Quantum Dynamics

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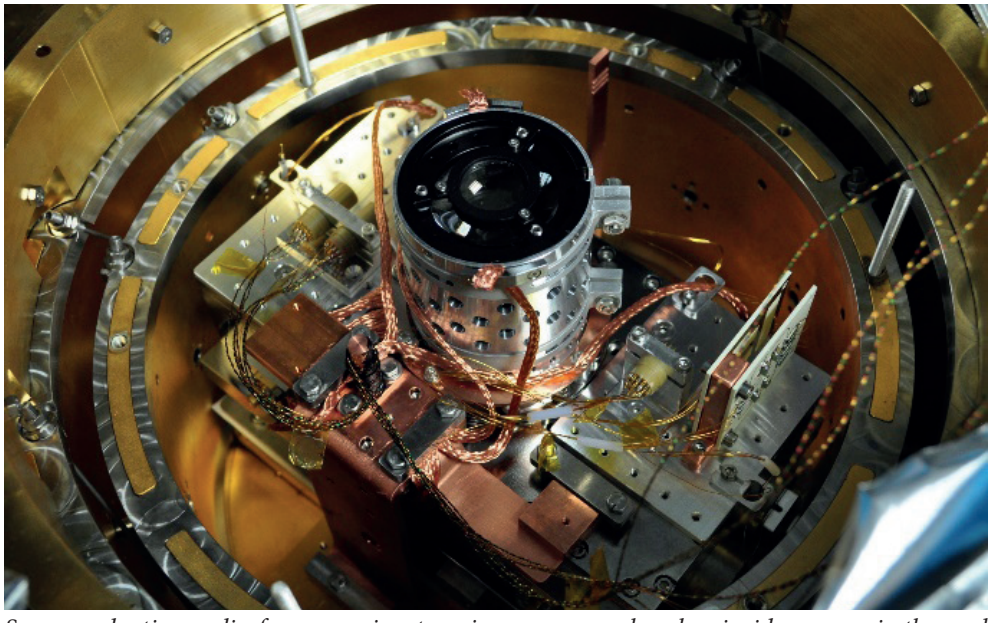
2.1 Highest Precision

An extremely precise atomic balance: PENTATRAP consists of five identically constructed Penning traps arranged one above the other. In these traps, two ions can be simultaneously measured in comparison. In order to minimise uncertainties, the ions are moved back and forth between different traps.

Ion Traps

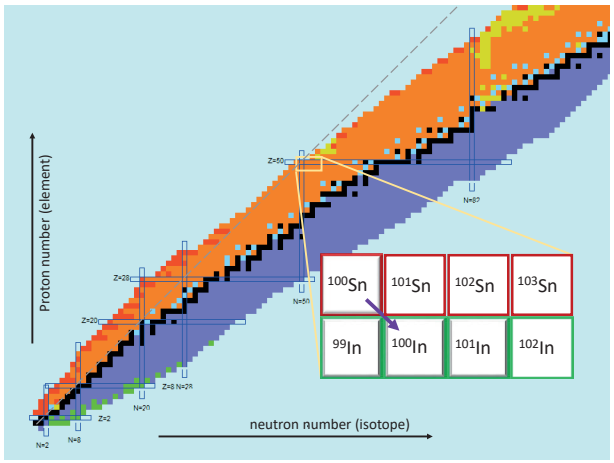
Ions can be stored in traps by the superposition of electric and magnetic fields in an extreme vacuum. Penning traps allow storage of a single ion that performs a characteristic oscillating motion in the trap. The ion's mass and further properties like magnetic moments of the bound electron in highly 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 nuclei 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 the framework of the BASE collaboration at CERN a new method for sympathetically cooling of protons using laser-cooled beryllium ions was implemented. In the future, this method will also be applied to antimatter. Recently, the charge-to-mass ratios of antiprotons and protons were found to be identical to eleven significant digits.

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 a novel superconducting trap for precise frequency measurements. Crystals of highly charged ions can be prepared with laser light and kept almost undisturbed over a longer period of time: a promising realisation of long-lived qubits. It is part of the BMBF quantum technology project Many-Frequency Control of Ultra-stable Qubits in Superconducting Ion Traps which started at the MPIK on 1 September 2021.



Superconducting radio-frequency ion trap in a vacuum chamber inside cryogenic thermal shields for operation at 4 K.

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



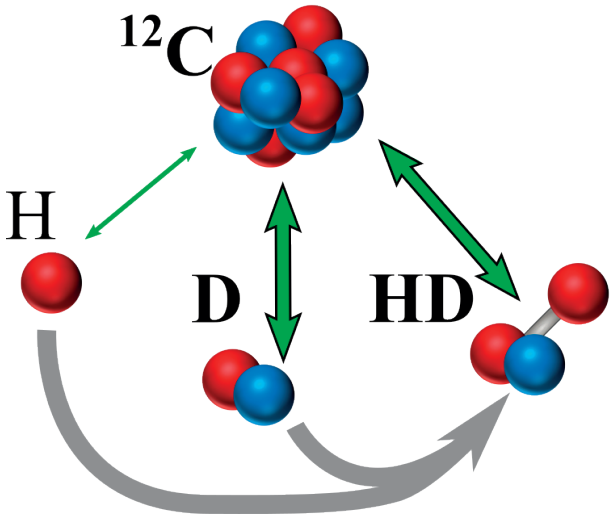
The exotic tin-100 and some of its measured and calculated neighbouring isotopes as an enlarged section of the nuclear chart (nuclei marked in black are stable, the other colours stand for different decay modes; the blue open bars mark the magic numbers and the dashed grey line connects nuclei with the same proton and neutron number). The purple arrow shows the decay of tin-100 to indium-100.

Penning-trap mass spectrometry at MPIK allowed to perform world-record measurements of the atomic masses of the electron and hydrogen isotopes. Both the proton and deuteron masses were found to be smaller than the previously accepted values. 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 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.

High-precision measurement of the deuteron's atomic mass



Summary of all currently measured light ion masses referenced to ^{12}C with the LIONTRAP experiment (green arrows). The agreement of the HD mass based on its constituents' masses and the binding energy (grey arrows) realises an independent internal cross check.

The rest masses of the proton, deuteron, triton and helion are very important to verify our understanding of physics for example to test quantum electrodynamics (QED) and search for 5th forces. The most precise 3-body QED tests compare rovibrational energies of the HD^+ molecular ion with their corresponding theoretical predictions at the low 10^{-11} level [1]. For these tests, accurate particle masses at the same level are required as input parameters. Recent inconsistency in these masses, determined with different mass spectrometers known as light ion mass puzzle, strongly calls for additional independent measurements.

The deuteron mass measurement was carried out at the LIONTRAP experiment, where the proton's atomic mass has already been determined with a relative precision of 3×10^{-11} [2]. Here a non-destructive comparison of the cyclotron frequencies of a single trapped highly charged carbon ion and the ion of interest is carried out. The inhomogeneous magnetic field has been the dominant systematic uncertainty of the proton mass measurement, but it was reduced by a factor of 50 for the deuteron campaign. This enabled the so far most precise mass measurement in atomic mass units resulting in a relative uncertainty of only 9×10^{-12} [3]. Although the value for the deuteron mass is 5σ smaller than the previous literature value, it is confirmed by another direct measurement of the HD^+ mass. Additionally, it also reduces the light ion mass puzzle and agrees with the recent HD^+ spectroscopy results [1]. This measurement encourages further investigations of the remaining light ion masses at the LIONTRAP experiment.

References:

[1] M. Germann et al., Phys. Rev. Res. 3, L022028 (2021), DOI: 10.1103/PhysRevResearch.3.L022028
[2] F. Heiße et al., Phys. Rev. A 100, 022518 (2019), DOI: 10.1103/PhysRevA.100.022518
[3] S. Rau et al., Nature 585, 43-47 (2020), DOI: 10.1038/s41586-020-2628-7

How magnetic is ^{3}He?

The hyperfine structures of hydrogen-like ions are a unique gateway to access nuclear magnetic moments and nuclear structure. Hence, while eliminating the ignorance of essential links in metrology due to insufficiently known moments, at the same time these ions provide complementary insight into the dynamic and static properties of the inner nucleon. The very recently started ^{3}He experiment exploits these characteristics to provide a new standard for absolute precision magnetometry and determine the nuclear charge and current distribution of ^{3}He.

To this end, a novel Penning-trap experiment was designed and built. Using novel techniques the advanced four Penning-trap system enables non-destructive measurements of the nuclear quantum state and is prepared to perform sympathetic laser cooling of single, spatially separated ions to sub-thermal energies [1].

In the first measurement campaign, ^{3}He was investigated by exciting microwave transitions at 140 GHz likewise 4 GHz between the ground-state hyperfine states. This enabled us to determine the nuclear *g*-factor, the electronic *g*-factor and the zero-field ground-state hyperfine splitting of $^3He^+$ with a precision of 5×10^{-10}, 3×10^{-10} and 2×10^{-11}, respectively [2].

Our measurement constitutes the first direct determination of the $^3He^+$ nuclear magnetic moment. The result is of utmost relevance for absolute precision magnetometry, as it allows the use of He NMR probes as an independent new standard with much higher accuracy. In addition, the comparison to advanced theoretical calculations enables us to determine the size of the ^{3}He nucleon with a precision of 2.4×10^{-17} m.

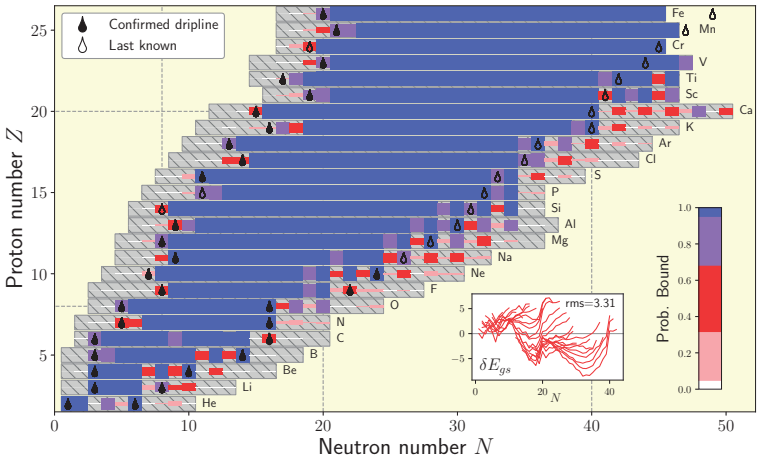
In future, the magnetic moment of the bare $^3He^{2+}$ nucleus will be measured employing sympathetic laser cooling. To this end, a novel 13-pole Penning trap for an improved coupling of spatially separated ions as well as a Penning trap for improved detection fidelities for the nuclear spin state have been designed and built [3].

References:

[1] A Mooser et al., J. Phys.: Conf. Ser. 1138, 012004 (2018), DOI: 10.1088/1742-6596/1138/1/012004
[2] A. Schneider et al., Nature 606, 878 (2022), DOI: 10.1038/s41586-022-04761-7
[3] A. Schneider et al., Ann. Phys. 531, 1800485 (2019), DOI: 10.1002/andp.201800485

Strong interactions and exotic nuclei

In the physics of nuclei, there have been rapid advances in the past years combining ab initio many-body methods with nuclear forces based on chiral effective field theory of the strong interaction, QCD. We have proposed and developed a powerful and flexible ab initio approach, the in-medium similarity renormalization group (IMSRG), which has enabled systematic calculations of nuclei up to 100 nucleons. Using the IMSRG, we explored the limits of existence of nuclei, the proton and neutron drip lines, from the light through medium-mass nuclei, based on a chiral effective field theory interaction and including theoretical uncertainties [1]. This first global ab initio calculation up to iron is shown in the figure. Remarkably, where the drip lines are known experimentally, our predictions are consistent within estimated uncertainties.



Global ab initio calculations of nuclei up to iron [1]. The gray region indicates all 700 nuclei calculated with the IMSRG, while the colour and height of each square corresponds to the estimated probability that a given nucleus is bound. This is compared to the experimentally confirmed drip lines or the last known isotope.

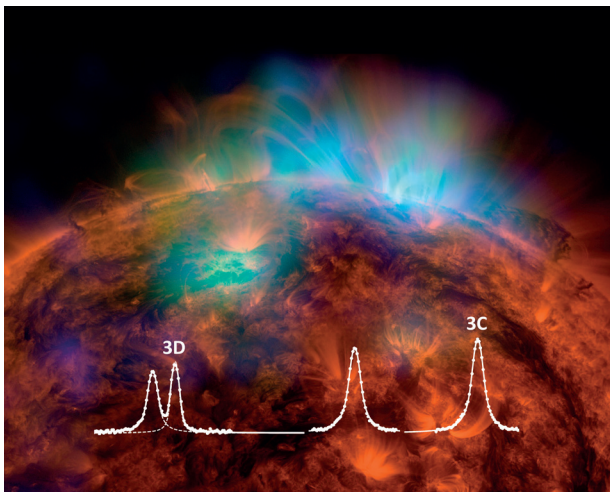
These advances have great synergies with the experimental frontiers in exotic nuclei, as neutron-rich nuclei are sensitive to new aspects of nuclear forces. For example, three-nucleon forces are key to explain the limits of bound isotopes and how magic numbers emerge. Recent highlights of our joint experiment-theory investigations focused on nickel and indium isotopes: We have explored the evolution of charge radii from $^{58\text{-}70}$Ni [2], the emergence of the doubly magic nature of ^{78}Ni [3], and the masses of indium isotopes near ^{100}Sn [4].

References:

[1] S.R. Stroberg, J.D. Holt, A. Schwenk, J. Simonis, Phys.Rev.Lett. 126, 022501 (2021), DOI: 10.1103/PhysRevLett.126.022501
[2] S. Malbrunot-Ettenauer et al., Phys.Rev.Lett. 128, 022502 (2022), DOI: 10.1103/PhysRevLett.128.022502
[3] R. Taniuchi et al., Nature 569, 53 (2019), DOI: 10.1038/s41586-019-1155-x
[4] M. Mougeot et al., Nature Phys. 17, 1099 (2021), DOI: 10.1038/s41567-021-01326-9

Highly Charged Ions – Matter under Extreme Conditions

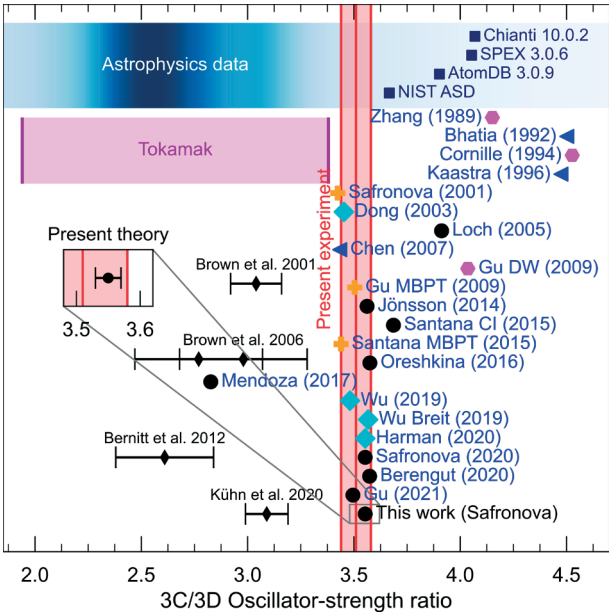
In fact, most of the visible matter in the Universe is assumed to be highly ionized. Analysis of the observed light (visible, UV, or x-ray) from these ions needs support by theoretical structure calculations which are often not accurate enough to determine, e. g., the temperature of the hot environment. The controlled production of highly charged ions (HCI) 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 resolved a key astrophysical problem.



X-ray fluorescence spectrum of highly charged iron ions, background: the Sun in x-ray light (© NuStar, NASA).

The cryogenic ion trap CryPTEx provides efficient cooling of trapped HCIs by means of laser-cooled Be^+ ions 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 next generation of CryPTEx focuses on extremely stable trapping conditions by isolating mechanical vibrations. Furthermore, a novel superconducting Paul trap resonator will enable precise localisation and strong confinement of HCIs in low-noise trapping potentials. With PENTATRAP, tiny differences in mass between different quantum states are measured providing new insights into heavy atoms. Thereby, a previously unobserved quantum state in rhenium was discovered, which could be interesting for future atomic clocks. At ALPHATRAP, the ground-state *g*-factors of highly charged ions can be measured with uncertainties at the 10^{-12} level. This provides an impressively precise test of the Standard Model of particle physics, allowing conclusions regarding the properties of nuclei and setting limits for new physics and dark matter.

A tale of two lines



Our new experimental data (vertical band) confirm recent large-scale calculations that demonstrated numerical convergence and unequivocally point to problems in earlier measurements, astrophysical observations, and theory.

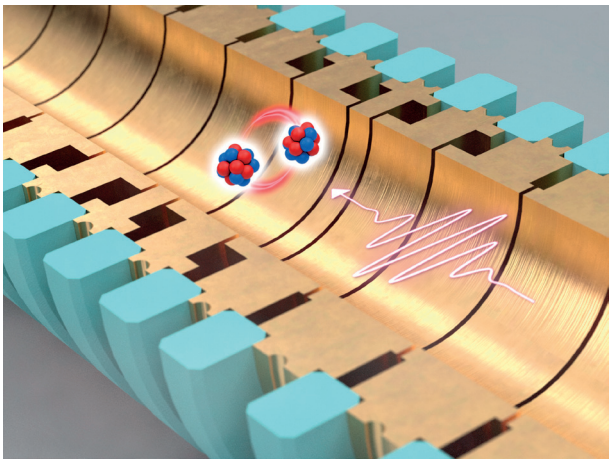
Megakelvin plasmas are ubiquitous in the Universe and drive its evolution together with gravitation. X-ray observatories already in space, and upcoming missions aim at diagnosing them. To do so, theoretical models have to be benchmarked by experiments simulating those conditions. Highly charged ions (HCI) of iron (Fe) are crucial for this: Near black holes, x-rays from Fe^{25+} ions are the last visible spectroscopic signal of matter, and soft x-ray lines from Fe^{16+} dominate in a multitude of other hot objects. Plasma diagnostics based on Fe^{16+} is an essential tool, in spite of having suffered from a puzzle over four decades: Astrophysical and laboratory data on its two strong transitions dubbed 3C and 3D showed intensity ratios incompatible with calculations. This casted doubt into the general methods needed for calculating many-electron ions. In a series of experiments [1,2], we performed measurements at PETRAIII (DESY) with a miniature electron beam ion trap (EBIT) storing Fe^{16+}. We reached the highest ever reported soft x-ray resolution, and revealed extended wings of the lines, containing hitherto unaccounted-for intensity. With unprecedented accuracy, these results confirmed advanced atomic structure theory calculations. Two independent, numerically massive calculations reached convergence and agreed mutually, as well as with the experiment. This finally solves the long-standing mystery, and proves that such state-of-the art calculations for complex many-electron systems including quantum electrodynamics corrections and relativistic mass shifts can be trusted at the 3% level.

References:

[1] S. Kühn et al. PRL 124, 225001 (2020), DOI: 10.1103/PhysRevLett.124.225001
[2] S. Kühn et al., 129, 245001 (2022), DOI: 10.1103/PhysRevLett.129.245001

Measurement of the bound-electron *g*-factor difference in coupled ions

From the beginnings of quantum electrodynamics, the electron's magnetic moment has played an extraordinary role for the understanding of the fundamental laws of atomic and subatomic physics. The precise determination of the *g*-factor, which expresses the magnetic moment relative to the Bohr magneton, has enabled stringent tests of the foundations of the Standard Model. By binding the electron to a highly charged nucleus, we are able to extend these tests to the regime of the strongest electromagnetic fields. With the ALPHATRAP experiment at MPIK we have pushed the precision to 11 significant digits, where a growing number of tiny, but fundamentally interesting effects becomes visible. At that point, unavoidable fluctuations of the magnetic field impede further progress. Recently however, we have achieved a conceptual breakthrough by crystallising two ions in the same trap (in this case two isotopes of hydrogenlike neon), which largely rejects magnetic field fluctuations. This new technique allowed us to directly determine the difference of *g* for the two ions with 13 digits sensitivity, 2 orders of magnitude better than ever before. At that level, exciting new effects, such as the QED recoil, can be observed. The measured value still agrees with the theoretical prediction even at that precision. This means the Standard Model not only, once again, passes with flying colours – but, beyond that, our result strongly bounds the properties of dark matter candidates.



Two hydrogenlike neon ions, one $^{22}Ne^{9+}$ and one $^{20}Ne^{9+}$, are trapped on a common crystal orbit around the trap centre. This way, both ions see exactly the same magnetic field, enabling us to measure the difference of their magnetic moments with utmost precision.

Reference:

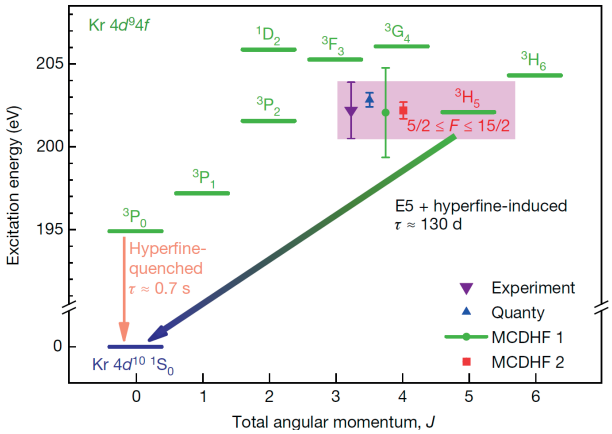
T. Sailer, V. Debierre, Z. Harman, F. Heiße, C. König, J. Morgner, B. Tu, A. V. Volotka, C. H. Keitel, K. Blaum & S. Sturm, Nature 606, 479–483 (2022), DOI: 10.1038/s41586-022-04807-w

Detection of metastable electronic states by Penning trap mass spectrometry

Modern clocks and frequency standards range from ensembles of neutral particles trapped in optical lattice clocks to individual, singly charged ions confined in Paul traps. With a fractional frequency accuracy of 10^{-18}, such clocks enable stringent tests of fundamental symmetries (for example, Lorentz invariance), geodetic measurements and searches for new physics and dark matter.

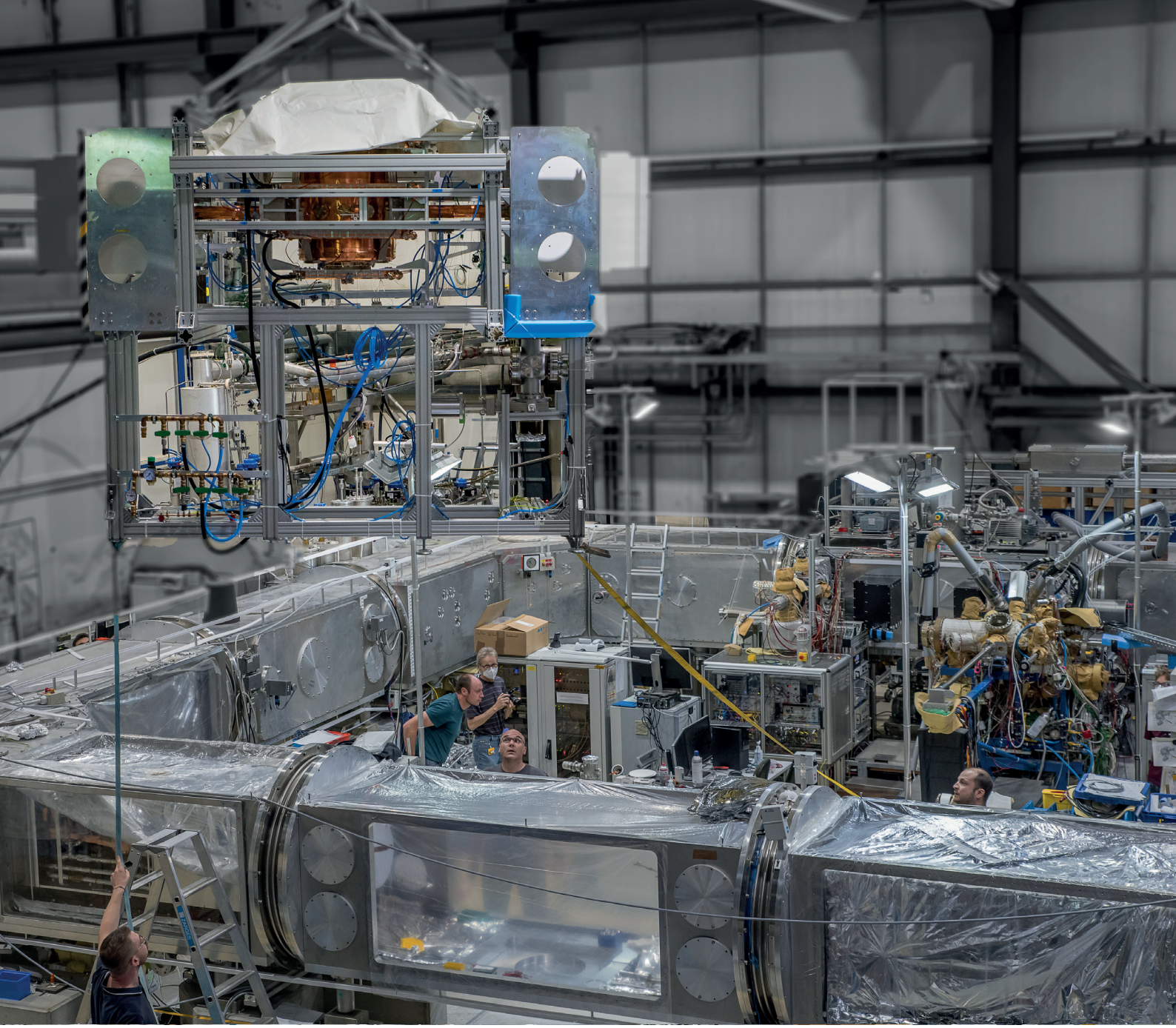
A further boost of precision is offered by next-generation atomic clocks based on highly charged ions (HCIs). The compact size of HCIs in comparison with atoms makes them less sensitive to external field fluctuations. Although electronic binding energies in HCIs typically amount to several kiloelectronvolts and inter-shell transitions usually appear in the x-ray region, there are also intra-shell fine and hyperfine transitions in the optical and ultraviolet range. Such transitions can be accessible to frequency combs and thus their frequencies can be measured very precisely.

However, insufficiently accurate atomic structure calculations hinder the identification of suitable transitions in HCIs and thus call for alternative methods of searching for such transitions. Recently we have demonstrated how high-precision Penning-trap mass spectrometry directly identifies a suitable clock transition in HCIs by measuring the mass difference between the atomic ground and a metastable state in rhenium ($^{187}Re^{29+}$), providing a non-destructive, direct determination of an electronic excitation energy. The result is in agreement with advanced calculations. We use the high-precision Penning-trap mass spectrometer PENTATRAP to measure the cyclotron frequency ratio of the ground state to the metastable state of the ion with a precision of 10^{-11}. With a lifetime of about 130 days, the potential soft-x-ray frequency reference at 4.96×10^{16} hertz (corresponding to a transition energy of 202 electronvolts) has a linewidth of only 5×10^{-8} hertz and one of the highest electronic quality factors (10^{24}) measured experimentally so far. The low uncertainty of our method will enable searches for further soft-x-ray clock transitions in HCIs, which are required for precision studies of fundamental physics.



Reference:

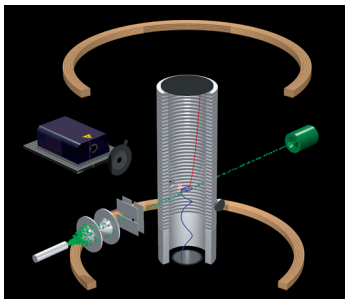
R. Schüssler et al., Nature 581, 42-46 (2020), DOI: 10.1038/s41586-020-2221-0



2.2 Atomic and Molecular Dynamics

The Cryogenic Reaction Microscope (CSR-ReMi) is transferred by crane from its nearby pre-assembly area, and guided by the team to its foreseen place in the Cryogenic Storage Ring (CSR), where it is now integrated for final checks before commissioning.

Reaction Microscopes and Absorption Spectrometers



Scheme of a reaction microscope.

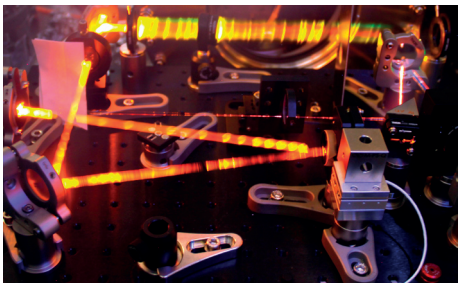
Reaction microscopes – "the bubble chambers of atomic and molecular physics" – have been developed and are continuously improved at MPIK. Ultra-short intense laser pulses or particle beams induce a break-up of simple molecules. The fragment ions and electrons are caught by means of electric and magnetic fields and recorded by large-area time- and position-sensitive detectors. From the reconstructed trajectories of the fragments, their complete momentum vectors, and thus the geometry and dynamics of the molecules before their break-up, can be determined ("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, built and recently integrated. It will be a key instrument for the worldwide unique possibilities to investigate slow and cold ions in the CSR.

Time-resolved attosecond absorption spectroscopy, a complementary technological in-house development, is based on the principle of Fraunhofer: shining a broadband ("white") light source through a medium reveals characteristic dark (absorption) lines in the spectrum. With dedicated vacuum apparatuses, the quantum dynamics of small atoms and molecules are investigated down to attosecond time scales, employing light from the visible to the extreme ultraviolet (XUV) and soft x-ray range, reflecting the coherent excited-state dynamics of the system.

Ultra-short Laser Pulses – the Microcosm in Extremely Slow Motion

How does a quantum system evolve in time and is it possible to visualise 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 fs = 10^{-15} s), while the electrons which mediate the chemical bond are even faster: here, attoseconds (1 as = 10^{-18} s) are the characteristic time scale.

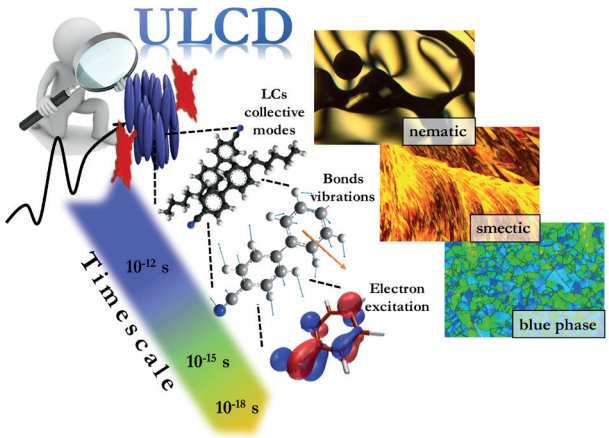
A key tool for time-resolved experiments are ultra-short intense laser pulses which are used to steer the atomic or molecular dynamics with extremely high precision. At MPIK's laser laboratories, phase-controlled laser pulses shorter than 5 femtoseconds at intensities of up to about 10^{16} W/cm^2 are available for experiments. Even shorter pulses of some attoseconds duration are generated by extreme non-linear optics. The resulting coherent high-harmonic radiation in the extreme UV range produces isolated as well as trains of attosecond pulses, which can be precisely timed relative to the broadband infrared to visible pulses of our laser systems, 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 in absorption spectroscopy setups and reaction microscopes, this allows for direct and time-resolved observation (and control) of nuclear and electronic quantum motions in chemical reactions.



Visualised beam path of an experiment for laser control of molecules.

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. Rotation of a molecule triggered by a laser pulse is used to measure the timing of the reaction that takes place in a second laser pulse. Such a "rotational clock" is a general concept applicable to sequential fragmentation processes in other molecules.

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. The broad-band spectrum of ultra-short laser pulses is used to detect the characteristics of the excitation processes within atoms and molecules – while their natural dynamics is disturbed by the intense fields of the laser. The electron pair of a helium atom can be specifically controlled in such a way that the energy levels are shifted for only fractions of the excited-state lifetime. In a molecule such as sulfur hexafluoride, the electronic exchange interaction, a pure quantum effect of a many-electron system, can be precisely controlled with a strong laser pulse on an ultra-fast time scale.



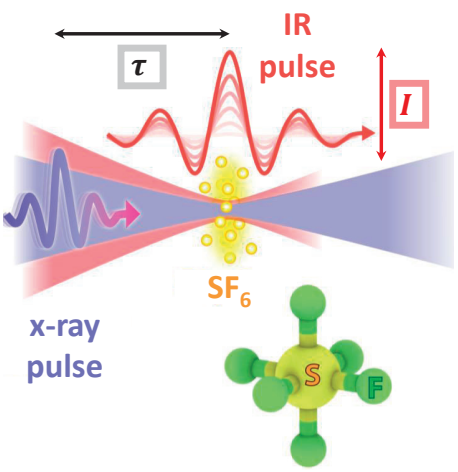
Research on ultra-fast liquid-crystal dynamics.

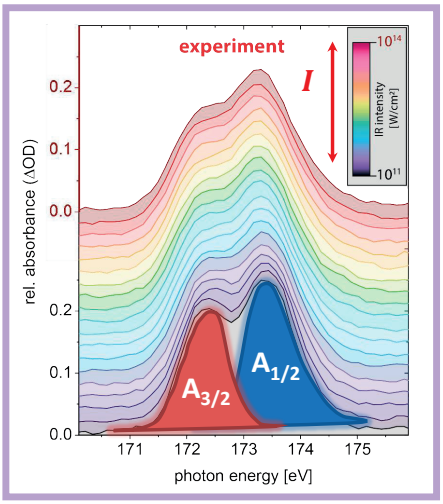
Experiments at the free-electron laser FLASH in Hamburg demonstrate strongly-driven non-linear interactions of ultra-short extreme-ultraviolet (XUV) laser pulses with atoms and ions. Here, powerful excitation of an electron pair in helium competes with the ultra-fast decay, which temporarily may even lead to population inversion. Resonant transitions in doubly charged neon ions were shifted in energy, and observed by XUV-XUV pump-probe transient absorption spectroscopy. Being specific to characteristic atomic transitions of core-level electrons, in such experiments the nuclear dynamics of atmospherically relevant molecules such as diiodomethane and oxygen can be time resolved.

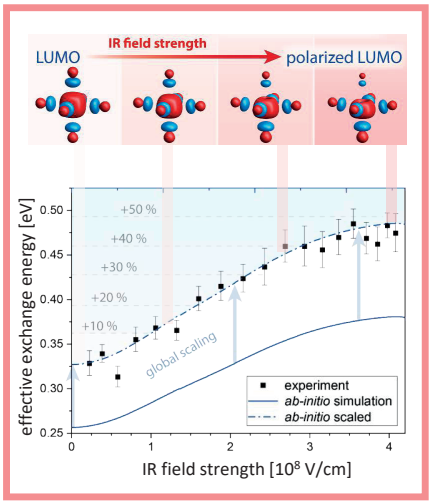
A new topic is ultra-fast liquid-crystal dynamics focusing on the understanding of such phenomena across LCs as phase transition from isotropic (liquid) to liquid crystal and solid phase. This investigation allows to define the degree of localisation in collective molecular dynamics excited with phase-stable THz pulses

Laser-controlled structural and electronic dynamics in SF_6

The laser-controlled structural and electronic dynamics of the SF_6 molecule is measured in the vicinity of the sulfur $L_{2,3}$ x-ray absorption edge with attosecond time-resolved absorption spectroscopy. The doublet lineshape of this transition (shown in the centre of the figure) connects a deeply bound (localised) core orbital with the (delocalised) valence orbitals of the molecule. Both quantum-mechanical spin-orbit and exchange interactions of the electrons play a role. Hereby the laser polarizes the valence orbitals of the molecule, which leads to an effective increase of the electronic exchange interaction between the valence and the core electrons in SF_6 [1], which is shown in the figure on the right. Femtosecond time-resolved x-ray absorption spectroscopy is further demonstrated to be sensitive to extremely small-scale molecular vibrations, which are quantified with a precision of only 14 femtometers [2].







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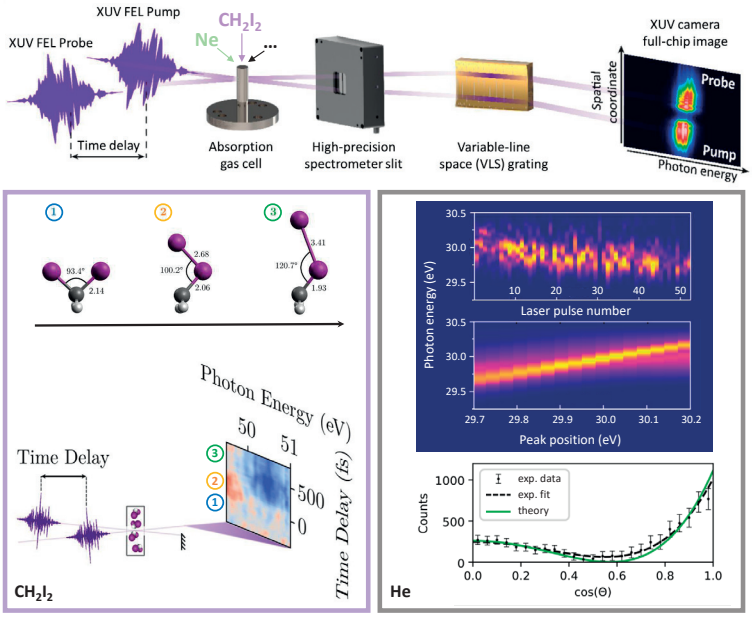
[2] P. Rupprecht et al., arXiv:2207.01290 [physics.chem-ph] (2022), DOI: 10.48550/arXiv.2207.01290

XUV-optical non-linear spectroscopy of small atoms and molecules at FLASH

A new beamline for time-resolved site-specific XUV-pump–XUV-probe transient absorption spectroscopy of atomic and molecular dynamics has been developed and successfully operated at the Free-Electron-Laser FLASH [1]. By measuring the time-delayed transmission through an absorbing neon (Ne) gas, the spectro-temporal profile of the FEL pulses can be quantified in situ [2]. The structural dynamics of diiodomethane (CH_2I_2) has been time resolved along a non-trivial dissociation pathway, both initiated and probed locally, resonant to transitions at the iodine atoms [3]. The apparatus for XUV-optical spectroscopy is now permanently installed behind the reaction microscope (ReMi) at Beamline FL26 at FLASH. This unique combination allows the coincident detection of ions, electrons and photons (spectra) on a single-shot level, enabling new experiments of fundamental AMO science and non-linear light-matter interaction. Sorting the spectral distribution of statistically fluctuating FEL pulses, the resonant photoelectron angular distribution of a two-photon-two-electron transition in helium has been resolved by high-resolution digital tuning within the much larger average FEL bandwidth [4].



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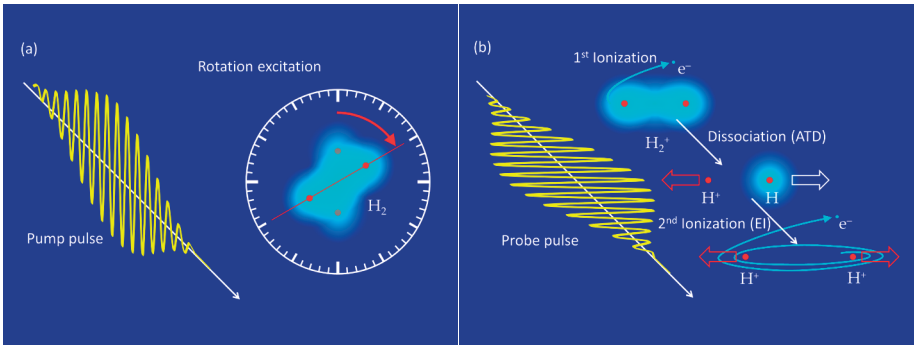
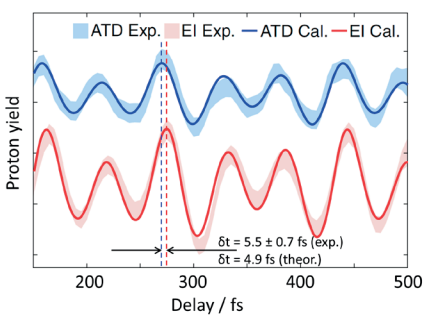
A molecular “rotational clock” for the observation of fragmentation mechanisms

How does a molecule break apart in an intense laser field, and what are the time scales of processes that contribute? To answer this question and to measure the timing of fragmentation steps for the model system of molecular hydrogen, H_2, the rotation of the molecule was used as an “internal clock”.

Essentially two fragmentation pathways contribute, the so-called “above threshold dissociation” (ATD) and “enhanced ionization” (EI). While the underlying mechanisms are reasonably well understood, the temporal sequence and the timing with respect to the first ionization step have not yet been measured. Both processes happen at different internuclear separations and they are both sensitive to the orientation of the molecular axis relative to the direction of the laser electric field – they happen most likely for parallel orientation. In our experiment, a weak femtosecond pump pulse excites the molecular rotation and a stronger probe pulse, which follows with a variable time delay, then ionizes the molecule and triggers the fragmentation (left figures). The two pulses are polarized perpendicular to each other. The experimental yields of ATD and EI events show an almost regular up and down, corresponding to the rotation of the molecule (right figure). In a closer analysis, however, a slight delay of approx. 5.5 fs is observed for EI compared to ATD. Using theoretical model calculations, further details can be extracted and the experimental results are very well reproduced. The “rotational clock” is a general concept that can be applied to fragmentation processes in other molecules as well.



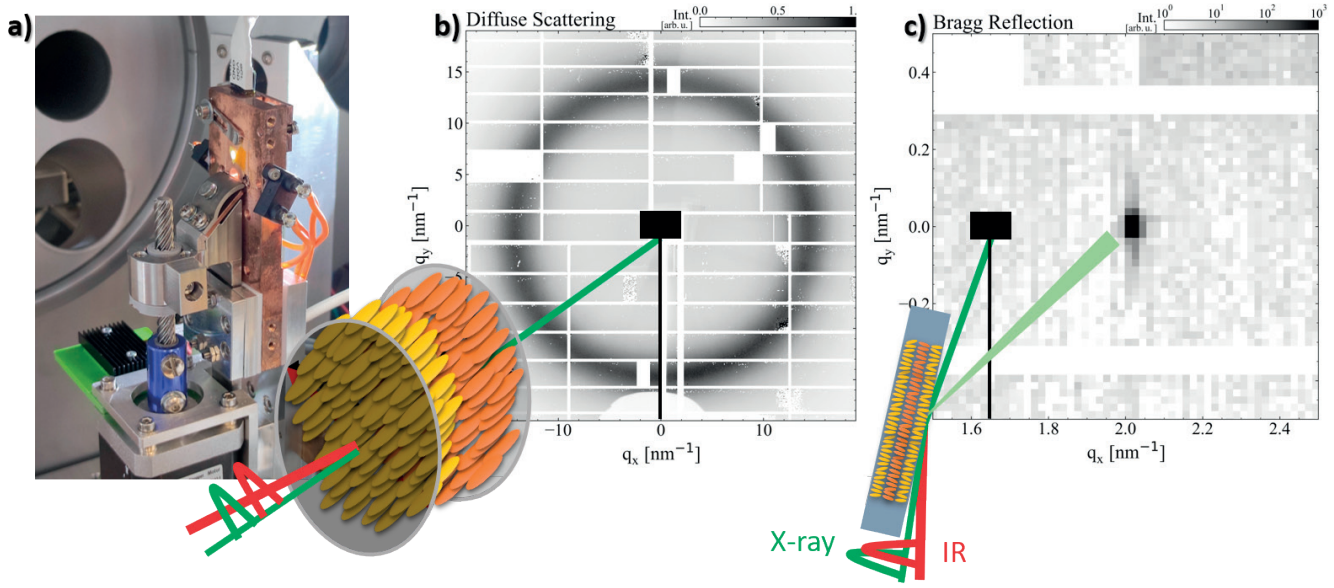


Reference:

Y. Mi et al.; Phys. Rev. Lett. 125, 173201 (2020), DOI: 10.1103/PhysRevLett.125.173201

Looking at liquid crystal dynamics from a hard x-ray point of view

Liquid crystals (LCs) are fluid-like liquids but they exhibit solid-like ordering at the molecular scale. To understand LC molecular dynamics at the picosecond time scale triggered via non-resonant excitation, we performed near-infrared pump (800 nm, 50 fs), hard x-ray diffraction probe (100 fs, 8.9 keV) on free standing films of octylcyanobiphenil (8CB) which presents a layered structure (smectic A) in the range 22 to 34°C. The key idea is to follow the impact on the 8CB x-ray diffraction pattern when low-frequencies collective molecular vibrations are laser-induced, with the aim to look at the short-range interaction among neighbouring molecules. A dedicated holder was built in collaboration with the workshops at the institute (panel a). Two different geometries were used: transmission where the diffuse scattering ring (panel b) is sensitive to intermolecular distances, and Bragg diffraction (panel c) sensitive to the smectic A inter-layer spacing.



Atomic and Molecular Collisions – Billiard Game with Quantum Balls

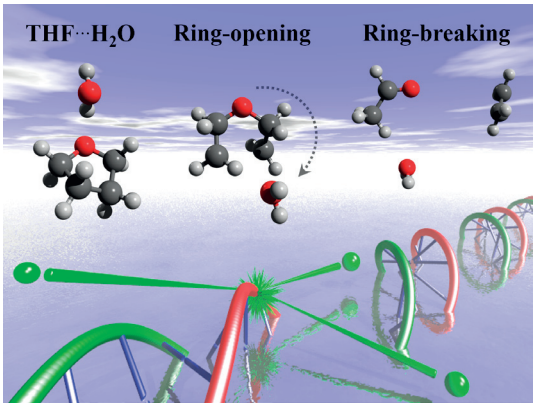
Research on correlated quantum dynamics represents one of the great challenges in contemporary science. Researchers at the MPIK explore quantum dynamics on a fundamental level, starting from a limited number of few interacting particles in atoms and molecules, and extending to more complex finite quantum 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.

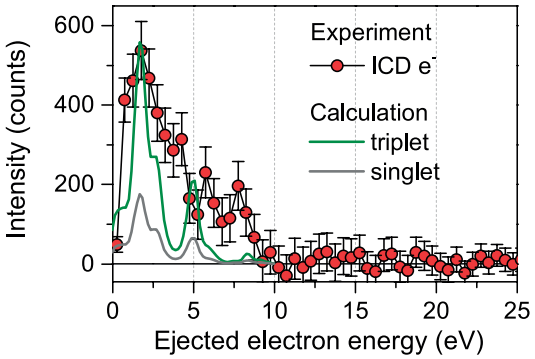
Ultra-cold Dynamics – Investigating Exotic Quantum Gases

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.

Amplification of electron-induced lesions in smallest condensed aggregates of organic molecules

Research on the elementary interactions of energetic radiation with organic and biologically relevant molecules can advance applications in medical treatment like radiation therapy and help to improve its effectiveness. Using multi-particle imaging spectrometers (reaction microscopes) we study the impact of electrons on molecular pairs (dimers) and larger clusters and could find hitherto unrecognised reaction pathways. For the tetrahydrofuran-water dimer (THF···H_2O) indicated in the upper figure we observed efficient molecular ring break for ionization of even the least bound electron where a single tetrahydrofuran monomer would stay intact [1]. We showed that the rearrangement of the water molecule bound to the THF ion releases sufficient vibrational energy to overcome the relevant reaction barriers. Similarly, in ionized methanol dimers new and fast dissociation channels were found. These outpace and, therefore, suppress slow channels occurring in the monomer like the "roaming" of an abstracted H_2 molecule in the vicinity of the methanol [2]. Lastly, we identified an intermolecular energy transfer and a subsequent Coulomb explosion reaction in weakly bound aromatic benzene pairs [3]. This intermolecular Coulombic decay (ICD) can be a blueprint for similar reactions across the hydrogen bonds of DNA base pairs or in folded protein chains. The lower figure shows the spectrum of the emitted ICD electrons. All observations were supported by theory and ab initio molecular dynamics simulations.





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[1] E. Wang et al., Nat. Comm. 11, 2194 (2020), DOI: 10.1038/s41467-020-15958-7

[2] E. Wang et al., Phys. Rev. Lett. 126, 103402 (2021), DOI: 10.1103/PhysRevLett.126.103402

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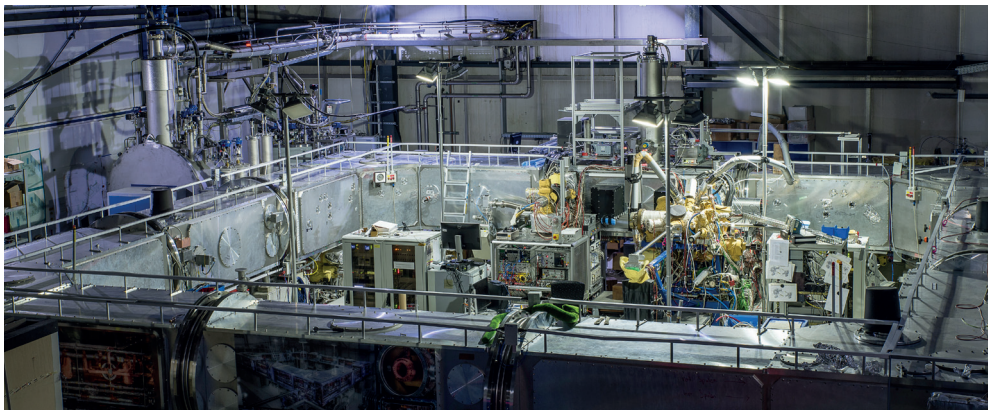
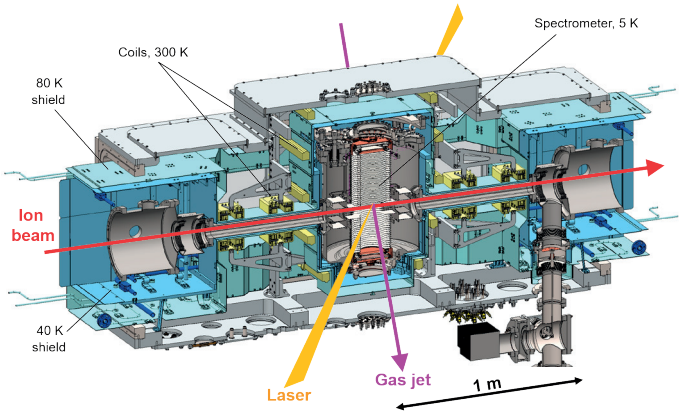
The Cryogenic Storage Ring

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. Besides beam diagnostics, instrumentation comprises particle detectors, a laser system, and the novel reaction microscope. The innovative mechanical concept of the CSR was developed and realised in close cooperation with MPIK's engineering design office and precision mechanics shop.

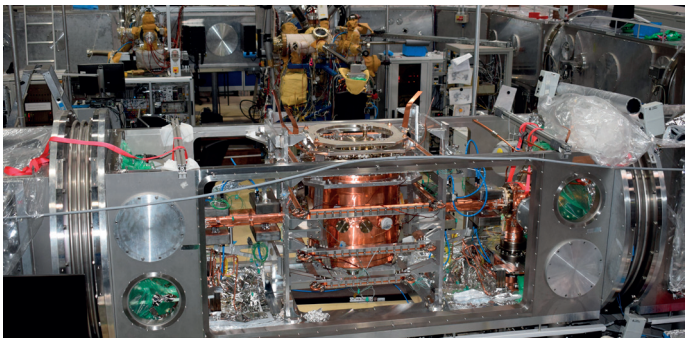


The cryogenic storage ring CSR with the electron cooler.

The CSR-ReMi - a reaction microscope for molecular break-up studies in the CSR



Design of the CSR-ReMi with its main components including projectile and target beams.



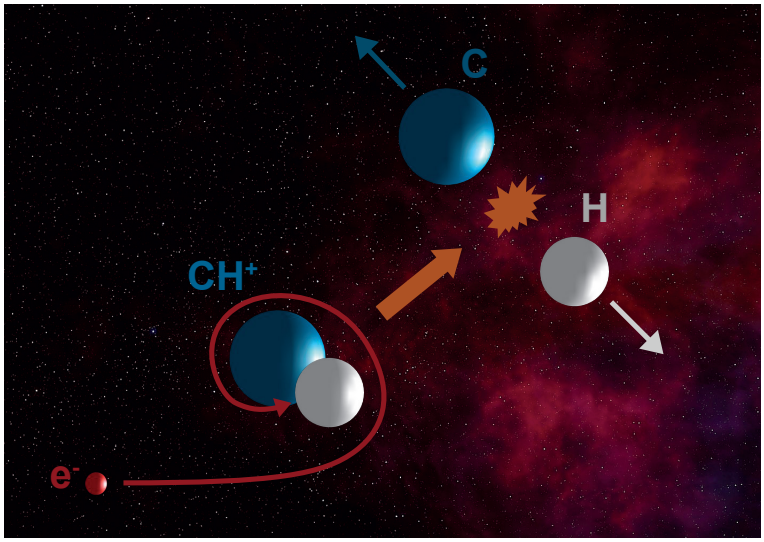
Photograph during CSR-ReMi installation into the storage ring.

Reaction microscopes (ReMi) are combined electron and ion spectrometers for energy and angular resolved detection of fragments emerging from individual collision events. In order to use this powerful technique for experiments with beams of slow and cold molecular ions and clusters inside the electrostatic cryogenic storage ring CSR, we designed a dedicated in-ring spectrometer, the CSR-ReMi. It is the first cryogenic reaction microscope worldwide (see figures).

The implementation of the fully constructed machine into the CSR started in May 2022. After its integration, which is expected to be finalised at the beginning of 2023, first test experiments with stored cold molecular ions are envisaged for spring 2023. Possible experiments include reactions like electron or even proton transfer, photodetachment or collisional ionization, and molecular break-up reactions of any kind.

The CSR-ReMi will significantly widen the spectrum of scientific applications at the CSR infrastructure. As a next step, in combination with a high-power femtosecond laser, time-resolved experiments on state-prepared molecules using established pump-probe techniques are planned. In all cases, the CSR-ReMi will be instrumental to unravel the dynamics of molecular fragmentation and relaxation processes.

Laboratory Astrophysics – the Chemistry of Space



Dissociative recombination of a methylidine ion in interstellar molecular clouds. A free electron traversing through space is attracted and captured by the positively charged CH^+ ion. The energy released in this process blows the molecule apart into a carbon (C) and hydrogen (H) atom.

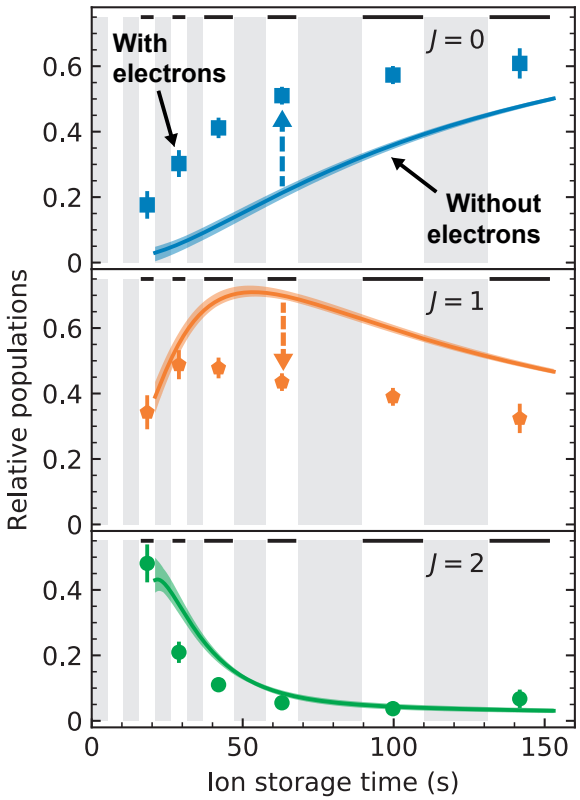
The gas-phase chemistry in interstellar clouds is driven by reactions involving ions and radicals which are created in collisions with photons and cold electrons. Here, the H_3^+ molecular ion plays a key role. The break-up of molecules after capture of an electron, called dissociative recombination, can be studied in detail in the cryogenic storage ring CSR. There, 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 neutral-atom beam setup at the CSR combines ground-state atoms with cold molecular ions, and thus provides access to this largely unexplored class of processes under true interstellar conditions.

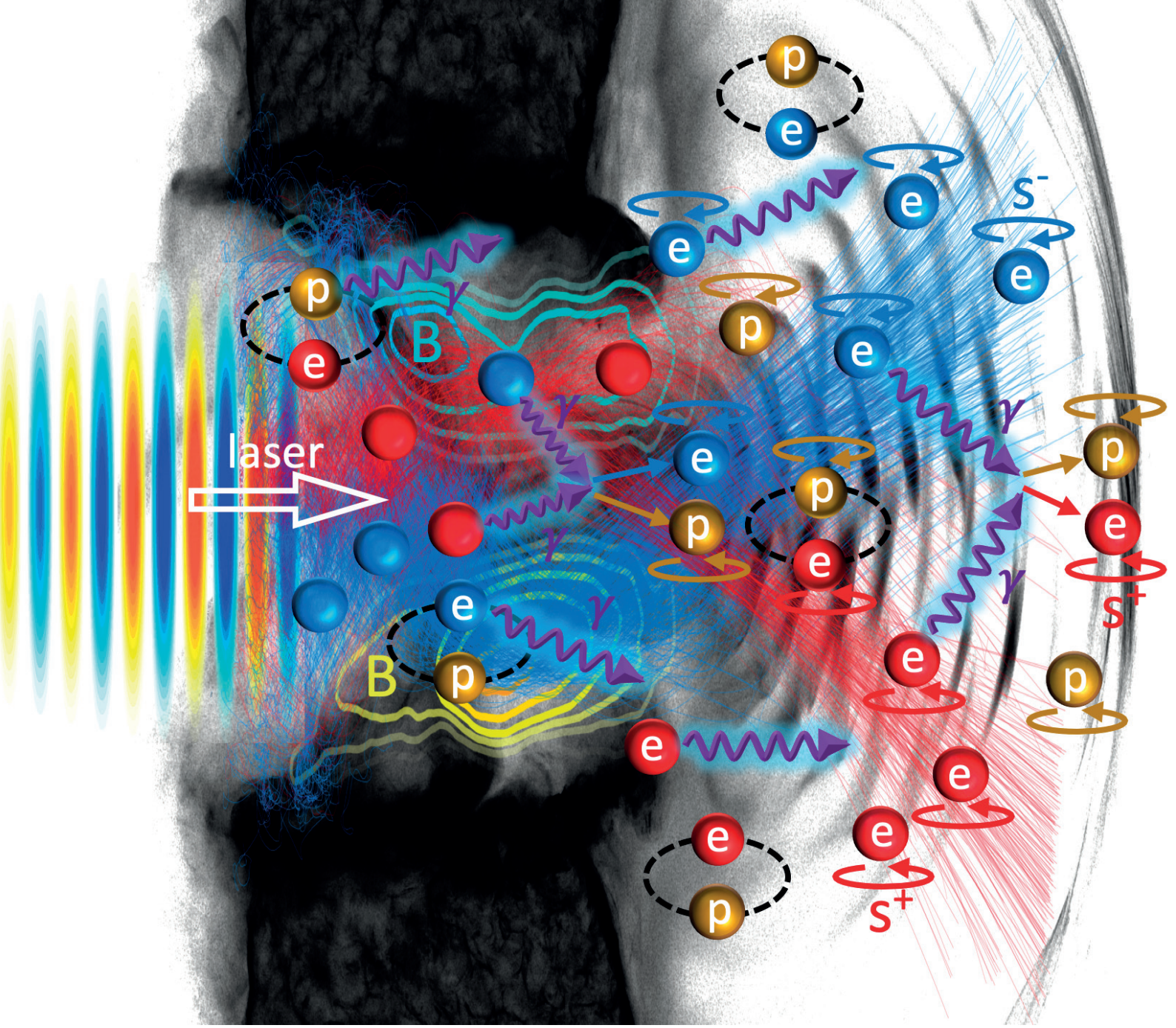
Laser spectroscopic measurements on molecular ions in the CSR using the electron cooler give new insight into quantum transitions modified by collisions with surrounding electrons under space-like conditions.

Laser probing of the rotational cooling of molecular ions by electron collisions

A lone molecule free in cold space will cool down by slowing down its rotation – it will spontaneously lose its rotational energy in quantum transitions, typically only once in many seconds. This process can be accelerated, slowed down, or even inverted by collisions with surrounding particles. In an experiment at the cryogenic storage ring (CSR), the rate of rotational state transitions due to encounters between molecules and electrons was measured by bringing isolated charged molecules in contact with electrons under controlled conditions at about 26 K. Thus, they could make this rate – only known by complex calculations so far – high enough to be quantitatively determined in an experiment at last. To this end the occupation of rotational energy levels in methylidine ions (CH^+) was probed by laser spectroscopy during up to 10 minutes of storage in CSR. The time evolution of the CH^+ rotational population has been acquired without the electron interactions, i. e., involving only radiative cooling in the low radiation field of CSR. This was then compared to the case with electron interactions to yield the pure electron-induced rate. The result represents the first experimentally derived rates for electron-induced rotational molecular transitions, while the derived values match well the so far unverified theoretical calculations. The electron-induced rates of rotational level changes are crucial in analysing, e. g., the faint signals of molecules in space detected by radio telescopes, or in predicting level-dependent chemical reactivity in dilute cold plasmas.



Reference:
Á. Kálosi et al., Phys. Rev. Lett. 128, 183402 (2022), DOI: 10.1103/PhysRevLett.128.183402



2.3 Matter in Extreme Fields

Extremely intense laser pulses impinging on matter: Electrons are accelerated resulting in gamma photons, magnetic fields, radiative reaction and lepton pairs being generated. Important information on the internal dynamics and generated electromagnetic fields is gained from the ejected lepton polarizations.

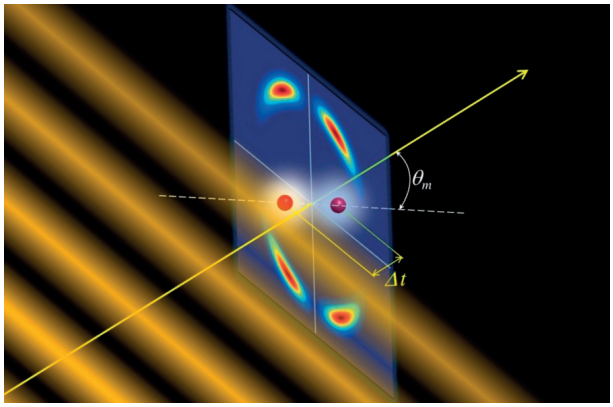
Matter in Strong Laser Fields – at the Frontiers of Feasibility

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 these 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 involving often also quantum electrodynamics and nuclear physics.

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. However, the accurate description of the sub-barrier dynamics can significantly modify this simple picture. Recent studies have identified a new pathway of strong-laser-field-induced ionization of an atom which takes place via recollisions under the tunnelling barrier. The interference of the direct and the under-the-barrier recolliding quantum orbits is shown to induce a measurable shift of the peak of the photoelectron momentum distribution which can be associated with a time delay. Based on a simple model without critical approximations, we showed that other conflicting approaches for the tunnelling time delay, as e. g., the backpropagation method, can be reconciled with our sub-barrier recollision theory. We also investigated the role of quantum features in the direct vicinity of the tunnelling exit and explained why not all experiments could find a non-vanishing tunnelling time.

The non-dipole description of the sub-barrier dynamics is also very important because it yields a non-zero momentum of the electron along the laser propagation direction at the tunnel exit. This modifies the photon momentum sharing in strong-field ionization as confirmed by recent experiments of Reinhard Dörner. Further studies revealed the counterintuitive nature of the Coulomb field of the atomic core for the non-dipole sub-barrier dynamics. Despite its attractive nature, the sub-barrier Coulomb field increases the photoelectron non-dipole momentum shift along the laser propagation direction. The scaling of the effect concerning the principal quantum number and angular momentum of the bound state indicate that the signature of Coulomb-induced sub-barrier effects can be identified in the asymptotic photoelectron momentum distribution via a comparative study of the field-dependent longitudinal momentum shift for different atomic species.

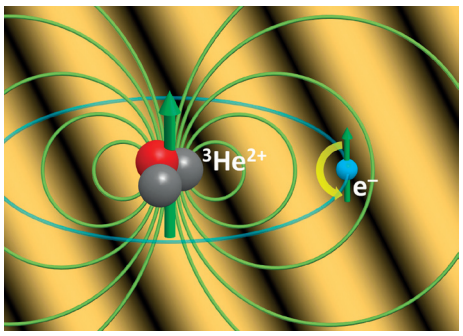
In addition, the non-dipole strong-field ionization time delay in a molecule due to the finiteness of the light speed manifests itself in the longitudinal momentum distribution, featuring double-slit interference (see figure), which efficiently encodes the molecular structure and laser parameters. The delay depends essentially on the tunnelling exit distribution rather than the molecular bond length – with corresponding consequences for the interference patterns.



Ionization of a diatomic molecule in an intense laser field tilted by an angle θ_m to the laser beam. The light wave (yellow) hits the two atoms of the molecule with time delay (Δt). The molecule acts as double-slit creating the interference structure in the photoelectron distribution (blue).

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.

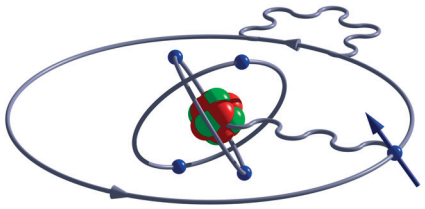


Schematic view of the $^3He^+$ ion's external and internal magnetic interactions. Background: microwave radiation.

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 laser-generated relativistic plasma. Here, a first-principle derivation of the underlying equations has been put forward.

Very strong fields also prevail in the vicinity of heavy nuclei. 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 binding energy or magnetic moment of electrons. On the one hand, comparison with precision experiments permits validation of QED predictions, while on the other hand theory helps to determine magnetic dipole moments like for $^3He^{2+}$ or natural constants like the electron mass: its value became by an order of magnitude more accurate.

Precision theory of ionic quantum dynamics



In a highly charged ion, besides the mutual relativistic interaction of electrons (in blue), QED effects such as the self-interaction of electrons via quantum fluctuations of the photon field (wave line) have to be taken into account, together with effects arising from nuclear structure.

We predict the properties of highly charged ions (HCI) to high precision. One purpose of our large-scale modelling is to support or guide local experiments with trapped and stored ions. In an experiment with the PENTATRAP Penning-trap setup, a long-lived, excited electronic state in a Re ion has been determined measuring the mass difference of the ion in its ground and excited states and using Einstein's $E = mc^2$ formula [1]. Our atomic theory helped to identify the corresponding levels and provided the lifetime of the metastable state. Such a non-destructive determination of a transition energy is anticipated to help the discovery of HCI transitions suitable for constructing future atomic clocks. Similar experiments, together with our calculations, yielded the Q value of the β-decay of ^{187}Re, relevant for the determination of the neutrino mass [2].

Another property which can be measured to high accuracy is the magnetic moment of the electron bound in HCI. We have put forward the use of this quantity in new physics searches [3]. This idea was implemented in an experiment performed with the ALPHATRAP setup, in which the difference between the magnetic moments of two Ne isotopes was measured with a remarkable precision [4]. The combination of theory and experiment provided a novel way of proving the existence of a hypothetical fifth force. Furthermore, our theory also enables to extract the magnetic properties of ^3He from high-precision experimental data, relevant for the calibration of new nuclear magnetic resonance probes [5].

Besides HCI, anions are also excellent systems for benchmarking precision atomic theory. Long-lived metastable states of the Si ion were studied at the cryogenic storage ring by selective photodetachment. The measured lifetimes of the extremely weekly bound excited states are in a very good agreement with our predictions [6].

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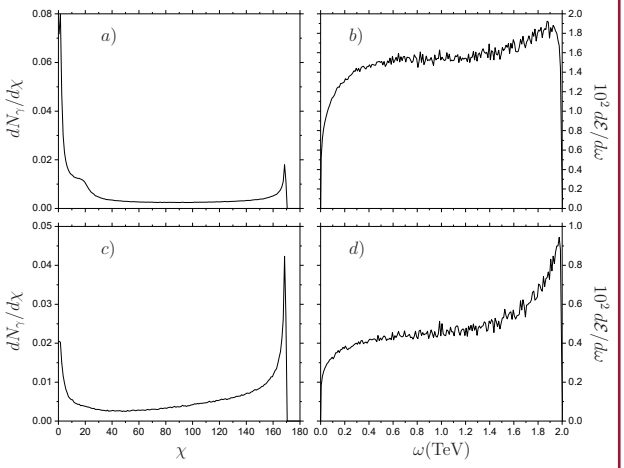
Testing strong-field QED close to the fully non-perturbative regime using aligned crystals

Strong-field QED is the theory of quantum processes occurring in the presence of intense background electromagnetic fields. The scale of the field strength is determined by the so-called critical field of QED $F_{cr} = m^2c^3/\hbar|e|$, which is of the order of 10^{16} V/cm (10^{13} G) in the electric (magnetic) case. The interaction between the background field and the electrons/positrons has then to be taken into account exactly in the calculations, whereas the interaction between electrons/positrons and photons can be commonly treated perturbatively. However, the Ritus-Narozhny conjecture states that if electrons/positrons experience field strengths of the order of $F_{cr}/\alpha^{3/2} \approx 1600\ F_{cr}$ in their rest frame, with $\alpha \approx 1/137$ being the fine-structure constant, their effective coupling with photons becomes of the order of unity. In this respect, QED would become a strongly coupled theory. We have shown that channelling radiation by ultra-relativistic electrons with energies of the order of a few TeV on thin tungsten crystals allows one to test the predictions of QED close to this fully non-perturbative regime by measuring the angularly resolved single-photon intensity spectrum. The proposed setup features the unique characteristics that essentially all electrons undergo at most a single photon emission and experience at the moment of emission and in the angular region of interest the maximum allowed value of the field strength, which at 2 TeV exceeds F_{cr} by more than two orders of magnitude in their rest frame (see figure, where θ_c is the critical channelling angle, i. e., the maximum angle between the initial electrons' momentum and the crystal atoms lines such that the electrons remain channelled).



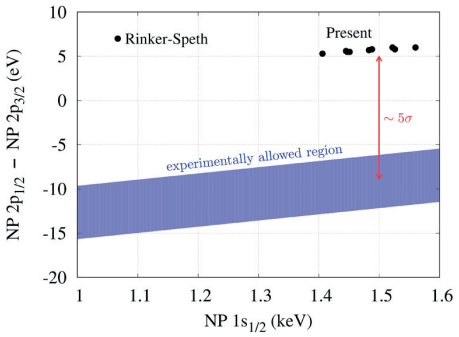
Left panels: Emitted photon number distributions as functions of the electron quantum non-linearity parameter χ (i. e., the crystal electric field in the rest frame of the electrons in units of F_{cr}) at the moment of emission, accounting for all emitted photons (a) and for the photons emitted with an angle larger than $\theta_c/2$ with respect to the z axis (c). Right panels: Average energy emitted per unit of photon energy, accounting for all emitted photons (b) and for the photons emitted with an angle larger than $\theta_c/2$ with respect to the z axis (d).

Reference:

A. Di Piazza, T. N. Wistisen, M. Tamburini, U. I. Uggerhøj, PRL 124, 044801 (2020), DOI: 10.1103/PhysRevLett.124.044801

Muonic fine-structure anomaly

In terms of the Standard Model, a muon is the second-generation lepton, identical to an electron except for its mass, and the interaction with other charged particles can be described by QED. Being captured by a nucleus, it forms a muonic atom, where the Bohr radius of muonic orbitals is 207 times smaller than that in electronic atoms, which causes high importance of QED effects and enhanced sensitivity to nuclear structure. A combination of the theoretical predictions for the energy levels and experiments measuring the muonic atomic spectra enabled the determination of nuclear parameters like charge radii, electric quadrupole and magnetic dipole moments. However, for over 40 years there has been a disagreement between theory and experiment, called muonic fine-structure anomaly.



Theoretical values (dots) of the nuclear polarization correction for muonic zirconium in relation to the experimentally allowed range for Δ2p as a function of the same contribution for 1s state.

Accessing the nuclear parameters is only possible with a thorough inclusion of the nuclear theory. In this respect, the most challenging effect to describe is the intricate interplay between muonic and internal nuclear degrees of freedom, which is known as nuclear polarization (NP) effect. First, the effects of nuclear excitations on atomic properties were described in a field-theoretical framework. Then, the nuclear ground state has been obtained from the Skyrme-Hartree-Fock procedure. Finally, complete nuclear excitation spectra are computed by means of the random-phase approximation and incorporated into calculations of the nuclear-polarization corrections to energy levels of muonic atoms [1]. The calculation uncertainty has been estimated by analysing the nuclear model dependence. The obtained results were applied to the long-standing problem of the fine-structure anomalies in heavy muonic atoms. We have considered muonic zirconium, tin and lead atoms. Our results [1] show that in all cases the nuclear polarization effect is unlikely to be responsible for the anomaly, contrary to what has been believed for over 40 years. Our recent rigorous predictions for self-energy corrections evaluated the leading-order QED predictions. Finally, excluding the long-suggested QED solutions to the puzzle [2], stimulates the search for other explanations, including physics beyond the Standard Model.

References:

[1] I. A. Valuev, G. Colò, X. Roca-Maza, C. H. Keitel, and N. S. Oreshkina, Phys. Rev. Lett. 128, 203001 (2022), DOI: 10.1103/PhysRevLett.128.203001

[2] N. S. Oreshkina, Phys. Rev. Research 4, L042040 (2022), DOI: 10.1103/PhysRevResearch.4.L042040

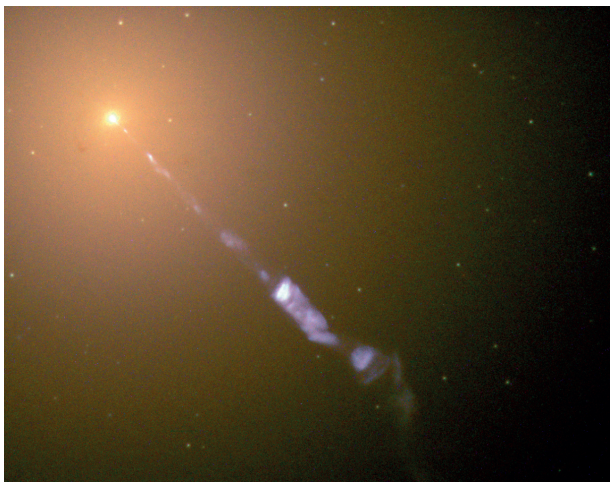
The theoretical calculations also help to determine nuclear properties from the measured data, and to set new limits for new physics and dark matter. A further motivation of such studies is that measurements of the magnetic moments of highly charged ions are anticipated to yield a new independent value of the fine-structure constant, i. e., the fundamental constant defining the strength of any kind of electromagnetic interactions in the Universe.

A long-standing problem of fine-structure anomalies in muonic atoms is revisited by considering level splittings in various muonic atoms. The results for μ-^{208}Pb suggest that the resolution to the anomalies is likely to be rooted in some previously unaccounted-for contributions or even more exotic explanations, including physics beyond the Standard Model.

Our relativistic atomic structure calculations have also contributed to the resolution of a conundrum that has puzzled astrophysicists for four decades. The intensity of two important, strong x-ray lines of a certain highly charged iron ion in astrophysical data and in laboratory experiments did not agree with predictions, obscuring the interpretation of astronomical observations. A recent EBIT measurement at the high-brilliance PETRA III synchrotron facility (see p. 30) finally agrees with our large-scale calculations and with other modern theoretical results.

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

With increasing intensities of laser systems, 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 to possibly preventing the generation of extremely strong laser pulses and we have put forward means to prevent those via suitably chosen focus areas. In particular, various concepts have been developed to generate polarized intense lepton and gigaelectronvolt (GeV) gamma beams. They are based on spin-dependent radiative reaction and are likely to find applications in high-energy, solid-state and astrophysics. As another example, the relativistic spin dynamics of electrons in extreme fields accompanying strong-field QED processes provides a new avenue for in-situ probing the plasma dynamic characteristics in ultra-strong laser fields. The ejected electron spin provides a new degree of freedom to extract information on the structure and magnitude of different components of the transient plasma fields, as well as to test the instability character in the plasma.



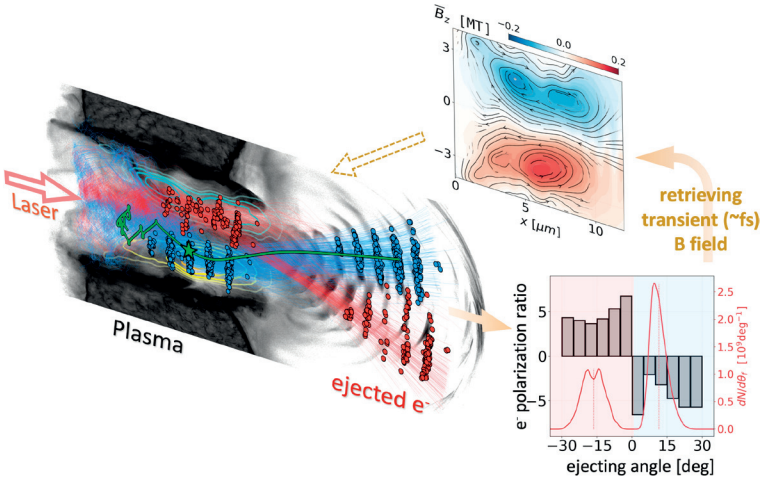
Black hole-powered jet of electrons and sub-atomic particles streams from the centre of galaxy M87 (Hubble).

Already to date, highly intense laser fields enable the acceleration of particles to energies up to the order of 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 ultra-relativistic lepton beams consisting of electrons and positrons in equal amounts as well as gamma rays. Most recently, an international team lead by MPIK researches has put forward a novel theoretical concept by showing that a pulsed, ultra-relativistic electron beam crossing a sequence of thin aluminium foils both self-focuses, therefore increasing its density, and efficiently yields a collimated gamma-ray pulse with more photons per unit volume than electrons in a solid. Such extremely dense beams enable physics studies of matterless photon-photon interactions ranging from matter-antimatter creation, light-by-light scattering, and searches of possible new physics in the interaction of photons with yet undiscovered particles, to laboratory astrophysics studies. Here, dense neutral relativistic jets of electron-positron pairs are generated from the intense gamma-ray pulse. In turn, these electron-positron jets enable unprecedented studies of the collective processes that are thought to shape high-energy astrophysical environments such as gamma-ray bursts and blazar jets, thus providing insights into these exotic systems.

Plasma self-diagnostics based on particles' polarization information

Ultra-relativistic plasma is an extreme state associated with violent gamma-photon emission, radiation reaction, and radiative spin polarization effects. Such an energetic and overdense plasma state drastically evolves at a time scale as short as a few femtoseconds, which disables conventional diagnostic methods relying on the external optical probe or changed particle radiography. Recently, we proposed a novel idea to utilise the polarization information of spontaneously ejected electrons or gamma-ray photons to retrieve the in-situ plasma dynamics.

By employing the unsymmetric electrons' spin-polarization signal, a non-trivial transient self-generated plasma magnetic island structure accompanied with current vortexes could be identified, see figure [1]. The result manifests that the spin signal offers a new degree of freedom to retrieve the structure and magnitude of the transient plasma magnetic fields. On the other side, the angular polarization pattern of emitted gamma photons could facilitate to decipher the electrons' in-situ acceleration status inside the plasma channel. In particular, the acceleration or deceleration status can be detected via the photons' polarization [2]. Also plasma instabilities can be characterised via the outgoing electrons' polarization state [3].



Left: spin-polarized electrons are ejected from the plasma irradiated by an intense laser pulse. Lower: angular distribution of e⁻ spin-polarization. Upper: identified transient self-generated plasma magnetic fields.

The results open the door for efficient plasma diagnostics based on spin polarization, which inherently exists in ultra-relativistic plasma phenomena, e. g. in cosmic ray radiation, stellar object evolution, and laser/plasma interaction.

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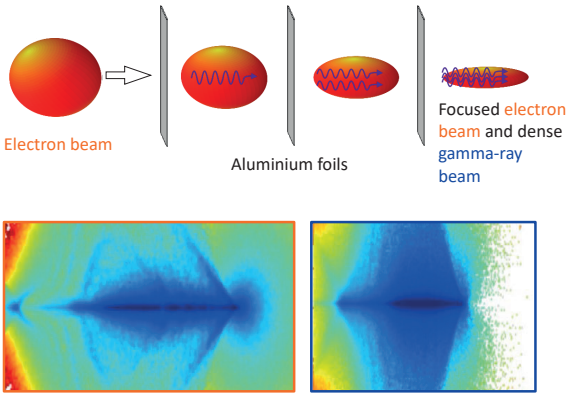
[1] Z. Gong, K. Z. Hatsagortsyan, C. H. Keitel, Phys. Rev. Lett. 127, 165002 (2021), DOI: 10.1103/PhysRevLett.127.165002
[2] Z. Gong, K. Z. Hatsagortsyan, C. H. Keitel, Phys. Rev. Res. 4, L022024 (2022), DOI: 10.1103/PhysRevResearch.4.L022024
[3] Z. Gong, K. Z. Hatsagortsyan, C. H. Keitel, Phys. Rev. Lett. in press (2022), arXiv:2212.03303 [physics.plasm-ph]

Generation of extremely dense gamma-ray pulses in beam-beam and beam-multifoil collisions

The generation of high-energy, dense and collimated photon beams is of great interest both to fundamental and applied research. With experimental implementation ongoing at the FACET-II accelerator facility at Stanford, under our division's leadership in [1] it was demonstrated with fully 3D simulations that a pulsed, ultra-relativistic electron beam crossing a sequence of thin aluminium foils both self-focuses and efficiently yields a collimated gamma-ray pulse with more photons per unit volume than electrons in a solid [1]. After passage of 20 foils, more than 30% of the electron beam energy is converted to gamma rays. This occurs because the strong electromagnetic field accompanying the ultra-relativistic electron beam is "back-reflected" as the beam crosses the foil surface. Similarly to an electromagnetic wave colliding with a mirror, at the foil surface the total electric field acting on the beam is nearly zero, while the total magnetic field is nearly doubled. This strong azimuthal magnetic field focuses the electron beam radially and triggers collimated high-energy photon emission. Efficient high-energy photon production with the possibility of precision studies of strong-field QED has also been demonstrated in beam-beam interaction [2].



Schematic setup (upper panel) and density distribution of the generated gamma-ray pulse (blue frame) and of the focused electron beam (orange frame) after the electron beam has crossed 16 consecutive aluminium foils.

Extremely dense beams enable physics studies of matterless photon-photon interactions such as light-by-light scattering and searches of possible new physics in the interaction of photons with yet undiscovered particles [3]. Dense gamma-ray beams also pave the way to matter-antimatter creation from light, as well as relativistic laboratory astrophysics studies. Here, dense neutral relativistic jets of electron-positron pairs are generated from the dense gamma-ray pulse.

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[2] M. Tamburini and S. Meuren, Phys. Rev. D 104, L091903 (2021), DOI: 10.1103/PhysRevD.104.L091903
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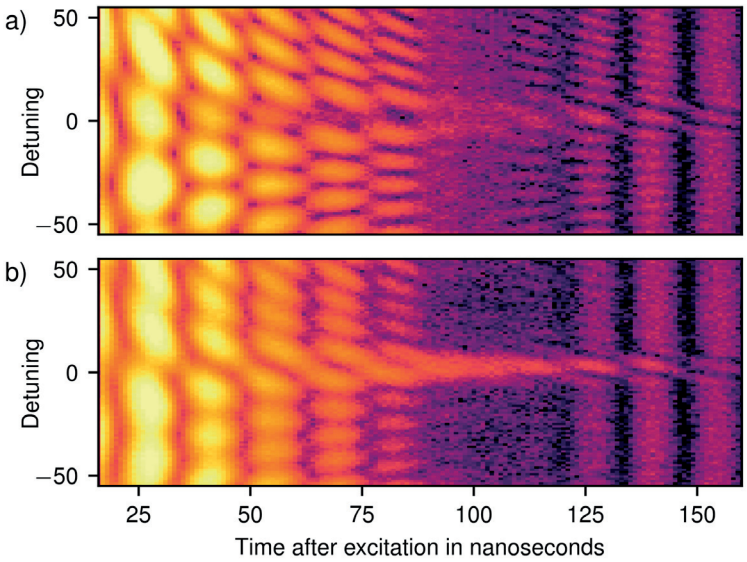
Extreme Light-Matter Interaction – Precisely Controlling and Probing Nuclear Transitions



Observed x-ray interference structures encoding the nuclear dynamics, as function of time after excitation and detuning between control field and target nuclei. (a) Measurement data for the case of enhanced emission, (b) for the case of enhanced excitation.

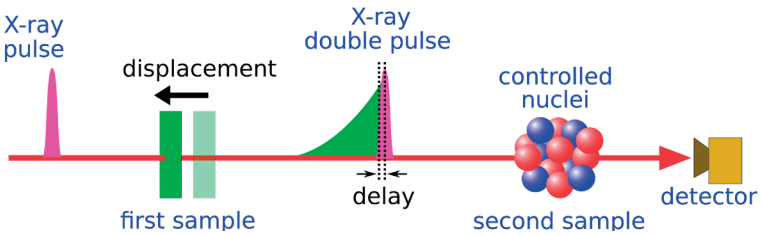
Quantum optics with x-ray light emerged in the last years as a new field. Of particular interest are certain atomic nuclei that only interact with x-rays with an extremely well-defined photon energy, due to an effect discovered by Rudolf Mößbauer at the precursor institute of MPIK in 1958. Spectroscopy – 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.

Using suitably shaped x-ray light, nuclear excitations have been coherently controlled for the first time – with a temporal control stability of a few zeptoseconds. This forms the basis for new experimental approaches for the engineering of complex nuclear quantum states and for the exploration of time-dependent phenomena with nuclei.

In parallel, an ab-initio theory has been developed for thin-film x-ray cavities with Mössbauer nuclei, a key platform for exploring nuclear quantum dynamics. With this theory, the as-yet open inverse problem of directly determining cavity structures providing a desired quantum optical functionality has been tackled.

Coherent x-ray optical control of nuclear quantum dynamics

Modern experiments on quantum dynamics can control the quantum processes of electrons in atoms to a large extent by means of laser fields, often based on coherence and interference phenomena. However, towards energies of hard x-rays, the intrinsic broadening of electronic resonances due to competing electronic processes limits the lifetimes of superpositions of atomic states, thereby rendering such control challenging. Resonances in Mössbauer nuclei are distinct from electronic x-ray resonances by their exceptionally narrow line widths, owing to the recoilless absorption and emission of photons. The narrow width has the advantage of desirably long coherence lifetimes, but on the other hand severely limits the possibility to strongly drive or control nuclear x-ray transitions even at modern x-ray sources. This raises the question, if the nuclear dynamics can coherently be controlled with suitably shaped x-ray fields. In Ref. [1], we have demonstrated such control at the European Synchrotron ESRF (Grenoble, France) in cooperation with researchers from DESY (Hamburg) and the Helmholtz Institute/Friedrich Schiller University (Jena). In the experiment, a sample enriched with the iron isotope ^{57}Fe was irradiated with short x-ray pulses from the synchrotron and, due to the energy sharpness, release this excitation comparably slow in form of a second x-ray pulse (figure). For the control, the sample is displaced quickly by a small distance, thereby inducing tiny delays between the two x-ray pulses, with a stability of only few zeptoseconds. We then used this double-pulse to control the dynamics of a second nuclear target. This target is excited to a coherent superposition of ground and excited state by the first pulse. The second pulse continues this nuclear dynamics, controlled by the mutual pulse delay. By varying this delay, the dynamics could be switched between further excitation of the nuclei and de-excitation of the nuclei, thereby demonstrating the control of the quantum-mechanical state of the nuclei (cf. figure above). These results open the toolbox of coherent control, which has been successfully established in optical spectroscopy, to atomic nuclei – providing new possibilities and perspectives, e. g. by preparing nuclei in particular quantum states towards more precise tests of fundamental physics and more accurate clocks, see also [2].



Schematic setup for the coherent control of nuclear dynamics.

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