildup of a Fano resonance in real time, measured by strong-field-gated attosecond XUV absorption spectroscopy.

In most cases, a "pump-probe" scenario is applied, where the first "pump" laser pulse prepares the system in the desired way and starts the time evolution which is then probed by the second laser pulse. Molecular motions like vibration and rotation can thus be traced. Observing chemical reactions in real time at femtosecond resolution is a very promising research area. In combination with reaction microscopes even the ultrashort time span needed for an isomerization reaction – rearrangement of atoms within a molecule – which is essential also in eye-vision or photosynthesis could be observed.

To observe the motion of electrons, however, even shorter light pulses on the order of attoseconds are required. One possibility therefore is the generation of high harmonics of an intense femtosecond laser. This way, the requested pulse durations of less than 100 attoseconds at wavelengths of some 10 nanometers can be reached nowadays. The helium atom represents a prototype for the correlated motion of electrons. Both its electrons can be excited by absorption of extreme-

ultraviolet attosecond pulses. Another femtosecond laser pulse time-dependently probes the thus generated two-electron wave packet which can be reconstructed with the support of calculations that are based on known static wave functions. Laser pulses even allow to steer this electronic 'couple dance'. In the future, a directed manipulation of the electron pairs in molecules may influence chemical reactions and enable hitherto impossible syntheses.

Spectroscopy – the measurement of the absorption and emission of light as it interacts with matter – is one of the most important tools of physics. Line spectra are observed in the case of resonant interaction. Under certain conditions, they interfere with a continuous background and asymmetric line shapes ("Fano profiles") emerge. This can be illustrated as the superposition of coupled oscillations. Using ultrashort laser pulses, it is possible to control the temporal evolution and thus the quantum interference – for example to achieve the transformation of a spectral absorption into an emission line and resolve the ultrafast formation of a Fano resonance on the femtosecond timescale.



Photoionisation of matter is a subject of research since 1905 when Einstein discovered the quantum nature of light. The question about the intrinsic time-scale of photoionisation, however, has only been discussed after the discovery of attosecond laser-physics a few years back. How much time does it take for an electron to leave an atom after the absorption of a single photon? Answers to this question are now in reach with most modern methods. Time-delays are measured with the streaking scheme where a short XUV-pulse leads to the emission of a photoelectron which is then driven or probed by a superimposed long-wavelength laser field. The resulting photoionisation timedelay is the sum of the quantum mechanical Wigner time-delay (Δt_{wigner}) and a contribution arising from a coupling of the electron with the probe laser in the presence of the atomic Coulomb field. In an experiment at the free-electron laser FLASH in Hamburg we succeeded in measuring the Coulomb-Laser contribution only exploiting a XUV pump - Terahertz (THz) probe scheme. At a photon energy of 59.4 eV, neon 2p and valence shake-up photoelectrons were emitted into the THz probe-field with 152 µm wavelength. Using a reaction microscope, the 3D momentum vector of the photoelectron was measured as a function of the pump – probe delay-time. We observed a time shift due to the Coulomb-laser coupling between direct 2p photoelectrons and slow shake-up electrons of up to 100 fs depending on the

electron energy. Overall, the experimental results are in agreement with theoretical predictions for the Coulomb-probefield coupling induced time-shifts in atomic photoemission. *Reference:*

G. Schmid et al., Phys. Rev. Lett. 122, 073001 (2019), DOI: 10.1103/PhysRevLett.122.073001

A new mechanism in strong-field ionization of molecules

Strong-field photoionisation of atoms and molecules with short laser pulses is usually described as an electronic tunnelling or multi-photon transition from a bound molecular valence-state into the continuum. But is this the most probable ionization mechanism in the case of a molecule like H₂, or do other more subtle ionization pathways contribute significantly? To answer this question, we measured the 3D photoelectron momentum distribution in coincidence with the ionic fragments for strongfield ionization of H₂ using a reaction microscope.

Two major fragmentation channels can be distinguished. The molecule may stay either bound leading to H⁺, (bound ionization), or it fragments into a proton and a neutral H atom after the ionization (dissociative ionization). Surprisingly, we observed a significant enhancement of photoelectrons in the low-energy regime for the bound-ionization channel. By further inspection of well known asymmetries in electron emission using two-colour laser pulses, this enhancement was found to be due to a delayed electron emission, at times longer than the pulse duration. The results are in perfect agreement with autoionisation of vibrationally excited states in neutral H₂, that are populated by the interaction with the strong laser pulses. Vibrational energy in the nuclear motion is effectively transferred to the electronic system leading to the emission of a low-energy electron on time-scales that are longer than 10 femtoseconds.

H, ionizatior excitation laser field dissociatio autoionization H_2^4 0.0 0.2 0.4 0.6 0.8 8рπ (a) $\nu'=1 (H_2^*) \rightarrow \nu=0 (H_2^+)$ (a) 8pg 9pc (b) $\nu'=2 (H_2^*) \rightarrow \nu=1 (H_2^+)$ (b) (c) $\nu'=2 (H_2^*) \rightarrow \nu=0 (H_2^+)$ Relative intensity 1.00 0.75 0.0 0.4 0.2 0.6 0.8 Electron energy (eV)

Reference:

References:

Y. Mi et al., Phys. Rev. Lett. 118, 183201 (2017), DOI: 10.1103/ PhysRevLett.118.183201

Attosecond precision by resonant transient absorption

Using moderately strong ultrashort optical laser pulses, it is possible to control the most fundamental dynamics of electrons bound in matter at the femtosecond and attosecond timescale. Such measurements are realized with the technique of extreme-ultraviolet (XUV) attosecond transient absorption spectroscopy. A dedicated vacuum beamline has recently been upgraded with an in-situ XUV beam splitter for recording precision time-resolved absorption spectra of ultrafast electron dynamics [1]. For the case of autoionising doubly excited states in helium, a new method of extracting the laser-driven dipole response in real time has thus been demonstrated [2]. With even higher laser intensities, xenon is strongfield ionized which leads to sub-cycle attosecond oscillations in the ionization yield. By deliberately changing the dispersion of the probing XUV attosecond pulse, a precision and accuracy of only a few attoseconds has been achieved with transient absorption spectroscopy [3]. This opens new perspectives for attosecond delay measurements across resonant atom-specific transitions in atoms and molecules.







When atoms are subject to external electromagnetic fields, their quantum structure and dynamics can be distorted. With intense short-wavelength extreme-ultraviolet (XUV) light from the Free-Electron Laser in Hamburg (FLASH) it is possible to selectively couple to the innermost electronic structure of atoms, here carried out in two recent measurements in neon (left) and helium (right). In these experiments, characteristic energy shifts and changes of the shape of spectral absorption lines have been observed, which reveal XUV-induced ac Stark shifts of atomic energy levels. Supported by theoretical modelling, for instance in helium a transient population inversion of a correlated two-electron state was achieved [1]. On the other hand, in neon, transient coherence effects through plasma diffraction of ionized electrons on the few-femtosecond timescale have been revealed [2]. Observing such ultrafast resonant nonlinear effects in the XUV spectral range is a decisive step towards the quantum control of chemical reactions in general, with atomic site selectivity within a molecule. *References:*

[1] C. Ott et al., Phys. Rev. Lett. 123, 163201 (2019), DOI: 10.1103/PhysRevLett.123.163201
 [2] T. Ding et al., Phys. Rev. Lett. 123, 103001 (2019), DOI: 10.1103/PhysRevLett.123.103001

Colliding Atoms and Molecules – Billiard Game with Quantum Balls

Research on correlated quantum dynamics represents one of the great challenges in contemporary science. Researchers at the MPIK explore quantum dynamics on a fundamental level, starting from a limited number of few interacting particles in atoms and molecules, and extending to more complex finite quantum systems such as clusters or even biomolecules. Bombardment with charged particles (electrons, ions) is a key method for the study of these quantum systems. Novel multi-coincident imaging techniques developed at MPIK provide comprehensive information about few-body quantum dynamics and allow a test of theories for such reactions. Electron impact plays an important role in the environment, for example in the upper atmosphere and in interstellar space, as well as in technical plasmas and in radiation biology. During a collision, a molecule may break up into several fragments; this plays a crucial role in biological tissues, since, e. g., the DNA molecule can be altered chemically or even be destroyed.

Ultracold Dynamics – Investigating Exotic Quantum Gases



Scheme of the magneto-optical trap combined with a reaction microscope.

Very cold atomic gases with quantum properties are accessible by means of laser cooling. Lithium atoms behave as bosons or as fermions depending on the choice of their mutual interaction. In the bosonic regime weakly bound atom pairs form, the mutual distance of which is experimentally controllable. This exotic form of matter is investigated with a reaction microscope. By ionization of all atoms in bound pairs or in few-particle systems and determination of all ion momenta, it is possible to deduce the initial spatial configuration of the particles. Here practically instantaneous ionization is done by an intense femtosecond-pulsed laser beam. Whether and how the quantum state of the gas influences its ionization dynamics is also of interest.

Ultrafast energy transfer in hydrated biomolecules

Damage of biological tissue by ionizing radiation relies to a large part on secondary particles such as radicals, ions and the abundantly produced slow electrons which can cause, e.g., DNA single and double strand breaks. We have observed that the aqueous environment in biological tissue can enhance molecular ionization and fragmentation via a hitherto unrecognized damage mechanism called intermolecular Coulombic decay (ICD). As a model system to study this process we use complexes consisting of one water and one tetrahydrofuran molecule (THF) which is a surrogate for deoxyribose in the DNA backbone. In the first step the water molecule is ionized by electron impact in the oxygen inner-valence shell (I). Then, within a few tens of femtoseconds the inner-valence vacancy is filled by an outer-valence electron and the released energy is transferred to the neighbouring THF molecule ionizing it as well (II). The charged ions repel each other and gain kinetic energy in the subsequent Coulomb explosion (III). Altogether two energetic ions (see the experimental momentum correlation diagram of



the H_2O^+ and THF⁺ ions) are produced in addition to the three reactive secondary electrons all of which can cause further damage in the vicinity. Thus, dense and therefore particularly harmful ionization clusters of several ionization processes within a volume typical for a biomolecular system like DNA can be produced. Future studies aim at determining the relative importance of ICD with respect to other damage mechanisms. Furthermore, we want to test theoretical calculations which indicate that ICD can be triggered by inner valence ionization of carbon atoms in organic molecules as well. Therefore, ICD is expected to be a widespread phenomenon in loosely bound organic matter that can provide a functional mechanism for the direct damage of biomolecules such as DNA. *Reference:*



Momentum of H_2O^+ (a.u.)

X. Ren et al., Nature Physics 14, 1062 (2018), DOI: 10.1038/s41567-018-0214-9

The Cryogenic Storage Ring

In the electrostatic cryogenic storage ring, CSR, beams of cold molecular ions of any size and highly charged ions can be investigated essentially without any influence of the environment. This is achieved by purely electrostatic ion optics, keeping the ring under extremely low pressure and at a temperature of a few degrees above absolute zero. The ions are produced in dedicated ion sources and injected into the ring by high voltages of up to 300 kV. In addition, a device for injecting beams of neutral atoms is attached to the CSR. An electron cooler improves the stored ion beam quality, and the electrons are available as reaction partners. The innovative mechanical concept of the CSR was developed and realised in close cooperation with MPIK's engineering design office and precision mechanics shop.



Copper strips distribute the cold among the chambers in the ultracold storage ring CSR.

Internal cooling of molecules in the Cryogenic Storage Ring

Storage-time dependent photodetachment at the CSR sensitively probes important properties of diatomic molecules that could not be accessed in earlier measurements. These include the molecular dipole moment and the precise free-space radiative lifetime of rotationally excited levels, which becomes very long for the lowest excited quantum states. With a laser tuned in its wavelength very close to the detachment threshold of the OH⁻ anion, the detachment signal can individually probe



the population in excited rotational levels while they are decaying. This becomes possible as levels live shorter the higher they are excited: of the levels the laser can photodetach, only the lowest one will be left after sufficient storage time. For more than a few minutes of storage, the remaining levels are J = 0 and 1, only. A detailed analysis of the CSR data, taken for 20 min of storage at 9 laser wavelengths, yielded the

relative photodetachment cross sections and the population fraction remaining in J = 1, which also quantified the blackbody field in the CSR. Indeed, this revealed that stimulated emission through the blackbody field accelerated the decay of the J = 1 level by about 12%. Only with this information its natural lifetime, excluding blackbody effects, could be extracted. As the first measurement of its kind, this revealed as much as 10% deviation from the rotational lifetimes predicted for OH⁻ from quantum chemical calculations and yielded the dipole moment of this molecule with as little as 1.5% uncertainty.

Reference: C. Meyer et al., Phys. Rev. Lett. 119, 023202 (2017), DOI: 10.1103/PhysRevLett.119.023202

Laboratory Astrophysics – the Chemistry of Space

One puzzling question is the formation of organic compounds in interstellar clouds. The gas-phase chemistry is driven by reactions involving ions and radicals which are created in collisions with photons and cold electrons. Here, the H_3^+ molecular ion plays a key role. The break-up of molecules after capture of an electron ("dissociative recombination") can



The network of cosmic chemistry in interstellar clouds.

be studied in detail in storage rings. In the new cryogenic storage ring CSR, for the first time, conditions are reached that correspond to interstellar temperatures where many types of internal motion are in fact frozen in molecular ions.

The positive ions of interest range in size from small atoms and molecules up to organic compounds. Also, negatively charged molecular ions (anions) are of interest here as they represent an important source of slow electrons. Provided sufficient inner excitation (vibration), they can literally "evaporate" electrons. Moreover, collisions with neutral atoms are also of great importance for astrochemistry. A novel neutral-atom beam setup at the CSR has recently been commissioned. It combines ground term atoms with cold molecular ions, and thus, for the first time, provides access to this largely unexplored class of processes under true interstellar conditions.

State-resolved dissociative recombination measurement improves predictions on primordial HeH*

Molecules play an important role in the formation of the first stars. This is due to their ability to cool gas clouds by the collisional excitation of molecular rotational levels and subsequent radiative emission. The cooling leads to gravitational collapse of the cloud, an inevitable step for star formation. From the few elements available in the early Universe, namely H, He, and traces of Li, only a handful of molecules could combine. Here HeH⁺ is an important candidate for a strong molecular coolant given its large dipole moment and predicted abundance. The abundance calculations rely on molecular reaction data for various formation and destruction processes, in particular at low (~10 K) temperatures. Here the dominant HeH⁺ destruction channel is via capturing a free electron, a process called dissociative recombination (DR). For this reaction all previous data have been obtained in room-temperature experiments, i. e., for ions with several rotational states populated. Given the

resonant nature of DR, however, the DR reaction rate was predicted to be strongly rotational-state dependent for many ions. To resolve this issue we have performed HeH⁺ DR measurements using the newly implemented electron cooler beam in the Cryogenic Storage Ring (CSR). In the CSR radiation field, where ~99% of the relevant spectral density represents the 6K wall temperature and 1% radiation leakage from 300 K, HeH⁺ ions relax radiatively to reach >92% in the J = 0 ground state within ~50 s. From the radiative cooling model and the detailed time-dependent measurement of the HeH⁺ recombination rate we derive rotationalstate resolved absolute rate coefficients. At ~10 K the new HeH⁺ destruction rates are >20 times lower than the data used in early Universe models previously. Correspondingly, the primordial HeH⁺



abundance at redshifts relevant to the formation of the first stars and galaxies should be >20 times higher than thought so far. *Reference:*

O. Novotný et al., Science 365, 676-679 (2019), DOI: 10.1126/science.aax5921

Low-temperature rates for key reactions of interstellar gas-phase water formation

The formation of water in the diffuse interstellar medium proceeds through a series of ion-neutral reactions involving the light molecular ions OH^+ and H_2O^+ . The full reaction chain reads as follows,

$OH^+ + H_2 \rightarrow H_2O^+ + H_1$	(1
$H_2O^+ + H_2 \rightarrow H_3O^+ + H_2$	(2
$H_{3}O^{+} + e \rightarrow H_{2}O + H_{2}$	(3

and it is terminated by the electron-recombination of H_3O^+ , resulting in the formation of interstellar water in the gas phase. Besides the relevance of water for life as we know it, it is also an important reservoir of oxygen in space. Furthermore, the recent detections of OH⁺ and H_2O^+ ions by the Herschel space telescope have been used to constrain important param-

eters like the cosmic ray ionization rate and the fraction of atomic to molecular hydrogen. For this purpose, accurate reaction rates for the above reactions are needed. Previous analyses have used rates derived at room temperature, which were extrapolated to interstellar temperatures (10 K to 100 K). We have measured the rate coefficients of reaction (1) and (2) in a cryogenic 22-pole ion trap, between 20 K and 150 K. Our experimental results reveal significantly faster low temperature reaction rates in both cases. These findings are in excellent agreement with calculations of a collaborating theory group from Cyprus and the USA, who use a modern ring-polymer-molecular-dynamics (RPMD) approach, which takes into account



quantum effects that prove to be crucial at low temperatures. Our results show that extrapolation can introduce large uncertainties and errors into the interpretation of observational data, and that quantum effects should not be neglected at low temperature, even in the case of seemingly simple barrier-less ion-neutral reactions. *Reference:*

S. Kumar, F. Grussie, Y.V. Suleimanov, H. Guo, H. Kreckel, Sci. Adv. Eaar3417 (2018), DOI: 10.1126/sciadv.aar3417



2.3 MATTER IN EXTREME FIELDS

According to quantum electrodynamics the properties of matter and even of vacuum are altered by intense laser fields.

Matter in Strong Laser Fields – at the Frontiers of Feasibility

The investigation of the interaction of matter with laser pulses and x-ray sources by now has reached a level at which fundamental aspects such as the quantum nature of both light and matter, relativity and couplings among the involved particles have become key issues and substantial challenges alike. Theory helps to explore the effects of extremely strong fields, even though this partly will be reached experimentally only in the near future. This requires the search for solutions of the many-body time-dependent Schrödinger and Dirac equations. Furthermore, quantum electrodynamics, nuclear effects and pair creation are considered.

One typical topic of interest is the fully relativistic understanding of quantum processes during tunnel ionization of an atom in a very strong field. A simple model of this process claims that the electron tunnels instantaneously through the laser-generated quantum barrier and appears at its exit with vanishing momentum. As a consequence, the complete momentum carried by the absorbed photons would be transferred to the ion. Meanwhile it has however been demonstrated that the above simple model needs to be corrected and that this momentum is shared by the electron and ion following a complete quantum relativistic calculation.

The question for the time span the electron needs for tunnelling is however controversial to date: Does this take time or is it instantaneous? Theoretical considerations based on a concept published by Nobel laureate Eugene Wigner in 1955 predict a finite tunnelling time. A recent joint theoretical and experimental study at MPIK using a refined model succeeded in translating the Wigner time into an observable quantity. An accurate analysis of electrons emerging from noble gases in ultrashort circularly polarized laser pulses gave evidence of a finite tunnelling time up to 180 attoseconds ($1 \text{ as} = 10^{-18} \text{ s}$). Our more recent calculations intuitively explain Wigner's tunnel exit momentum via a higher-order strong-field approximation with interferences of recolliding trajectories under the tunnel barrier.



Tunnel effect in a circularly polarized laser field: The electron escapes from the atom through the potential barrier in the presence of the strong laser field. The "simple-man" (instantaneous) and Wigner (finite time) models predict different electron trajectories.

Signatures of under-the-barrier dynamics in tunnelling ionisation



electric field direction

Schematic picture of laser-induced tunneling ionization: (dashed) the direct trajectory, and (solid) the under-thebarrier recolliding trajectory. The interference of the direct and the rescattered trajectories induces a shift of the peak of the photoelectron momentum distribution. The tunneling is nonadiabatic, when the energy is not constant during the tunneling process.

In a strong laser field ionization of an atom takes place via tunnelling of the electron from the atomic bound state into the continuum through the potential barrier formed by the atomic potential and the laser field. Although the under-the-barrier dynamics is a small part of the whole laser-electron interaction in this process, it imprints its gentle signatures in the photoelectron momentum distribution at the detector. High resolution of momentum detection, better than 0.01 atomic units, is required to observe these signatures. One of these signatures is the time delay in the attoclock, which recently has been measured in a mixture of gases in [1]. In the latter, the systematic experimental errors have been canceled by measuring the difference of the time delay for two gases. While there are debates how to interpret the attoclock time delay, we put forward a simple straightforward interpretation in [2]. The time delay is equivalent to a shift in the transverse momentum distribution of the attoclock and we have shown that the latter emerges when the interference of the direct ionization path with the under-the-barrier recolliding one is accounted for, see Figure. We have also predicted another signature of the under-the-barrier dynamics due to the

non-dipole effect of the laser magnetic field. The peak of the electron momentum distribution along the laser propagation direction is shifted forward at the tunnel exit during the under-the barrier wave packet formation. It has a consequence for the photon momentum partition between the ion and the electron in the ionization process. Our fully relativistic prediction has been recently confirmed by an external group in an ultrahigh precision measurement in [3]. *References:*

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Extreme Light-Matter Interaction – Precisely Controlling and Probing Nuclear Transitions



Control of x-ray light via mechanical motion of a resonant target. Before motion (top), the light scattered by the target (blue) extinguishes the excitation (red). After the motion (bottom), the scattered light is displaced and the waves enhance each other (magenta).

Quantum optics with x-ray light emerged in the last years as a new field. Of particular interest are certain atomic nuclei that only interact with x-rays with an extremely well-defined photon energy, due to an effect discovered by Rudolf Mößbauer at the precursor institute of MPIK in 1958. Spectroscopy – the measurement of the absorption and emission of light as it interacts with matter – of such precise nuclear transitions forms the basis for numerous applications across the natural sciences. Establishing coherent and quantum control of these nuclei is crucial for future applications, but remains a big challenge due to the lack of intense x-ray driving fields with a small energy spread.

To tackle this problem, a joint theoretical and experimental study showed that the macroscopic motion of a sample can shuffle light intensity within the spectrum of given x-ray pulses such that it is enhanced at the desired energy. A recent theoretical and experimental follow-up work exploited such improved x-ray pulses to coherently control the quantum dynamics of matter. In particular, the controlling pulses could switch the dynamics of nuclei in a second target between the elementary processes of absorption and stimulated emission. In parallel, the theory for a first application of the motion control was developed, which allows one to measure correlations between different observables of a quantum mechanical system without the usually unavoidable perturbing back action of the measurements on the system's dynamics.

Extremely narrow nuclear transitions are also of interest for accurate measurements. A prime example is the nuclear clock based on thorium-229, which promises to improve the accuracy of the best atomic clocks available today. Recently, a team at the LMU Munich measured with increased precision the previously uncertain nuclear transition energy. The extraction of this energy from the experimental data required simulations performed at MPIK. Now lasers specifically designed to excite the nuclei can be constructed, fuelling fundamental research based on extremely precise time measurements.

Coherent x-ray-optical control of nuclear dynamics

Mößbauer nuclei feature resonances with extremely narrow spectral line widths, which form the basis for a broad range of applications across the natural sciences. On the other hand, the narrow linewidth causes a lack of strong resonant x-ray driving and control fields for the nuclei. To address this issue, we developed and implemented a method to simulate strong control fields in certain settings using precisely controlled mechanical motions of a nuclear target. This motion allows us to impose a time-dependent phase on the x-ray pulse. In a first experiment together with the group of Ralf Röhlsberger (DESY, Hamburg), we used the mechanical control to enhance the resonant intensity of given x-ray pulses, by redistributing off-resonant photons onto the resonance [1]. Such optimised pulses allow for stronger driving of the nuclei, and enhanced signal rates. After having demonstrated this control over the x-ray light, in a follow-up experiment, we in turn demonstrated the control of nuclear dynamics [2]. For this, we generated tunable x-ray double-pulse sequences via the motion. This double-pulse was then used to drive the nuclei in a second target. After the first pulse excited the nuclei, we controlled the relative phase of the second pulse to switch the subsequent nuclear dynamics between stimulated



Experimentally recorded time- and energy-resolved spectra of nuclear resonances. The upper and lower panels compare two different motions applied to the nuclei. The effect of the motion on the spectra is clearly visible in the modifications of the interference structures.

emission and further absorption. Using a multi-dimensional spectroscopic measurement technique, we demonstrated the exceptional few-zeptosecond stability of this coherent control. As future applications, we envision advanced spectroscopy methods and pump-probe techniques using the extremely narrow nuclear transitions.

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Towards a nuclear clock with ²²⁹Th

Throughout the entire nuclear chart, ²²⁹Th presents the lowest-lying excited state at approximately 8 eV, being accessible for vacuum-ultraviolet lasers. This metastable state offers the unique opportunity of a nuclear clock which is expected to outperform present technology. Such a clock would have a large variety of applications, ranging from relativistic geodesy over dark matter research to the observation of potential temporal variation of fundamental constants.

Understanding how such an anomalous low-lying state could occur in this actinide isotope is paramount for the prediction of its properties. We have developed a theoretical model which can reproduce the level structure of the entire energy spectrum of ²²⁹Th up to 400 keV. This model shows that the isomeric state occurs due to a very fine interplay of the collective motion of the even-even core, which presents quadrupole-octupole deformation (see Fig.), and the single odd neutron [1, 2]. The Coriolis interaction occurring between core and odd neutron renders possible the nuclear clock transition.

The very recent direct measurement of the clock transition energy yielded 8.28 ± 0.17 eV. It employed spectroscopy of the internal conversion (IC) electrons emitted in-flight during the decay of the excited nucleus in neutral ²²⁹Th atoms [3]. In the IC process, the ²²⁹Th nucleus transfers its energy to the electronic shell, ionizing an outer shell electron (see Fig.). Due to the complicated electronic level schemes occurring in the experiment, our IC calculations were essential to extract the nuclear tran-



a) IC decay ejects the 7s electron. b) Nuclear quadrupoleoctupole deformation of the collective core and additional odd neutron.

sition energy from the measured electronic spectra. This energy determination is the starting point for high-resolution nuclear laser spectroscopy and the development of a nuclear optical clock of unprecedented accuracy. *References:*

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Strong-Field Quantum Electrodynamics – Modifying the Vacuum

In the language of quantum electrodynamics (QED), the electromagnetic interaction is described as the exchange of so-called virtual photons between charged particles. Another consequence of this theory is the fact that there is no empty space, i.e., the vacuum can be pictorially described as being filled with virtual particles. Though their existence is only allowed for a very short time – given by quantum uncertainty – the presence of virtual particles can be detected by high-precision experiments. At the same time, QED is the to date best tested theory in physics at all.

Of particular interest is the QED in extremely strong fields. Those fields will influence the charged virtual particles in the quantum vacuum such that the vacuum becomes polarized changing its optical properties. Our theories deal also with the fundamental question of pair production, spin dynamics and radiation reaction. In the latter case a charged particle

is accelerated in an electromagnetic field and emits electromagnetic radiation which in turn acts back on the particle's motion. Intense laser fields can help to test experimentally the underlying equations. Quantum aspects of radiation reaction in electron dynamics should show up in studies using already available laser systems. This is also of importance for many-particle ensembles like a lasergenerated relativistic plasma.

Very strong fields also prevail in the vicinity of the nuclei of heavy elements. High-precision QED calculations of the inner structure of matter for especially highly charged trapped and stored ions are of particular relevance for our institute. The interplay of theory and experiment significantly contributes to the determination of fundamental properties such as the magnetic moment of the electron. On the one hand, comparison with precision experiments permits validation of QED predictions, while on the other hand theory helps to determine natural constants like the electron mass: its value became by an order of magnitude more accurate.



In highly charged ions (HCI), the inner-shell electrons experience extreme electromagnetic fields of the nucleus. Effects of relativity and of quantum electrodynamics (QED) are boosted in these ions as compared to their neutral counterparts, rendering HCI an ideal system for testing the validity of the Standard Model in strong fields. In high-precision calculations, we predict the structural properties of HCI considering the self-interaction of electrons and corrections due to the virtual creation and annihilation of dilepton pairs. In many-body systems, the electrons also exchange photons between each other, and such correlation effects are treated by large-scale atomic structure calculations and by QED perturbation theory.



Of particular interest is the magnetic moment of HCl, which can be measured to ultimate precision in Penning traps, by employing the newly constructed ALPHATRAP setup of the Institute. Beyond successful tests of QED, these studies yield values of fundamental constants. In order to further improve the determination of the electron mass with a record-breaking precision, we put forward further ionic systems [1], and push forward the boundaries of theory by evaluating multi-loop QED loop corrections in a non-perturbative manner, i. e. in a framework which is valid in strong Coulomb fields [2]. Another fundamental access to QED and electronic structure effects is given by the investigation of energy levels. This became recently possible by means of precision mass spectroscopy: the electronic binding energy can be directly determined by comparing the

masses of different ions. We perform high-precision calculations of the binding energy of the outermost electron of the Xe¹⁷⁺ ion, and find a perfect agreement with the very first measurement of this kind performed with the recently built PENTATRAP experimental setup of the Institute [3]. Besides testing our understanding of the structure of HCI, such studies are expected to deliver improved mass differences in nuclear physics as well as a refined test of the mass-energy equivalence. *References:*

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Scheme of the QED contributions to the

electronic structure of highly charged ions:

the electrons (blue balls) interact with each

other and themselves via virtual photons (wave lines). In the field of the nucleus, also

particle-antiparticle pairs (blue and red

balls) may be virtually created.

High-Energy Laser Physics and Laboratory Astrophysics – Cosmic Accelerators in the Laboratory Scale

With increasing laser intensity, the underlying physics has been continuously transferring from atomic to high-energy physics. Our many-particle quantum, quantum plasma and semiclassical particle-in-cell (PIC) codes incorporate especially radiative reaction, spin dynamics, pair cascades as well as deviations from the locally constant field approximation. Cascades of electron-positron pairs were seen as a risk to generate extremely strong laser pulses and we have put forward means to prevent those via suitably chosen focus areas. In particular, most recently various concepts have been developed to generate polarized intense lepton and gamma GeV beams. They are based on spin-dependent radiative reaction and are likely to find applications in high-energy, solid-state and astrophysics.

Already to date, highly intense laser fields enable the acceleration of particles to energies up to the order of giga-electronvolts (GeV). This opens the possibility to reproduce physical conditions in the laboratory, as they prevail in extreme astrophysical processes. In close collaboration with external experimental groups, MPIK researchers developed models for the production of ultrarelativistic lepton beams consisting of electrons and positrons in equal amounts as well as gamma rays. Thereby, the conversion of bremsstrahlung to electron-positron pairs could be identified as a substantial mechanism and in addition the emission of the gamma rays to significantly slow down the ultrarelativistic electrons. The investigation of such highly energetic processes on laboratory scale is of great importance also for astrophysics: Cosmic gamma-ray bursts for example, to our present knowledge emerge from the extremely collimated ultrarelativistic leptonic jets which are emitted along the rotation axis of certain types of collapsing stars.



Laboratory production of ultrarelativistic electron-positron beams by laser-accelerated electrons hitting a metal target.

Quantum radiation reaction and strong-field QED beyond the local constant field approximation

A pioneering experiment has been carried out where ultra-relativistic electrons (energy up to 2 GeV) head-on collided with an ultra-intense laser beam (intensity $I_0 \approx 10^{21}$ W/cm²) [1], with our team being responsible of the theoretical interpretation of the experimental data. Our numerical simulations were of crucial importance to identify the experimental regime of interaction approaching for the first time the so-called

regime of interaction approaching for the first time the so-called "quantum radiation-reaction regime" in laser-electron collision, where each electron emitted several high-energy photons. Our quantum simulations, unlike pure classical ones, provided good agreement with the experimental data. Moreover, our simulations hinted to a failure of the so-called local constant field approximation (LCFA) which has always been implemented in quantum numerical codes and which consists in treating the emission process as perfectly localized in space and time.

Following up on the above results, we developed the first scheme to go beyond the LCFA in the infrared part of the photon emission spectrum [2, 3], where non-local effects are more severe (see Figure). The result is a fully local scheme to investigate the emission of radiation beyond the LCFA in arbitrary background electromagnetic fields. The improvement of the model developed in [2, 3] as compared with the LCFA is clear from the numerical example in the figure, where the LCFA spectrum differs from the exact one by more than one order of magnitude in the infrared part.



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Laser-induced polarisation of electron and positron beams

Relativistic polarized electron and positron beams are fundamental experimental tools to test symmetry properties in physics. Recently we have shown a way to polarize electron and positron beams with currently available realistic laser fields [1, 2]. Nonlinear interaction of electrons with an elliptically polarized laser field with a small ellipticity has been shown to result in splitting of the beam with respect to polarization due to the spin dependence of radiation reaction, which yields



An intense linearly polarized two-color laser pulse head-on collides with an unpolarized relativistic electron beam, resulting in emission of photons, which decay into polarized e^+ and e^- , with spin parallel and anti-parallel to the laser's magnetic field direction, respectively. up to 70% polarization [1]. Moreover, in strong external fields the electron-positron pair production probabilities possess much higher asymmetry with respect to the spin of the created particles than the radiation. The latter property is harnessed for generation of highly polarized positrons in a two-colour asymmetric laser field [2]. The pair generation is a nonlinear process, which is strongly suppressed in the minor cycle of the asymmetric field, and allows (see figure) up to 60% polarization of the generated positrons. Furthermore, a way of generating high-intensity polarized electron beams is shown in [3], employing molecular gas targets polarized in a laser field. In such a target, polarized electrons can be accelerated to high energies in the wakefield of an intense laser pulse. While the

magnetic field generated in the wake may cause beam depolarisation, we find conditions under which the accelerated electrons are strongly bunched along the beam axis, where the role of the magnetic field is minor. Limiting the depolarisation to around 10%, in this way polarized electron currents in the kiloampere range are achievable.

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