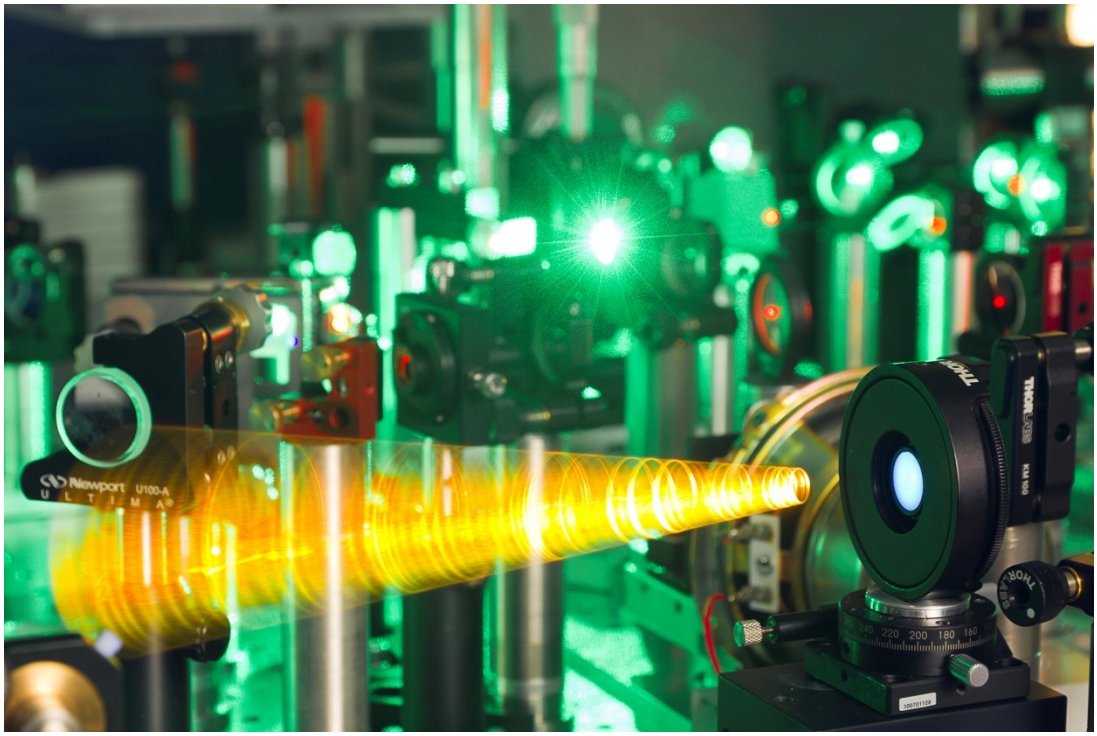
Proposal to Establish a DFG Priority Programme

Quantum Dynamics in Tailored Intense Fields (QUTIF)



Superfluorescence cone behind a single-cycle parametric amplifier

Initiators

Manfred Lein (Coordinator)
Institute for Theoretical Physics, Leibniz Universität Hannover

Gerhard Paulus (Vice-Coordinator)
Institute of Optics and Quantum Electronics, Friedrich-Schiller Universität Jena

Uwe Morgner
Institute for Quantum Optics, Leibniz Universität Hannover

Jan Michael Rost
Max Planck Institute for the Physics of Complex Systems Dresden

Regina de Vivie-Riedle
Department Chemie, Ludwig-Maximilians-Universität München

Marc Vrakking
Max Born Institute Berlin

Proposal to Establish a DFG Priority Programme

Quantum Dynamics in Tailored Intense Fields (QUTIF)

Summary

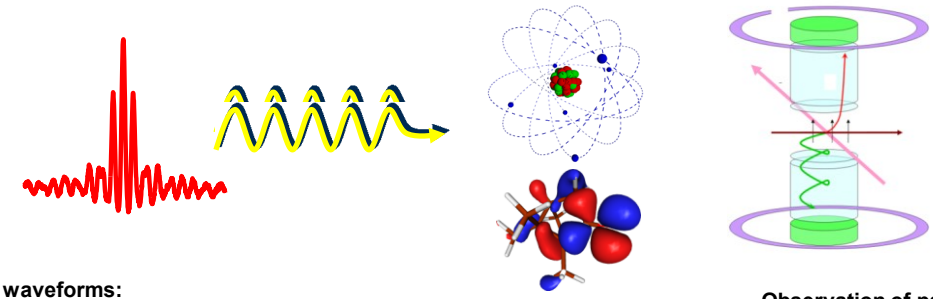
The development of light-pulse generation is reaching a stage at which fully controllable intense electric fields are available to drive microscopic systems far away from their natural states, but under well-defined conditions so that quantum mechanical dynamics prevails. Phase-stabilized ultrashort pulses with arbitrary polarization state and a broad range of wavelengths offer the possibility to subject atoms, molecules, clusters and solids to precisely known forces of sufficient strength to be comparable to, or even larger than the inner-atomic forces. The most important innovations along this way have been the generation of strong few-cycle and single-cycle pulses, the advent of efficient sources in the mid-infrared regime and advanced methods of polarization control. The essential difference to the light sources widely used ten years ago lies in the capability to shape not only the light pulse envelope but also the instantaneous electric field. Tailored intense fields provide control over electron dynamics, leading for example to the control of electron localization in molecules, controlled molecular excitation and to the manipulation of electron trajectories in strong-field ionization and high-harmonic generation, facilitating attosecond pulse production. Controlled electrons imply Ångström scale ultrafast imaging methods. This is realized in the form of laser-induced electron diffraction and high-harmonic spectroscopy with important applications in chemistry such as the tracing of non-adiabatic dynamics near conical intersections. Multicolour polarization-controlled irradiation makes it possible to orient and manipulate polar molecules and to direct currents with attosecond time precision. The area of high-harmonic generation is currently revolutionized by the use of such new light fields. Similarly, charged-particle spectroscopy, where electron and ion detection techniques such as velocity-map imaging and reaction microscopes are employed as state-of-the-art tools, is beginning to use tailored intense fields for studying quantum systems in unprecedented detail.

In this Priority Programme, the dynamics of strongly perturbed quantum systems is investigated with tailored radiation fields on the femtosecond and attosecond time scale. By combining experimental and theoretical expertise and bringing together the fields of optics, quantum dynamics and chemistry, we will achieve milestones such as the control and observation of subfemtosecond charge migration and the laser-based recognition and manipulation of chiral molecules. In order to watch microscopic phenomena with minimal disturbance by their environment, the main focus lies on gas-phase systems. On the atomic physics side of this Programme, fundamental issues are the interplay between multielectron interactions and light-induced dynamics as well as the boundary between classical and quantum physics. The attosecond temporal structure of laser-induced ionization will be analysed with a range of approaches going beyond existing attoclock and two-colour high-harmonic spectroscopy methods and extending these methods to multielectron dynamics. We study the electron spin in the strong-field regime, in particular the generation of spin-polarized electrons from laser ionization. The physics of molecular systems in intense few-cycle, multicolour and polarization controlled light pulses is a mostly unexplored territory. We investigate the launch and observation of ultrafast charge migration – an electronic effect occurring faster than nuclear motion. Electron wave-packet dynamics will be controlled with tailored light, for example by exploiting the wavelength dependence of light-molecule interactions to suppress or enhance multiorbital dynamics or to reveal the low-energy structure in photoelectron spectra from mid-infrared irradiation. Laser-induced orientation of molecules will be established and used for applications. In the realm of chemistry, tailored fields hold new opportunities for controlling chemical dynamics by nonresonant and resonant dynamic Stark shifts, tracing electron dynamics with attosecond precision and the recognition of the absolute configuration of chiral molecules. Via high-harmonic spectroscopy of molecules, we achieve ultrafast imaging of structure and dynamics on the sub-atomic length scale. The application of strong fields to solid-state systems and clusters in a quantum mechanical, i.e. non-plasma, regime holds even more open questions. Strong-field processes in unconventional media such as exploding droplets doped with nanoparticles and in laser-induced filaments imply new perspectives such as alternative attosecond pulse sources and high-harmonic generation in inhomogeneous near fields. As a result of this Priority Programme, the control of microscopic processes with light will be taken to a new level, both in terms of temporal and spatial resolution as well as regarding the variety of investigated systems.

1. Overview

Light is our most important tool to observe and manipulate the microscopic world. The main reason is that the electromagnetic field of light waves exerts forces that we can control extremely well. These forces affect in the first place the electrons inside matter and indirectly the dynamics of all other degrees of freedom. The emerging capability to generate waveforms that are tailored at the level of the instantaneous electric field on a subfemtosecond time scale and with arbitrary polarization opens a novel type of laser-matter science. Unlike the multi-cycle averaged effect of conventional laser pulses, these waveforms can exert controlled instantaneous forces in arbitrary direction. The physics and chemistry of quantum systems in the presence of tailored strong light form the central theme of this Priority Programme. Specifically, the typical light intensities of interest are in the range from 10 TW/cm^2 to 1 PW/cm^2 where the applied field strength is already comparable to the inner-atomic forces but mild enough to avoid immediate destruction of the target systems. Concerning the timescales, an 800 nm laser field has an optical period of 2.6 femtoseconds. Thus, by tailoring fields with sub-cycle and sub-femtosecond precision to electronic and nuclear dynamics, they provide access to *attosecond* chemistry and physics (1 attosecond = 10^{-18} seconds). Furthermore, attosecond pulses generated from strong-field interactions via high-harmonic generation can probe directly the temporal evolution of quantum systems with unprecedented speed. Photons, electrons and ions are emitted from irradiated systems and carry the information about microscopic dynamics. This information is often encoded in a non-obvious manner, requiring careful modelling of the underlying phenomena. By bringing together the know-how of scientists in waveform synthesis, theoretical quantum physics and chemistry as well as photon and particle detection (see figure below), we will reach breakthroughs such as the observation of ultrafast electronic charge rearrangement in atoms and molecules, or the recognition of the handedness of chiral molecules. Now is the ideal time to form a DFG-funded national network in view of the strong international competition and the world-wide current efforts to exploit the new light sources for microscopic control and imaging.

Quantum Dynamics in Tailored Intense Fields (QUTIF)



Tailored waveforms:
Thomas Baumert (Uni Kassel)
Tobias Brixner (Uni Würzburg)
Eleftherios Goulielmakis (MPQ Garching)
Ferenc Krausz (LMU München)
Franz Kärtner (Uni Hamburg)
Uwe Morgner (Uni Hannover)
Marcus Motzkus (Uni Heidelberg)
Thomas Pfeifer (MPI-K Heidelberg)
Gerhard Paulus (Uni Jena)
Eberhard Riedle (LMU München)
Christian Spielmann (Uni Jena)
Matthias Wollenhaupt (Uni Oldenburg)

Theory:
Dieter Bauer (Uni Rostock)
Dassia Egorova (Uni Kiel)
Volker Engel (Uni Würzburg)
Thomas Fennel (Uni Rostock)
Stefanie Gräfe (Uni Jena)
Misha Ivanov (MBI Berlin)
Christoph Keitel (MPI-K Heidelberg)
Manfred Lein (Uni Hannover)
Nina Rohringer (CFEL Hamburg)
Jan Michael Rost (MPI-PKS Dresden)
Ulf Saalmann (MPI-PKS Dresden)
Alejandro Saenz (HU Berlin)
Robin Santra (CFEL Hamburg)
Armin Scrinzi (LMU München)
Rüdiger Schmidt (TU Dresden)
Olga Smirnova (MBI Berlin)
Regina de Vivie-Riedle (LMU München)

Observation of particles / photons:
Bernd Abel (Uni Leipzig)
Markus Drescher (Uni Hamburg)
Reinhard Dörner (Uni Frankfurt)
Peter Hommelhoff (Uni Erlangen)
Matthias Kling (LMU München)
Reinhard Kienberger (TU München)
Milutin Kovačev (Uni Hannover)
Karl-Heinz Meiwes-Broer (Uni Rostock)
Robert Moshammer (MPI-K Heidelberg)
Adrian Pfeiffer (Uni Jena)
Horst Rottke (MBI Berlin)
Claus Ropers (Uni Göttingen)
Katrin Siefermann (IOM Leipzig)
Thorsten Uphues (Uni Hamburg)
Marc Vrakking (MBI Berlin)
Karl-Michael Weitzel (Uni Marburg)
Helmut Zacharias (Uni Münster)

2. State of the art

Below, the recent development of strong-field science is outlined. The description shows that German groups are very strong in this area and that a large part of the pioneering experimental and theoretical activities are based in this country. There is also a substantial number of group leaders who have recently moved to Germany, e.g. Olga Smirnova, Misha Ivanov, Robin Santra, Franz Kärtner, Stefanie Gräfe and Marc Vrakking. In 2013, some of the QUTIF initiators have already started bringing together the relevant researchers in international workshops held in Germany:

- The 523rd Heraeus Seminar *"High-Harmonic Spectroscopy"* organized by Manfred Lein, Milutin Kovačev, Uwe Morgner and Hans J. Wörner took place in the Physikzentrum Bad Honnef, 28 Jan – 1 Feb 2013.
- The Atomic Physics Workshop with focus days on *Quantum Dynamics in Tailored Intense Fields*, organized by Jan Michael Rost, Uwe Morgner and Manfred Lein, will be held at the Max Planck Institute for the Physics of Complex Systems in Dresden, 25 – 29 Nov 2013.

Tailored waveforms

The temporal shaping of a light wave requires that its frequency spectrum has a sufficiently broad bandwidth. The considerable bandwidth of femtosecond lasers motivated novel schemes of laser pulse shaping already 25 years ago [1]. As a result, it became possible to generate pulses of virtually arbitrary shape. The use of computer-controlled light modulators has enabled a novel type of coherent control, which resulted in a remarkable series of papers in Science and Nature. Examples have been the control of chemical reactions [2], quantum control of energy flow [3], and selective bond dissociation [4]. With polarization shaping, a new class of tailored pulses was introduced [5], where additionally the light polarization is varied on ultrafast time scales.

As a decisive step beyond earlier work, we are now able to shape not only the envelope but the entire waveform. The generic example is a few-cycle pulse, for which the carrier-envelope offset determines the temporal evolution of the electromagnetic wave. Few-cycle pulses can relatively easily be generated directly with an oscillator [6] or using hollow-fibre compression [7]. Carrier-envelope stabilization experienced a breakthrough with the invention of the self-referencing method [8,9]. While this technique has become the standard, significant progress in terms of precision and single-shot capabilities has been made with the stereo-detection of photoelectrons in opposing directions [10,11] as invented by Paulus and co-workers.

To realize the full potential of tailored waveforms for direct control of electron dynamics, one must achieve large differences in amplitude for subsequent half-cycles and control the carrier-envelope phase. To this end, there are four options of various degrees of complexity. In the simplest case, a harmonic is added to the carrier wave with adjustable amplitude and phase [12,13]. The far-reaching potential of this method has been recognized only recently [14]. Considerably more effort is needed to apply phase-controlled [15] or phase-tagged [16] few-cycle pulses, which make the interpretation of the data easier. The third option is the control of the polarization with sub-cycle time-dependence. To date, this has mainly been used to generate isolated attosecond pulses [17,18]. The most sophisticated approach is the coherent combination of several spectral bands in order to reach a total bandwidth well in excess of an optical octave. This allows for synthesizing waveforms of virtually arbitrary temporal evolution [19,20], just as it is known from radio-frequency technology or acoustics. Concerning the precision timing of laser pulses, sub-attosecond precision has recently been achieved with a pulse shaper [21].

The application of tailored waveforms to steer electronic dynamics in atoms, molecules and selected model systems together with the most advanced photon and particle detection devices is at the core of this proposal. Tailored waveforms also facilitate the generation of attosecond extreme ultraviolet pulses with an unprecedented flexibility in order to realize attosecond-pump attosecond-probe spectroscopy. At the same time, precision pulse shaping in the visible and infrared regime remains important to address those chemical processes that are difficult to initiate with extreme ultraviolet light. The control of polarization leads naturally to chemical applications regarding chiral molecules.

High-harmonic spectroscopy

High-harmonic generation (HHG) has been a steadily growing field of research since the late 1980s and has recently been revolutionized by the use of new light fields. In HHG, photons of a linearly polarized laser beam are converted into high-frequency photons while interacting with atoms or molecules. Following the

initial experimental demonstration [22] and the development of the theoretical understanding in the 1990s [23,24], considerable efforts were undertaken to apply HHG as a high-frequency radiation source. The coherence of harmonics can be exploited to form pulses with durations below a femtosecond. The first attosecond pulse trains [25] and isolated attosecond pulses [26] were demonstrated at the beginning of this millenium. Around the same time, the first experiments combining HHG with laser-induced molecular alignment were carried out [27]. Early laser-based alignment techniques used the adiabatic response of molecules. The impulsive post-pulse alignment using femtosecond pulses was developed by Vrakking and coworkers [28], offering the possibility to perform experiments on aligned molecules in the gas phase under field-free conditions. HHG from aligned molecules became a new opportunity to study molecular structure on ultrashort time scales, since the structure of molecules is imprinted on the spectrum of the emitted light [29,30]. A new research area, often termed as *high-harmonic spectroscopy* [31] started to develop. Most of its implementations exploit the three-step mechanism [32] composed of (i) laser-induced ionization, (ii) electron acceleration by the field and (iii) recombination with the parent ion. In the third step, an electron wave packet impinges on the parent ion and probes its state. The de Broglie wavelength of the returning electron is comparable to the size of molecules, implying spatial sensitivity on the atomic scale. Examples of high-harmonic spectroscopy are the molecular orbital tomography by Corkum and coworkers [30] and the probing of attosecond dynamics by chirp encoded recollision (PACER), i.e. the attosecond tracing of nuclear motion proposed by Lein [33] and experimentally realized by Marangos and coworkers [34]. PACER exploits the isotope effect as it compares the HHG spectra from hydrogen (H_2) and from heavy hydrogen (D_2). The idea has been extended to polyatomic molecules such as methane, water and ammonia [34-37]. PACER operates on the attosecond scale since the photon energy of the emitted harmonics is mapped on the electron excursion time, typically ranging from about zero to two femtoseconds. This phenomenon – known as attochirp – is predicted already within Corkum model [32]. It was demonstrated experimentally by Salières and coworkers [38]. A closely related imaging method exploiting coherent recollision electrons is the laser-induced electron diffraction [39].

McFarland et al. and Smirnova et al. showed that not only the highest occupied molecular orbital (HOMO) but also lower-lying orbitals contribute to the HHG spectrum [40,41]. This represented a paradigm shift away from the belief that optical laser fields couple essentially only to the most loosely bound electron. This implies an exciting consequence: ionization leaves a hole wave packet which can be probed by observing HHG. Hässler et al. combined this idea with molecular orbital tomography and measured images of the orbitals constituting attosecond wave packets [42]. Besides being an attosecond technique, the HHG-based imaging has an advantage over conventional diffraction-type methods such as X-ray diffraction: those require phase-retrieval algorithms to obtain spatial images, whereas Hässler et al. directly measured the harmonic phases and used them in the tomographic retrieval. Recently, a way to suppress the contributions of the more tightly bound electrons has been found: by moving to driving wavelengths in the mid-infrared regime, the measured spectra are compatible with a HOMO-only interpretation, which leads to easy valence-orbital tomographic imaging [43].

The importance for chemistry is underlined by Wörner et al.'s experiment, probing the dynamics of a molecular wave packet passing through a conical intersection in NO_2 molecules [44]. The non-adiabatically coupled electronic and nuclear motion at conical intersections [45] is crucial in important biomolecular reaction pathways. In a quite opposite direction, namely fundamental atomic physics, the exit time of the electron in tunnelling ionization of helium has been measured with attosecond-scale precision by Dudovich and coworkers using two-colour HHG with orthogonally polarized 800 nm and 400 nm fields [46]. The weak second-harmonic field perturbs the electron motion and gives information about the ionization dynamics. With theoretical support by Smirnova and Ivanov, it was shown that tunnelling follows precisely the dynamics predicted by the quantum-orbit model based on complex-time trajectories [47]. By numerical solution of the time-dependent Schrödinger equation, the Lein group has shown that also the *Keldysh tunnelling time,* i.e. the imaginary part of the complex ionization time, can be retrieved from two-colour HHG [48].

In recent work by Kovačev, Morgner and coworkers [49], high-order harmonics were observed from laser-induced filaments. In this case, HHG has given insight into the spatiotemporal evolution of the filament, which appears to be an alternative source of attosecond pulses. Furthermore, the Hannover group has recently revisited HHG from water microdroplets in a pump-probe setup [50]: a first pulse heats and expands the water droplets; thereafter harmonics are generated by a second pulse at a variable time delay. It has been shown that already at liquid-water densities, strong HHG is observed and can give information about the droplet expansion. In the future, droplets will provide a way to let strong fields interact with dopants such as nanoparticles in a controlled manner.

Strong-field particle spectroscopy

Numerous groups in the atomic and molecular physics community work with the same quantum systems and similar light sources as the HHG community, but there is a long tradition of detecting charged particles, i.e. ions and electrons, as a means to investigate microscopic dynamics. The detection of charged particles by cold target recoil ion momentum spectroscopy (COLTRIMS) was originally developed by the Frankfurt atomic physics group to measure recoil momenta in ion-atom collision physics [51]. In the year 2000, COLTRIMS began to revolutionize the study of laser-induced phenomena by providing ion momentum spectra for single and double ionization of He atoms by strong laser pulses [52,53]. The ability to detect ions and electrons in coincidence gave a new level of insight into the non-sequential double-ionization mechanism and gave the first direct experimental proof that rescattering is responsible for correlated emission of two electrons from atoms. By now, COLTRIMS has been applied to a multitude of questions in strong-field physics with atoms and molecules. An alternative technique for photoelectron momentum distributions, which does not provide coincident detection, but much faster acquisition of the data is the velocity-map imaging method (VMI) [54]. VMI measures the projection of the three-dimensional ion or electron distribution onto a detection plane. From the two-dimensional image, the full distribution can be obtained by Abel inversion provided that the distribution is cylindrically symmetric. In the case without symmetry, the three-dimensional object can be retrieved by a tomographic procedure, as pioneered in the Baumert group [55]. VMI is becoming a standard tool in strong-field science. One striking example is the measurement of molecular-frame three-dimensional momentum distributions from laser-aligned molecules, performed by Stapelfeldt and coworkers [56]. Another recent example is the control of the left-right directionality of ion emission in the fragmentation of small molecules by phase-stable few-cycle mid-infrared pulses, see the work by Znakovskaya et al. on D_2^+ molecular ions [57]. COLTRIMS and VMI are now established techniques, but substantial work is needed to combine them with the new available radiation sources. QUTIF will bring the relevant researchers together to reach the best possible scientific gain.

Electron dynamics

Both intense laser pulses and attosecond pulses can easily ionize matter, either due to the high field or due to the high frequency. This is one reason why the recent years have shown vigorous activity to resolve the ionization dynamics of atoms and molecules with attosecond temporal precision. As described above, in the context of HHG, ionization times have been determined by two-colour methods [46]. Other approaches employ the detection of ions and electrons. One possibility is the streaking method where an attosecond pulse ionizes the quantum system by one-photon absorption and the subsequent response of the electron to a strong infrared pulse gives information about the exit time of the electron. In this way, delays around 20 attoseconds have been measured for photoionization from different orbitals in Ne atoms [58]. Similar measurements were reported for Ar atoms [59]. These results have stimulated an outburst of theory papers aiming at explaining the unexpectedly large time delay [60-62]. Another, rather different method is the *attoclock* established by Keller, Dörner and coworkers for investigating tunnel ionization [63]. It is based on angular streaking with elliptically polarized few-cycle pulses. In this case, the laser field fulfils two purposes: it first ionizes an atom and then rotates the emission direction of the outgoing electron and the recoil ion. From the emission angle at the peak of the momentum distribution, one retrieves the time of ionization. The initial set of measurements was compatible with the concept of instantaneous tunnelling, i.e. there is no real delay between the maximum of the electric field and the birth of the electron in the continuum. Recent attoclock measurements suggest a small but non-zero real tunnelling delay time [64]. Concerning molecules, the ionization dynamics of H_2^+ and N_2 molecules has been investigated by the Dörner group with the help of circularly and elliptically polarized pulses [65,66]. Unexpected features have been revealed such as the importance of the initial momentum of the outgoing electron. This finding is important since in tunnelling the initial velocity of the electron is usually considered to be small. The knowledge about these initial conditions is essential for using the attoclock. The ionization of polar molecules has been studied with coincidence measurement of electrons and ions, confirming the sensitivity of ionization to the orbital shape and the importance of multiorbital contributions [67].

For ultimate applications in ultrafast chemistry and electronics, however, the dynamics of bound electrons is the essential handle to control the target systems. Via the response of bound electrons to strong fields (Stark effect), nuclear motion can be controlled [68,69]. It has been shown that stronger fields allow the tracing of dynamics on shorter time scales [70]. In July 2013, Vrakking and co-workers have traced molecular laser-induced polarization with extreme time precision by attosecond probing. Earlier this year, Wollenhaupt, de Vivie-Riedle and co-workers have demonstrated the use of precision pulse shaping for the controlled population of molecular target states. Krausz and co-workers have shown that the a.c. conductivity of solids

can be controlled at sub-laser-cycle time scales [71]. These examples underline the tremendous progress made in the last few years in the control and measurement of electron dynamics relevant for future applications.

Chiral molecules

Chirality plays an extremely important role in chemistry and pharmaceutics because different enantiomers of a drug can have entirely different effects on the human body. Polarization-controlled light is an essential tool for the study of chiral molecules. It has long been known that the handedness of chiral molecules can be recognized by their different absorption of circularly polarized light (circular dichroism) and by the opposite rotation of the light polarization (optical activity). In photoionization, a forward-backward asymmetry is present in the photoelectron momentum distributions in one-photon ionization with circular polarization [72,73]. In contrast to photoabsorption, this *photoelectron circular dichroism* can be understood within the electric dipole approximation [74]. It is therefore a large effect: the relative difference between the signals in forward and backward directions is of the order of 10 percent. However, one-photon ionization typically requires high frequency and thus synchrotron radiation. Baumert, Wollenhaupt and coworkers have recently demonstrated for camphor and fenchone molecules that an asymmetry appears also in electron distributions from *multiphoton ionization* with table-top laser pulses [75]. So far, the theoretical understanding of this effect is limited since the published theory covers only the one-photon case. Ongoing work in the Lein group shows that a strong-field ionization theory for chiral systems, based on quantum chemical bound-state calculations and approximate non-plane-wave continuum states, yields realistic results for the photoelectron circular dichroism. Already a few years earlier it was shown that circular dichroism is present in the ion yields from femtosecond laser ionization [76]. In September 2013, the experimental determination of the absolute molecular configuration of chiral molecules by coincident detection of five charged fragments after irradiation with high-repetition rate pulses has been reported [77]. Chiral molecules receive growing interest also in high-harmonic spectroscopy [78] and there are various proposals how to exert coherent chemical control on them [79,80]. In the future, we expect to use time dependent variation of the light polarization in order to control chiral species.

Mid-infrared fields

Another important new trend in laser-matter interactions concerns mid-infrared fields. The last few years have shown impressive progress in the generation of strong pulses in the wavelength range from one to two μm by optical parametric amplification. Important experiments have already been carried out with such pulses [81]: the energy range of the HHG spectrum has been extended substantially while the driving intensities could be kept moderate. This has led to the confirmation of phenomena which could not be clearly seen at 800 nm wavelength. For example, the presence of the Cooper minimum in HHG spectra from krypton atoms [82] and the giant resonance in spectra from xenon atoms [83] were demonstrated. Also, organic molecules have been studied successfully [84].

Besides the anticipated benefits (better applicability of trajectory-based models and usability for holography [85]), mid-infrared fields have revealed unexpected new effects. Most notably, a low-energy structure has been measured in the photoelectron energy spectrum [86]. It has been reproduced theoretically by Bauer, Vrakking and coworkers [87] and by Gräfe and coworkers [88]. The Rost group has explained the position of the enormous peak in terms of soft recollisions [89], as opposed to the more frequently discussed hard recollisions responsible for the emission of high-energy electrons. Although several groups have investigated this new mechanism, no calculations on molecules have been reported while there is clear experimental evidence of large differences between atoms and molecules [86]. An important aspect of mid-infrared pulses is the suppression of multielectron effects in HHG in sharp contrast to the more complicated situation at 800 nm [43]. Multielectron contributions pose a big challenge in molecular imaging and at the same time they open up the possibility to study electrons in inner subshells. Long wavelengths are advantageous in the PACER method for probing of molecular dynamics on the attosecond time scale [34], as they extend the accessible time window for probing. In previous work, only the very first moments of the initiated wave-packet motion could be probed. With mid-infrared pulses, complete cycles of molecular vibrations become accessible. This method provides the possibility to study quantum nuclear dynamics with sub-femtosecond resolution in the presence of a strong external field.

International collaborations and visibility of the QUTIF consortium

A very successful international community in strong-field quantum dynamics has emerged in recent years, particularly in Europe. Attosecond physics and laser-based imaging are increasingly covered by the top scientific journals such as Nature and Science. Most of the principal investigators in QUTIF have strong links to collaborators in other countries, many of them through coordinated EU funded networks. Selected cooperations of the QUTIF initiators that have led to joint published or submitted papers are listed below.

Manfred Lein:	– Jon Marangos (Imperial College London) – Camilo Ruiz (CLPU Salamanca) – Ignacio Sola (Universidad Complutense Madrid)
Uwe Morgner:	– Mette Gaarde (Lousiana State University) – Luis Plaja (Universidad de Salamanca)
Gerhard Paulus:	– Zenghu Chang (University of Central Florida) – Robert Jones (University of Virginia) – Pierre Agostini, Louis DiMauro (Ohio State University) – Andrius Baltuška (TU Wien)
Jan Michael Rost:	– Roland Guichard (UMPC Université Paris) – Nora Berrah (Western Michigan University)
Regina de Vivie-Riedle:	– Ali Alnaser (King Fahd University of Petroleum and Minerals, Saudi Arabia) – Eric Wells (Augustana College, South Dakota)
Marc Vrakking:	– Albert Stolow (NRC Ottawa) – Fernando Martin (Universidad Complutense Madrid) – Giuseppe Sansone (Politecnico di Milano) – Anne L'Huillier (Lund University)

3. Scientific objectives

The availability of new light sources producing *fully controllable intense* fields leads to new prospects in physics and chemistry because virtually arbitrary forces can be applied to microscopic systems. In this Priority Programme we drive *quantum systems* with strong fields, i.e. these systems are far away from their equilibrium state. In contrast to conventional spectroscopy, we are primarily interested in the properties of such strongly perturbed systems. In the language of quantum mechanics, ultrafast interactions imply little decoherence: during the interaction, the driven quantum systems suffer only little noise from their environment so that the full quantum mechanical behaviour is revealed. In the following we outline the three major objectives of QUTIF. For each objective, we state tangible milestones and ultimate goals that we regard as particularly important.

Control of electron dynamics is the grand unifying objective underlying most of the current research on intense-laser-matter interactions. Electrons are important because they cause the decisive forces in chemical reactions of molecules and they are responsible for conductivity. All matter that is relevant for the processes in our daily life consists of electrons and nuclei. Among these particles, electrons are by far the lightest ones and their motion takes place on the shortest time scales, from femtoseconds down to attoseconds. As they couple directly to the electric field of the applied laser pulses, light is the appropriate handle to steer electrons. Consequently, if the electrons are subject to suitably tailored external light, virtually all other processes of interest can be controlled as well since they are usually slower. Sudden changes in the charge distribution of extended systems such as polyatomic molecules can propagate within attoseconds through the system. Within QUTIF we launch and probe ultrafast charge migration with controlled infrared fields and attosecond pulses. Reaching the “holy grail” of attosecond-pulse-based science – the attosecond-pump attosecond-probe spectroscopy of complex correlated systems - will represent a major breakthrough within this Priority Programme. In solid materials and surfaces, we control currents and conductivity on the attosecond time scale and thus enter a new area of strong-field solid-state electronics [71]. The small mass of electrons also makes them ideal objects for studying quantum mechanical behaviour such as tunnelling and correlated dynamics in many-electron systems. Although the dynamics of bound electrons is most decisive for chemistry, a huge part of previous work has concentrated on ionization dynamics. It is a useful domain to study electron wave packets and it has brought up open questions concerning tunnelling ionization and multiple ionization. QUTIF will solve these fundamental questions and proceed further to

bound-electron dynamics relevant for chemistry and electronics.

Intermediate milestones:

- Reaching an understanding of tunnelling ionization: tunnelling time and exit velocity
- Observation of attosecond wave packets in complex molecules
- Sub-cycle control of ionization and dissociation

Ultimate goals:

- Control of practically relevant chemical dynamics with subfemtosecond precision
- Attosecond-pump attosecond-probe spectroscopy of complex electronic dynamics

Establishing strong-field imaging. A few pioneering studies have demonstrated the feasibility of Ångström-scale molecular imaging via high-harmonic generation [30,42] or laser-induced electron diffraction [39], which are both based on laser driven recollision of electrons with their parent ions. Since the de Broglie wavelength of recollision electrons is comparable to the size of atoms, molecular structure can be resolved. Moreover, since these methods operate on femto- and attosecond scales, one may image distortions in the presence of additional strong fields. Previously used laser pulses were not ideal due to the limitations in pulse generation. Novel mid-infrared pulses simplify the interpretation of harmonic spectra and photoelectron distributions substantially and facilitate clean molecular imaging. The manipulation by additional tailored fields, e.g. to drive Stark shifts in molecules or to orient/align molecules, provides a handle to study molecules relevant in physics, chemistry and biology. Entirely new applications such as recording the change of an electronic orbital during nuclear motion in a chemical reaction will be realized. When applied to conical intersections, quick changes in the orbital character will be measured. Strong-field imaging will be extended to new media, namely surfaces and clusters. Overall, tailored light pulses will make the vision of *Ångström scale imaging with femtosecond to attosecond time resolution* come true.

Intermediate milestones:

- Characterization of the strong-field response in the mid-infrared regime
- Efficient molecular orientation with two-colour fields or few cycle pulses

Ultimate goals:

- Imaging of orbitals while undergoing chemical reactions
- Ångström and attosecond-scale imaging of nuclear and electronic motion

***Manipulation of chirality in physics and chemistry*.** Chirality is one of the most intriguing topics in science and at the same time it is of high practical relevance in biochemistry. Our table-top laser-based research will be extremely useful for practical chemical applications to chiral molecules. This includes the recognition of the left-handed or right-handed variant of molecules as well as the selective manipulation of a desired enantiomer in a racemic mixture, potentially even the selective generation of enantiomers in laser-controlled chemical reactions. Within QUTIF, a huge progress in theoretical molecular physics will necessarily take place in order to achieve a strong-field treatment of chiral molecules. Experimentally, we proceed from few-photon to multiphoton processes and we use tailored light polarization. Addressing the electron spin is a further emerging aspect of chirality in strong-field interactions. We generate spin-polarized electron beams by shining circularly polarized pulses and polarization-controlled pulses on atoms and more complex systems. The prerequisite of spin polarization is the presence of spin-orbit coupling and the sensitivity of the strong-field response to the left- or right-handed sense of the orbital angular momentum. We will confirm and analyse these predictions.

Intermediate milestones:

- Quantitative theory of strong-field dynamics of chiral molecules
- Characterizing the effect of bound-state orbital angular momentum in the strong-field response

Ultimate goals:

- Photoionization-based measurement of the absolute configuration of chiral molecules
- Laser-based selection of enantiomers from racemic mixtures
- Spin-polarized electron emission by strong fields

To realize the above listed objectives, a full understanding of quantum systems in fields of arbitrary shape is needed. The response of atoms, molecules, clusters and surfaces to few-cycle pulses with fixed carrier-envelope phase, polarization-controlled fields, multicolour fields and mid-infrared fields is unexplored to a large extent and will most certainly provide many surprises. On the experimental side, we apply fully

controlled light pulses to a broad variety of target systems. This requires the collaboration of research groups with rather different backgrounds, such as pulse design and chemistry. QUTIF will stimulate the interaction of researchers with expertise in advancing light fields and those with long-standing knowledge about the illuminated targets. On the theory side, versatile tools need to be developed to treat the nonlinear response of few- and many-particle systems. Traditionally, strong-field theory has often considered only small systems and used simplified models. By combining state-of-the-art methods from theoretical chemistry and strong-field theory, we achieve quantitative descriptions of laser-driven polyatomic molecules with chemical accuracy. We place particular emphasis on the dynamics on a time scale below the optical cycle of the driving field as we are interested in the shortes time scales accessible with currently available light sources.

4. Work programme

The work plan ranges from fundamental atomic science to applied research on molecules and solids. Despite the broad range of targets, all projects share the idea of using tailored intense driving fields to address microscopic phenomena.

A. Fundamental problems in atomic physics

A1. ***Multielectron correlation*****.** The electron-electron interaction crucially influences both the electronic structure of atoms and their response to external fields. Double and multiple ionization is known to be dramatically enhanced by electron correlation via a rescattering mechanism where laser-driven electrons excite or knock out further electrons from atoms [52,53,90]. Correlation is responsible also for the decay of inner-shell vacancies and doubly excited states. This implies the appearance of Fano line shapes in atomic absorption spectra, which can be modified with laser fields [91]. We address the control of correlation in strong fields by applying pulses with well controlled polarization, in particular elliptical polarization, and by coincidence measurement of fragment momenta as well as attosecond absorption spectroscopy. Important unsolved questions will be answered, such as the breakdown of the independent electron approximation in sequential double ionization [92]. Theoretically, electron correlation is notoriously difficult to treat. Computational approaches will be developed for the combined treatment of correlation and strong perturbations. This includes the quantum mechanical simulation of two-electron dynamics in circularly polarized fields, which has not been reached to date.

A2. ***Dynamics at the boundary of classical and quantum physics.*** Strong-field phenomena such as high-harmonic generation are often interpreted in terms of classical trajectories [32,93,94]. Tunnelling ionization, although being a deeply quantum mechanical process, is thought to provide the starting conditions (time, position and velocity) for subsequent classical motion. This has caused heated debates concerning for example the existence of a non-zero real delay in tunnelling [63,64], the meaning of complex-valued trajectories [48] and the influence of non-adiabaticity due to the non-zero laser frequency [95,96]. To answer these questions, QUTIF goes beyond existing ways to investigate tunnelling [46,63]: we will exploit photoelectron spectra from few-cycle and two-colour irradiation with variable phase delay as well as polarization-resolved high-harmonic spectroscopy to deduce ionization times. Methods to determine the phase of tunnelling wave packets by interference with one-photon ionization wave packets [97] will be devised to gain additional insight into tunnelling. With respect to non-adiabaticity, the wavelength dependence of ionization and high-harmonic generation will be studied. Wavelengths from the deep ultraviolet to the mid-infrared frequency regimes are employed. As a result, QUTIF will establish the correct interpretation of classical concepts for the description of laser-induced phenomena.

A3. ***Electron spin in strong fields.*** The spin degree of freedom has mostly been ignored in strong-field science because laser fields do not couple directly to the spin as long as the electric dipole approximation is assumed. However, recent developments show that spin-orbit interactions are extremely important for strong-field dynamics: ionization can create bound-electron wave packets in the ion, oscillating at the spin-orbit frequency [98] or, according to analytical theory, ionization can even lead to strongly spin-polarized electron emission [99]. QUTIF will set up experiments to confirm spin-dependent ionization. The effect is a consequence of the dependence of ionization yields on the magnetic quantum number of the initial bound state, which we will characterize experimentally and theoretically. By numerical simulations we calculate the degree of spin polarization that can be obtained from various target systems (atoms and molecules.)

A4. ***Quantum dynamics in sculpted waveforms.*** By reaching the single-cycle pulse duration limit from phase-stabilized laser sources and optical parametric chirped-pulse amplification systems and by coherently adding different harmonics, pulse shaping has been extended to the instantaneous electric field itself [19,20].

This provides highly non-sinusoidal fields, giving rise to very new experimental prospects for control of light-matter interactions on the sub-femtosecond time scale [100]. Already proven concepts of waveform synthesis and new approaches will be made accessible for quantum dynamics projects. These include modifying the cutoff [32] and shaping the attochirp [38] in high-harmonic generation as well as launching electron wave packets with extremely confined starting time.

B. Molecular physics in strong controlled fields

B1. ***Attosecond charge migration.*** Not only the electronic excitation of atoms or molecules with broadband extreme ultraviolet pulses but also the sudden removal of an electron from a molecule results in electron dynamics on the attosecond time scale [101]. Electron dynamics implies charge migration over potentially large distances within a polyatomic molecule. This can lead, for example, to specific fragmentation at atomic sites remote from the site of the initial ionization much earlier than the remote site could be reached by a vibrational-coupling migration mechanism. QUTIF will work towards the detection of ultrafast charge migration using attosecond pulses. With additional laser fields, the electron dynamics will be modified, leading eventually to the ultrafast control of microscopic currents within molecules.

B2. ***Laser-induced alignment and orientation*****.** While impulsive laser-induced alignment of molecules has become a standard technique to investigate the molecular response in dependence of the orientation [28] [102], there have been few reports on laser-induced field-free orientation in the sense of head-versus-tail order in gases of non-inversion-symmetric molecules [103,104]. Impulsive methods have the advantage that the alignment field is off at the time of rotational revival, facilitating alignment-field-free experiments with aligned/oriented molecules. With phase-controlled multi-colour pulses and few-cycle pulses, we will develop molecular orientation as a standard tool in strong-field science. High-harmonic spectroscopy with oriented molecules will provide valuable information about molecular properties via measurement of even and odd harmonics of the driving laser field [105]. The alternative access to fixed-in-space molecules is the coincidence measurement of fragments and electrons. In this case, the direction of molecular fragmentation can be correlated with the momentum distribution of the electron. Aligned and oriented molecules offer opportunities for exciting new research: the delay in photoionization [58] will be studied as a function of molecular orientation. Different alignment angles of a molecule give the same photoelectron energy so that Coulomb-laser coupling effects [61,106] cancel out when measuring the photoemission time difference of different orientations. This simplifies the analysis as compared to the atomic experiment [58].

B3. ***Molecular tunnel ionization.*** The widely held assumption that only the highest-occupied molecular orbital (HOMO) is relevant in strong-field ionization and high-harmonic generation had to be given up in recent years [107]. It is now accepted that lower-lying orbitals contribute substantially to the strong-field response, especially when the HOMO contribution is suppressed by the structural symmetry of the orbital [41]. Nonetheless, for long wavelengths the lower-orbital effects are suppressed in high-harmonic generation [43]. QUTIF will investigate the presence of multiorbital dynamics in high-harmonic generation and photoelectron spectra over a wide range of wavelengths. Substantial work has been devoted in the last few years to the huge low-energy structure (LES) in the photoelectron spectra from *atoms* in mid-infrared fields, first described in 2009 [86]. Although the pioneering experiment included results on the LES of nitrogen molecules, there is no theoretical work nor understanding of the differences between the atomic and molecular LES. We will extend the LES theory to molecules and analyse the effect of molecular structure. Ionization at large internuclear distance is another extreme situation, encountered in noble-gas dimers [108]. It leads to double-slit interference which can be viewed as a probe of electron localization. When interference is observable, it facilitates diffraction-like imaging of the molecular geometry. In combination with coincident detection of electrons and nuclei, molecular ionization can be studied in even more detail. Photoelectron distributions in the molecular frame will give the crucial information to decide whether molecular tunnelling follows the atomic picture or whether there are additional effects on the exit time and exit velocity of the tunnelling electron.

C. Applications in chemistry

C1. ***Chiral recognition.*** As shown in recent experiments [75], the photoelectron momentum distribution from multiphoton ionization of chiral molecules irradiated by circularly polarized light is sensitive to the choice of enantiomer. The phenomenon is analogous to earlier work using one-photon ionization by synchrotron radiation [72]. Nevertheless, the multiphoton approach is more attractive for applications because it is a table-top technique. We will explore chirality in laser-driven electron dynamics and develop the first theory for strong-field ionization of chiral molecules. The sensitivity of ion yields, photoelectron distributions and high-harmonic generation spectra to chirality will be analysed. This work leads to immediate applications for the

recognition and selective manipulation of enantiomers. We will establish the recognition of enantiomers by the asymmetry in the electron distributions. Furthermore, gas-phase experiments even with quickly racemizing enantiomers become possible with coincidence methods: by detecting ions from a fragmenting enantiomer the handedness of the molecule is determined on the fly and thus the photoelectron distribution is recorded in correlation with the measured molecular geometry. Polarization-shaped pulses will be used to explore new avenues of controlling chiral molecules.

C2. ***Attosecond control and imaging of reactions.*** Although chemical reactions are essentially the rearrangement of atoms on the femtosecond time scale, in many cases the course of a reaction can be decided on a much shorter time scale by irradiation of attosecond pulses or by tailored few-cycle waveforms [45]. This has been demonstrated, for example, by the controlled left-right asymmetry in dissociative ionization of deuterium molecules [109] and by the selection of fragmentation pathways in polyatomic molecules such as ethylene using few-cycle-pulse-induced recollision [110]. With precision pulse shaping, charge oscillations have been controlled on the attosecond scale to address specific electronic excitations [111]. In QUTIF, we explore in detail the control of chemical reactions with tailored few-cycle pulses and eventually attosecond pulses. By controlling the carrier-envelope phase of few-cycle pulses, we manipulate the directionality of the electron dynamics and its recollision energy. A moderate approach to chemical control is provided by dynamic Stark control [68,69], which we will implement with tailored fields to achieve control during the entire evolution of a reaction. In a pioneering experiment in chemical high-harmonic spectroscopy, it has been shown that the dynamics of molecules near conical intersections can be traced in the harmonic emission [44]. Conical intersections are ubiquitous in polyatomic molecules and play an important role in biochemistry. To this end, we will perform pump-probe studies to trace the dynamics of small and large molecules. Typically, an ultraviolet photon is required to initiate quantum dynamics on an excited electronic state of a molecule. Therefore, tailored few-femtosecond UV pulses will be a key element in these studies. The coupled dynamics of electrons and nuclei beyond the Born-Oppenheimer approximation, as it occurs near conical intersections, will require dedicated theoretical work. In QUTIF, we will also pursue recollision-based methods for imaging of molecular structure and dynamics. This includes high-harmonic spectroscopy and laser-induced diffraction. An intriguing proposal is the molecular orbital tomography [30], where orbitals are reconstructed from measured amplitudes and phases of high-harmonic radiation from aligned molecules. In QUTIF, we will analyse the possibility to record the time dependence of an electronic orbital during a chemical reaction. In a femtosecond pump-probe scheme on aligned molecules with the high-harmonic spectrum as the probe signal, tomography reveals changes of the orbital as the nuclei undergo rearrangement. Particularly interesting will be the adiabatic or diabatic dynamics at avoided crossings or at conical intersections, where a drastic change of the orbital shape should be observed.

D. Extending strong-field quantum dynamics to new media

D1. ***Strong-field response of doped droplets.*** We investigate the response of exploding water droplets to short pulses [50,112] and particularly the effect of dopants inside the droplets, cf. [113,114]. These dopants may be molecules as well as solid state materials such as nanoparticles. We investigate high-harmonic generation and its dopant-size dependence. The droplet setup has the unique feature of providing a mass-limited target for strong-field interactions. The accessible range of water densities is extremely large, ranging from liquid water to thin vapours. The dynamics of water molecules in the near field of the laser-driven nanoparticles is an approach to perform high-harmonic generation in spatially inhomogeneous fields.

D2. ***Dynamics in filaments.*** Due to the formation of ultrashort spikes in laser-induced filamentation in gases, filaments are a promising alternative approach to attosecond pulse generation [49]. High-order harmonic generation from filaments and the underlying microscopic dynamics will be investigated. The methodology includes measurement of spectral data, spatially resolved far-field detection and the measurement of coherence times and dipole phases. Theoretically proposed excited Kramers-Henneberger states [115] will be studied experimentally. Filaments are of high practical interest as they are also relevant in laser manufacturing of solids. By analysing filamentation dynamics, QUTIF will answer open questions related to the pulse-shape dependence of nanostructure formation [116].

D3. ***Multiphoton physics at solid surfaces.*** Attosecond physics and strong-field quantum dynamics have focused almost exclusively on gas-phase systems (atoms and molecules) for a long time, but recent experiments on attosecond-resolved electron emission from surfaces [117] and rescattering at nanospheres [118] and nanotips [119] show that solids are becoming a new target of investigation. New experimental setups as well as thorough theoretical modelling will be devoted to study these new phenomena. We will investigate the applicability of typical strong-field concepts such as recollision, tunnelling ionization and nonrelativistic high-harmonic generation to the case of surfaces. The interaction of waveform-controlled few-cycle pulses with insulators can modulate the a.c. conductivity on a sub-laser-cycle time scale [71]. Along

these lines, we will study applications of strong fields to ultrafast electronic engineering.

D4. *Clusters.* Existing reports on intense-laser-cluster interactions have mainly discussed plasma formation and mechanisms of energy absorption [120]. As in solids, little is known [121] about the mechanism of high-harmonic generation or other quantum mechanical strong-field processes in clusters. Recent experiments suggest that coherent processes involving delocalized electrons exist in clusters [122]. By subjecting clusters to tailored fields such as elliptically polarized pulses and two-colour fields, new information about the harmonic generation mechanism and about the role of electron delocalization will be gained in QUTIF.

Methodology

Laser-based radiation sources in a wide range of wavelengths from the mid-infrared to the extreme ultraviolet will be applied. The most modern techniques will be used to control the light fields on the femtosecond and attosecond time scales: waveform synthesis, phase tagging, polarization control and attosecond pulse generation. From the perspective of detection, the tools used in QUTIF fall into the categories:

(i) *Strong-field particle spectroscopy: electrons and ions, typically from atomic or molecular multiphoton break-up processes, are detected.*

Presently, the two major experimental tools are the reaction microscopes based on COLTRIMS [51] for coincident detection of charged particles and VMI (54) for fast-acquisition measurement of momentum distributions. Recently, the first phase-dependent experiments using cold but fast molecular ion beams were reported [123]. In concept, they are similar to COLTRIMS, but they allow for using the most fundamental molecules such as H_2^+ and H_3^+. In addition, also neutral fragments can be detected.

(ii) *High-harmonic spectroscopy: the investigation of microscopic structure and dynamics by observation of high-order harmonic radiation.*

Using HHG not only as a light source, but as an observable to investigate fundamental processes in atoms and molecules [31,124] is a recent trend. Although Germany has very strong research groups in HHG and attosecond physics, only recently German groups have entered the emerging field of high-harmonic spectroscopy, which is suited to investigate both electronic motion and molecular dynamics on ultrashort time scales, from femtoseconds to attoseconds.

Our theoretical tools range from the most recent analytical methods [125] derived from the theories of Keldysh, Faisal, Reiss [126] and Perelomov, Popov, Terent'ev [127], to fully numerical methods involving the direct solution of the one-body and few-body time-dependent Schrödinger equation. Applying the numerical approach to problems with several dimensions will rely on efficient parallelization. To obtain quantitative results for molecules, data obtained from quantum chemical calculations is used as input information to strong-field theories.

5. Networking and training

Various types of network activities are planned to ensure scientific exchange, international visibility and training of PhD students and postdocs, see the detailed description below. We will ensure that all QUTIF events are accessible to disadvantaged network members by offering special support for family travel and for barrier-free access (with advice taken from Ingo Barth, deaf postdoc in Hannover). The information flow between the participating groups and the visibility to the public will be guaranteed by setting up a QUTIF website based in Hannover. It will display the QUTIF events, job offers and publications. A secured part of the webpage is used to exchange internal information and data between the partners.

Annual Meetings

QUTIF will hold

- a **kick-off meeting** at the beginning of the Priority Programme and
- **annual meetings I-VI** near the end of each completed year

where presentations from all participating groups are expected. Since the requested funding periods are two times three years, the annual meetings III and VI will summarize the status and achievements in comparison to the initial goals. Meetings I, II and IV will be **networking meetings**: in addition to status reports, these meetings will offer slots for extended presentations of existing or planned collaborations within QUTIF.

Promising ideas for new collaborations can receive special support from the steering committee on request. The desired benefit is an increased quantity of joint research in the second funding period. Note that several essential topics of joint work are already listed under Synergy in Section 6. Meeting V will be a special **meeting on future developments**, discussing the anticipated outcome of QUTIF, future trends and the potential continuation of collaborative research.

Coordinated training of young scientists

The training of the young researchers within QUTIF covers the three main aspects: scientific knowledge, personal skills and autonomy.

Four training schools will provide specific scientific training that is vital to this research field. All PhD students hired within QUTIF are expected to attend the schools. Tutorials will focus on both experimental and theoretical/numerical aspects of ultrafast laser physics, high-harmonic spectroscopy and strong-field particle detection. Lectures will be presented by members of the programme consortium and invited speakers. In particular, the topics include tailored waveforms, quantum dynamics of atoms, molecules and other media in strong fields, high-harmonic generation and attosecond physics. In addition, the schools contain elements of transferable skills: issues of confidentiality, copyright and ethical issues, understanding of funding and evaluation in scientific research, academic or commercial exploitation of research results, career planning and developing ways to improve employability (effective drafting of a curriculum vitae, applications, preparation of scientific presentations and interviews). To this end, QUTIF will invite speakers from funding agencies, industry and scientific journals. The schools will normally take place at one of the QUTIF institutions. The hosts will additionally provide training at the local laboratory. The training schools are held in the first, second, fourth and fifth year of the Programme. To keep the organization manageable, a part of the schools will be held in conjunction with the annual meetings.

The training of independence and autonomy of young researchers concerning in particular personal management, organization skills and mobility is achieved by the following instruments:

- **Two young-researcher meetings** will be organized solely by the young scientists at one of the partner sites. The principal investigators are excluded from this meeting, but the young researchers are encouraged to invite external speakers. The young-researcher meetings are planned for the third year, i.e. near the middle of the Priority Programme duration, and at the beginning of the sixth year.
- **Secondments** of PhD students will be encouraged. With application-based financial support from the central funds, young researchers visit the laboratories of other QUTIF members. The durations are flexible in a range from one week to several months.
- A **junior scientists' panel** headed by one representative will be formed. The panel is encouraged to have closed meetings at the annual QUTIF meetings The young scientists' representative has an advisory role towards QUTIF's coordination group.

International conferences

QUTIF will organize **two international conferences**. It is planned to combine each conference with one of the annual meetings of QUTIF, one in the second year and the other in the fifth year. The purpose of these conferences is to foster scientific exchange on the topics related to QUTIF and to guarantee the visibility of the network to the international strong-field community.

6. Justification

Novelty

The central element of this Programme is the use of electric fields tailored on the sub-femtosecond time scale in order to drive and control microscopic electronic processes and molecular dynamics. Going beyond the methods previously used in strong-field physics and femtochemistry, novel pulse-production techniques now facilitate the full control of the instantaneous electric field. This means that (i) the carrier-envelope phase in few-cycle pulses can be set arbitrarily, either by phase stabilization or by post-pulse sorting of the results, (ii) the polarization properties of laser fields can be tailored by combining sub-pulses with various different polarizations and well defined phase relations or by polarization shaping techniques. By controlling the instantaneous field, previously impossible manipulations are in reach, such as modification of molecular potential energy surfaces on a sub-femtosecond time scale. Well controlled fields are also the workhorse for making attosecond pulses, which are awaiting widespread use for real-time observation of electron

correlations in complex systems. Moreover, the production of strong ultrashort pulses in the mid-infrared regime is now becoming a widely used tool. After the past two decades were strongly dominated by the 800 nm wavelength of Ti:Sapphire laser systems, the area is now open for wavelength-dependent studies over a wide range. Many laboratories are setting up optical parametric devices to explore the mid-infrared wavelength range. One benefit of moving to mid-infrared sources is the better applicability of the three-step trajectory model of high-harmonic generation and thus the better suitability for high-harmonic spectroscopy schemes. Combining new light sources with the pulse-shaping expertise of the femtochemistry community leads to unprecedented opportunities in the control of microscopic dynamics.

Another important new development is the marriage of strong-field physics with chemistry. With our table-top laser-based methods for chiral recognition, QUTIF addresses one of the most important problems in chemistry. Traditionally the modelling of intense-field effects such as tunnelling ionization or high-harmonic generation has mostly focused on simple atoms or model systems. In contrast, recent experiments aim specifically at seeing signatures of the particular molecular structure in strong-field experiments, since the strong-field response provides rich information of the Ångström-scale structure and attosecond dynamics of the target system. Bringing together theories of quantum chemistry with strong-field physics is a challenge that we address in this Priority Programme. QUTIF places emphasis on the dynamics of quantum systems driven far away from their ground state. This distinguishes our proposal from many other activities in ultrafast molecular imaging, which usually focus on the imaging of matter in its natural state or under weak perturbations.

To summarize, the quantum dynamics in tailored intense fields is an **emerging research area**. Germany has the potential to be the leading nation in this development. To this end, a strong network must be established now within Germany to combine the existing expertise and capacities, some of which would otherwise remain isolated.

Interdisciplinarity

Physics and chemistry are the main disciplines involved in this Programme. From the physics viewpoint, strong fields offer the possibility to study multielectron dynamics on ultrashort time scales without decoherence. Controlling currents in electronic circuits by external fields leads to applications even in engineering. From the chemistry viewpoint, the interaction of controlled fields with molecular properties such as chirality and conical intersections provides new opportunities for control of fast reactions, chiral recognition and selection. Chirality plays an important role in biological systems: different enantiomers of a molecule can have entirely different effects on living beings. Our research may therefore have long-term implications for biology.

Among the research groups that are potentially involved in this Priority Programme (see the list in Appendix A), about 80% are in physics departments and 20% are in chemistry departments. It is expected that many fruitful long-term collaborations will result from this enterprise.

Synergy

German researchers are among the world leaders in the topical areas that represent the basis of this proposal, including pulse tailoring, attosecond physics, coincidence spectoscopy and coherent control. These groups follow a variety of very different approaches to closely related phenomena. This Priority Programme will foster collaborations of the strong-field communities in physics and chemistry. Only within such a network can the existing powerful experimental and theoretical methods be combined to reach the maximum possible gain of knowledge in this emerging field. Thus, QUTIF will boost the international visibility of German strong-field research.

One element of synergy arises from clear-cut new bilateral and multilateral collaborations of QUTIF participants. From the perspective of the initiators, the following activities are extremely promising:

- Paulus, Rost, Bauer and Gräfe will join forces to analyse mid-infrared ionization, in particular concerning the molecular effects which are not understood to date.
- Smirnova and Lein will cooperate via Ingo Barth (presently advanced postdoc in Hannover) on angular momentum dependence and spin dependence of strong-field dynamics. Together with the experimental colleagues (Vrakking, Moshammer, Drescher), schemes for experimental confirmation will be implemented.
- Baumert, Wollenhaupt and Lein plan to collaborate in order to reach a full understanding of the photoelectron distributions from chiral molecules, possibly supported by the Scrinzi group with respect to computational methods for circular polarization, by the Bauer group with respect to trajectory-type

modelling for chiral potentials, and by groups in quantum chemistry (Egorova, de Vivie-Riedle).

- Fennel, Kovačev, Morgner and Baumert will work together on filamentation and droplet dynamics, in particular to investigate the microscopic forces therein that drive quantum processes.
- Electron wavepackets at the subfemtosecond scale will be studied in experimental/theoretical joint work by Goulielmakis, Pfeifer, Santra and Rohringer on fundamental atomic systems and by Vrakking, Wollenhaupt, de Vivie-Riedle, Schmidt and Engel in molecules with coupled electrons and nuclei.

The other important element of synergy is the mixing of scientific communities. This cannot necessarily be pinned down to specific collaborations. For example, the coherent control community posseses deep knowledge in femtosecond pulse shaping of phase, amplitude and polarization (Brixner, Baumert, Motzkus, Wollenhaupt, Riedle), which will be of utmost value for utilizing waveform synthesis (Goulielmakis, Morgner, Kärtner, Paulus). A large network as proposed here makes sure that the available new light sources find their way to practical applications on relevant systems. On the theory side, quantum dynamics groups in chemistry (de Vivie-Riedle, Engel, Gräfe, Egorova) and in physics (Bauer, Ivanov, Lein, Saenz, Scrinzi) will be brought together by QUTIF with a huge potential for mutual benefit. To give a striking example of the existing obstacles for scientific exchange, this year's main conference in attoscience (ATTO2013, Paris, 8 – 12 July) was held in parallel to the most important femtosecond conference (FEMTO11, Copenhagen, 7 – 12 July).

7. Distinction from other Programmes

SFB 652 Rostock: *Strong correlations and collective effects in radiation fields: Coulomb systems, clusters, and particles*. The SFB in Rostock investigates primarily correlation and collective effects in many-particle systems such as clusters, molecular aggregates, nanoparticles, helium droplets and Bose-Einstein condensates. Hence, the target systems and the research focus have minor overlap with QUTIF.

SFB 631, München: *Solid-state based quantum information processing: physical concepts and materials aspects*. This SFB is devoted to quantum-information processing: the implementation of qubits and realization of qubit operations in solid-state systems. There is no overlap with our Priority Programme.

SFB/TR 49, Frankfurt/Kaiserslautern/Mainz*: Condensed Matter Systems with Variable Many-Body Interactions*. This SFB/TR treats ultracold gases and solid-state systems. It has no overlap with QUTIF.

SFB/TR 21, Stuttgart/Ulm/Tübingen: *Control of Quantum Correlations in Tailored Matter*. This SFB/TR treats ultracold spins, quantum phase transitions and hybrid quantum systems. It has no overlap with QUTIF.

SFB/TR 18, Düsseldorf/Jena/München: *Relativistic Laser-Plasma Dynamics*. Laser-plasma interactions in extremely intense fields and laser-based particle acceleration are at the heart of the SFB/TR 18. Quantum dynamics plays a minor role since the plasmas are well described by classical relativistic physics. As QUTIF works in the quantum mechanical non-relativistic regime, there is essentially no thematic overlap.

SFB 755, Göttingen: *Nanoscale photonic imaging*. This SFB pursues applied research on the imaging of biological systems down to the femtosecond scale and the development of X-ray optics for imaging. QUTIF reaches down to Ångström-scale imaging and attosecond time scales. Thus, there is essentially no overlap.

SFB 925, Hamburg: *Light induced dynamics and control of correlated quantum systems*. Quantum gases and strongly correlated systems form substantial parts of the Hamburg SFB. These topics are not covered by QUTIF. The SFB has also a component dedicated to ultrafast dynamics of small quantum systems, but the aspect of tailored fields plays a minor role. Hence, the relations to our Programme are limited. The SFB principal investigators Drescher und Santra are potential participants in QUTIF.

SPP 1391, Bielefeld/Kaiserslautern: *Ultrafast Nanooptics*. This Priority Programme has strong activities in the femtosecond spectroscopy of nanostructures, plasmon dynamics and coherent control of such systems. Although QUTIF makes efforts on investigating some selected condensed-matter systems, its main focus is on gas-phase targets and on quantum mechanical dynamics. The overlap is therefore very limited.

GRK 1355, Hamburg: *Physics with new advanced coherent radiation sources*. This Programme has some thematic overlap with QUTIF, but it is also concerned strongly with light sources outside the frame of QUTIF, such as synchrotrons, free-electron lasers or matter waves sources. Unlike QUTIF, the GRK is a training programme with limited resources for setting up research projects.

MAP: *Munich Centre for Advanced Photonics*. The MAP cluster of excellence puts strong emphasis on the development of photon-source and charged-particle source technology. MAP aims at recording

microscopic and mesoscopic phenomena with high temporal and spatial resolution, at biomedical imaging and at short-pulsed therapy. MAP is thus devoted to developing tools in photon science and to time-dependent spectroscopy of systems close to their natural state. In contrast, QUTIF focuses on the new phenomena taking place in strongly-driven systems with emphasis on quantum behaviour.

CUI: *Hamburg Center for Ultrafast Imaging*. This cluster of excellence aims at the imaging and control of a wide range of target systems including for example biomolecules or ultracold gases. The Programme has a strong focus on X-ray based imaging, while there is also some level of activity in laser-based methods. Tailored intense fields do not play a substantial role. Rather, the focus is on dynamics close to the natural or unperturbed state of the systems of interest. The overlap with QUTIF is small.

CORINF: *Correlated Multielectron Dynamics in Intense Light Fields* (EU FP7 ITN). There is a certain thematic overlap of QUTIF with the EU network CORINF, which involves several partners in Germany. However, CORINF is a *theory-only* training network with limited resources for setting up research projects. In contrast, QUTIF will strengthen the cooperation of experiment and theory. Tailored fields are not central in CORINF.

ATTOFEL: *Attosecond and free-electron-laser physics* (EU FP7 ITN). ATTOFEL is a EU training network with emphasis on light-source development, which is not the central goal of our proposal. ATTOFEL finishes in December 2013.

ELCH: *Electron dynamics of chiral systems* (LOEWE Programme Hessen). The small ELCH Programme has an element dedicated to chiral molecules in strong laser fields besides other activities using particle impact and one-photon excitation. Chirality is currently attracting the attention of many groups. QUTIF will help to form a network of these groups on a scale much beyond the local Programme in Hessen.

8. Coordination

The scientific and organizational coordination of QUTIF is handled by the *steering committee*. This includes the planning of the annual meetings, schools and international conferences, fostering scientific exchange and collaborations within the network and deciding on financial support for young scientists and for disadvantaged researchers. The academic experience of the committee members covers the relevant fields of experimental and theoretical physics as well as chemistry. The steering committee is available to give advice to participating researchers about the scientific programme of QUTIF as well as possible collaborations. The *Coordinator* and the *Vice-Coordinator* are the heads of the steering committee. They communicate between the DFG and the participants of the Programme. Their main task within the network is to encourage joint research.

To promote young scientists and to guarantee equal opportunity, the steering committee evaluates the research conditions for young group leaders and female researchers. Based on a semi-annual application procedure, the steering committee decides on small start-up grants for young scientists and on financial support of secondments. Furthermore, financial and organizational support will be given to researchers who are disadvantaged for reasons of gender or disability. Requests can be directed at any time to the steering committee. Gender-equality support covers childcare for researchers in QUTIF in cases such as conference travel, secondments or school holidays, and, if appropriate, home-office equipment. For families, QUTIF will contribute to job-related relocation costs and to increased conference travel costs.

Steering committee:

Manfred Lein (Coordinator)	Institute for Theoretical Physics, Leibniz Universität Hannover
Gerhard Paulus (Vice-Coordinator)	Institute of Optics and Quantum Electronics, Friedrich-Schiller-Univ. Jena
Uwe Morgner	Institute for Quantum Optics, Leibniz Universität Hannover
Jan Michael Rost	Max Planck Institute for the Physics of Complex Systems Dresden
Regina de Vivie-Riedle	Department Chemie, Ludwig-Maximilians-Universität München
Marc Vrakking	Max Born Institute Berlin

9. Funding periods

We request a Priority Programme with funding for **2 times 3 years.**

10. Estimation of required funding for the first three-year period

The project costs are estimated from experience in the initiators' research groups. Junior projects are targeted at young group leaders with higher need for investment. Investment costs cover laser systems and detectors for experimental projects. In theory projects involving heavy numerical research, servers for parallelized simulations are needed. To this end, we estimate 30000 EUR in each junior theory project and in three out of nine regular theory projects. About 85% of the hired personnel will be PhD students on 75% salary (estimated 45124 EUR p.a. by assuming a 5% increase relative to the current DFG number of 0.75x57300 EUR p.a.). About 15% will be postdocs on full salary (estimated 65205 EUR p.a.). This leads to average personnel costs of 144400 EUR in a three-year project. Experimental projects include also an estimated amount of 6000 EUR for student assistants. The temporary positions are at the postdoc level (65205 EUR p.a.). These are targeted at junior project leaders who apply for funding of their own position. The detailed individual amounts per project will be given in the project proposals.

The centre project covers the QUTIF meetings and schools, see Section 5 (module "Project-Specific Workshops", 20000 EUR p.a.), funds for gender equality, see Section 8 (module "Gender Equality Measures in Research Networks", 15000 EUR p.a.), start-up grants, see Section 8 (module "Start-Up Funding", 20000 EUR p.a.), scientific visitors and secondments of group members (module "Network Funds", 8000 p.a.) and other consumables for coordination (3000 p.a.). Personnel costs in the centre project cover a half-time office assistant position (67100 EUR for three years).

The estimation of required funding is summarized in the following table for the first three-year funding period. The amounts in experimental and theory projects are average numbers.

	Consumables	Invest	Personnel	Project sum	Total sum
16 projects - experiment	**60 000,-**	**100 000,-**	**150 400,-**	**310 400,-**	**4 966 400,-**
9 projects - theory	**15 000,-**	**10 000,-**	**144 400,-**	**169 400,-**	**1 524 600,-**
2 junior projects - experiment	**60 000,-**	**150 000,-**	**150 400,-**	**360 400,-**	**720 800,-**
2 junior projects - theory	**15 000,-**	**30 000,-**	**144 400,-**	**189 400,-**	**378 800,-**
2 temporary positions ("Eigene Stelle")	**--------**	**--------**	**195 615,-**	**195 615,-**	**391 230,-**
Centre project	**198 000,-**	**--------**	**67 100,-**	**265 100,-**	**265 100,-**
Total sum (€)					**8 246 930,-**

(All amounts are given in EUR.)

11. Bibliography

Author names from the QUTIF initiator group appear in **boldface**.

[1] A.M. Weiner, J.P. Heritage, E.M. Kirschner. *J. Opt. Soc. Am. B* 5, 1563 (1988).
[2] A. Assion, T. Baumert, M. Bergt, T. Brixner, B. Kiefer, V. Seyfried, M. Strehle, G. Gerber. *Science* 282, 919 (1998).
[3] J.L. Herek, W. Wohlleben, R.J. Cogdell, D. Zeidler, M. Motzkus. *Nature* 417, 533 (2002).
[4] R.J. Levis, G.M. Menkir, H. Rabitz. *Science* 292, 709 (2001).
[5] T. Brixner, G. Krampert, T. Pfeiffer, R. Selle, G. Gerber, M. Wollenhaupt, O. Gräfe, C. Horn, D. Liese, T. Baumert. *Phys. Rev. Lett.* 92, 208301 (2004).
[6] **U. Morgner**, F.X. Kärtner, S.H. Cho, Y. Chen, H.A. Haus, J.G. Fujimoto, E.P. Ippen, V. Scheuer, G. Angelow, T. Tschudi. *Opt. Lett.* 24, 411 (1999).
[7] M. Nisoli, S. De Silvestri, O. Svelto, R. Szipöcs, K. Ferencz, C. Spielmann, S. Sartania, F. Krausz. *Opt. Lett.* 22, 522 (1997).
[8] J. Reichert, R. Holzwarth, T. Udem, T.W. Hänsch. *Opt. Comm.* 172, 59 (1999).

[9] H. Telle, G. Steinmeyer, A. Dunlop, J. Stenger, D. Sutter, U. Keller. *Appl. Phys. B* 69, 327 (1999).
[10] **G.G. Paulus**, F. Grasbon, H. Walther, P. Villoresi, M. Nisoli, S. Stagira, E. Priori, S. De Silvestri. *Nature* 414, 182 (2001).
[11] T. Wittmann, B. Horvath, W. Helml, M. Schätzel, X. Gu, A. Cavalieri, **G.G. Paulus**, R. Kienberger. *Nature Phys.* 5, 357 (2009).
[12] D. Schuhmacher, F. Weihe, H.G. Muller, P.H. Bucksbaum. *Phys. Rev. Lett.* 73, 1344 (1994).
[13] **G.G. Paulus**, W. Becker, H. Walther. *Phys. Rev. A* 52, 4043 (1995).
[14] O. Raz, O. Pedatzur, B.D. Bruner, N. Dudovich. *Nat. Phot.* 6, 170 (2012).
[15] A. Baltuška, T. Udem, M. Uiberacker, M. Hentschel, E. Goulielmakis, C. Gohle, R. Holzwarth, V.S. Yakovlev, A. Scrinzi, T.W. Hänsch, F. Krausz. *Nature* 421, 611 (2003).
[16] N.G. Johnson, et al. *Phys. Rev. A* 83, 013412 (2011).
[17] Z. Chang. *Phys. Rev. A* 70, 043802 (2004).
[18] I. Sola, et al. *Nature Phys.* 2, 327 (2006).
[19] A. Wirth, M.T. Hassan, I. Grguraš, J. Gagnon, A. Moulet, T.T. Luu, S. Pabst, R. Santra, Z.A. Alahmed, A.M. Azzeer, V.S. Yakovlev, V. Pervak, F. Krausz, E. Goulielmakis. *Science* 334, 195 (2011).
[20] A. Harth, M. Schultze, T. Lang, T. Binhammer, S. Rausch, **U. Morgner**. *Opt. Express* 20, 3076 (2012).
[21] J. Köhler, M. Wollenhaupt, T. Bayer, C. Sarpe, T. Baumert. *Opt. Express* 19, 11638 (2011).
[22] A. McPherson, et al. *J. Opt. Soc. Am. B* 4, 595 (1987).
[23] A. L'Huillier, K.J. Schafer, K.C. Kulander. *J. Phys. B* 24, 3315 (1991).
[24] M. Lewenstein, P. Balcou, M. Y. Ivanov, A. L'Huillier, P.B. Corkum. *Phys. Rev. A* 49, 2117 (1994).
[25] P.M. Paul, E.S. Toma, P. Breger, G. Mullot, F. Augé, P. Balcou, H.G. Muller, P. Agostini. *Science* 292, 1689 (2001).
[26] M. Hentschel, R. Kienberger, C. Spielmann, G. A. Reider, N. Milosevic, T. Brabec, P. Corkum, U. Heinzmann, M. Drescher, F. Krausz. *Nature* 414, 509 (2001).
[27] R. Velotta, N. Hay, M.B. Mason, M. Castillejo, J.P. Marangos. *Phys. Rev. Lett.* 87, 183901 (2001).
[28] F. Rosca-Pruna, **M.J.J. Vrakking**. *Phys. Rev. Lett.* 85, 4024 (2001).
[29] **M. Lein**, N. Hay, R. Velotta, J.P. Marangos, P.L. Knight. *Phys. Rev. Lett.* 88, 183903 (2002).
[30] J. Itatani, J. Levesque, D. Zeidler, H. Niikura, H. Pépin, J.C. Kieffer, P.B. Corkum, D.M. Villeneuve. *Nature* 432, 867 (2004).
[31] H.J. Wörner. *Chimia* 65, 299 (2011).
[32] P.B. Corkum. *Phys. Rev. Lett.* 71, 1994 (1993).
[33] **M. Lein**. *Phys. Rev. Lett.* 94, 053004 (2005).
[34] S. Baker, J. Robinson, C.A. Haworth, H. Teng, R.A. Smith, C.C. Chirilă, **M. Lein**, J.W.G. Tisch, J.P. Marangos. *Science* 312, 424 (2006).
[35] J.P. Farrell, S. Petretti, J. Förster, B.K. McFarland, L.S. Spector, Y.V. Vanne, P. Decleva, P.H. Bucksbaum, A. Saenz, M. Gühr. *Phys. Rev. Lett.* 107, 083001 (2011).
[36] J. Förster, A. Saenz. *ChemPhysChem* 14, 1438 (2013).
[37] P.M. Kraus, H.J. Wörner. *ChemPhysChem* 14, 1445 (2013).
[38] Y. Mairesse, A. de Bohan, L.J. Frasinski, H. Merdji, L.C. Dinu, P. Monchicourt, P. Breger, M. Kovačev, R. Taïeb, B. Carré, H.G. Muller, P. Agostini, P. Salières. *Science* 302, 1540 (2003).
[39] C.I. Blaga, J. Xu, A.D. DiChiara, E. Sistrunk, K. Zhang, P. Agostini, T.A. Miller, L.F. DiMauro, C.D. Lin. *Nature* 483, 194 (2012).
[40] B.K. McFarland, J.P. Farrell, P.H. Bucksbaum, M. Gühr. *Science* 322, 1232 (2008).
[41] O. Smirnova, Y. Mairesse, S. Patchkovskii, N. Dudovich, D.M. Villeneuve, P.B. Corkum, M.Y. Ivanov. *Nature* 460, 972 (2009).
[42] S. Hässler, J. Caillat, W. Boutu, C. Giovanetti-Teixeira, T. Ruchon, T. Auguste, Z. Diveki, P. Breger, A. Maquet, B. Carré, R. Taïeb, P. Salières. *Nature Phys.* 6, 200 (2010).
[43] C. Vozzi, M. Negro, F. Calegari, G. Sansone, M. Nisoli, S. De Silvestri, S. Stagira. *Nature Phys.* 7, 822 (2011).
[44] H.J. Wörner, et al. *Science* 334, 208 (2011).
[45] M.F. Kling, P. von den Hoff, I. Znakovskaya, **R. de Vivie-Riedle**. *Phys. Chem. Chem. Phys.* 15, 9448 (2013).
[46] D. Shafir, H. Soifer, B.D. Bruner, M. Dagan, Y. Mairesse, S. Patchkovskii, M.Y. Ivanov, O. Smirnova, N. Dudovich. *Nature* 485, 343 (2012).
[47] P. Salières, B. Carré, L. Le Déroff, F. Grasbon, **G.G. Paulus**, H. Walther, R. Kopold, W. Becker, D.B. Milošević, A. Sanpera, M. Lewenstein. *Science* 292, 902 (2001).
[48] J. Zhao, **M. Lein**. *Phys. Rev. Lett.* 111, 043901 (2013).

[49] D.S. Steingrube, E. Schulz, T. Binhammer, M.B. Gaarde, A. Couairon, **U. Morgner**, M. Kovačev. *New. J. Phys.* 13, 043022 (2011).
[50] H.G. Kurz, D.S. Steingrube, D. Ristau, **M. Lein**, **U. Morgner**, M. Kovačev. *Phys. Rev. A.* 87, 063811 (2013).
[51] R. Dörner, V. Mergel, O. Jagutzki, L. Spielberger, J. Ullrich, R. Moshammer, H. Schmidt-Böcking. *Phys. Rep.* 330, 95 (2000).
[52] R. Moshammer, et al. *Phys. Rev. Lett.* 84, 447 (2000).
[53] T. Weber, H. Giessen, M. Weckenbrock, G. Urbasch, A. Staudte, L. Spielberger, O. Jagutzki, V. Mergel, M. Vollmer, R. Dörner. *Nature* 405, 658 (2000).
[54] A.T.J.B. Eppink, D.H. Parker. *Rev. Sci. Instr.* 68, 3477 (1997).
[55] M. Wollenhaupt, C. Lux, T. Krug, T. Baumert. *ChemPhysChem* 14, 1341 (2013).
[56] J. Maurer, D. Dimitrovski, L. Christensen, L.B. Madsen, H. Stapelfeldt. *Phys. Rev. Lett.* 109, 123001 (2012).
[57] I. Znakovskaya, P. von den Hoff, G. Marcus, S. Zherebtsov, B. Bergues, X. Gu, Y. Deng, **M.J.J. Vrakking**, R. Kienberger, F. Krausz, **R. de Vivie-Riedle**, M.F. Kling. *Phys. Rev. Lett.* 108, 063002 (2012).
[58] M. Schultze, et al. *Science* 328, 1658 (2010).
[59] K. Klünder, et al. *Phys. Rev. Lett.* 106, 143002 (2012).
[60] S. Nagele, R. Pazourek, J. Feist, J. Burgdörfer. *Phys. Rev. A.* 85, 033401 (2012).
[61] M. Ivanov, O. Smirnova. *Phys. Rev. Lett.* 107, 213605 (2011).
[62] A.S. Kheifets. *Phys. Rev. A.* 87, 063404 (2013).
[63] P. Eckle, A.N. Pfeiffer. C. Cirelli, A. Staudte, R. Dörner, H.G. Muller, M. Büttiker, U. Keller. *Science* 322, 1525 (2008).
[64] A. Landsman, M. Weger, J. Maurer, R. Boge, A. Ludwig, S. Heuser, C. Cirelli, L. Gallmann, U. Keller. arXiv:1301.2766v2 (2013).
[65] M. Odenweller, N. Takemoto, A. Vredenborg, K. Cole, K. Pahl, J. Titze, L.P.H. Schmidt, T. Jahnke, R. Dörner, A. Becker. *Phys. Rev. Lett.* 107, 143004 (2011).
[66] J. Wu, M. Meckel, S. Voss, H. Sann, M. Kunitski, L.P.H. Schmidt, A. Czasch, H. Kim, T. Jahnke, R. Dörner. *Phys. Rev. Lett.* 108, 043002 (2012).
[67] J. Wu, L.P.H. Schmidt, M. Kunitski, M. Meckel, S. Voss, H. Sann, H. Kim, T. Jahnke, A. Czasch, R. Dörner. *Phys. Rev. Lett.* 108, 183001 (2012).
[68] T. Frohnmeyer, M. Hofmann, M. Strehle, T. Baumert. *Chem. Phys. Lett.* 312, 447 (1999).
[69] B.J. Sussman, D. Townsend, M.Y. Ivanov, A. Stolow. *Science* 314, 278 (2006).
[70] M.F. Gelin, D. Egorova, W. Domcke. *J. Phys. Chem. Lett.* 2, 114 (2011).
[71] A. Schiffrin, et al. *Nature* 493, 70 (2012).
[72] N. Böwering, T. Lischke, B. Schmidtke, N. Müller, T. Khalil, U. Heinzmann. *Phys. Rev. Lett.* 86, 1187 (2001).
[73] I. Powis. *J. Phys. Chem. A* 104, 878 (2000).
[74] B. Ritchie. *Phys. Rev. A* 13, 1411 (1976).
[75] C. Lux, M. Wollenhaupt, T. Bolze, Q. Liang, J. Köhler, C. Sarpe, T. Baumert. *Angew. Chem. Int. Ed.* 51, 5001 (2012).
[76] H.G. Breunig, G. Urbasch, P. Horsch, J. Cordes, U. Koert, K.-M. Weitzel. *ChemPhysChem* 10, 1199 (2009).
[77] M. Pitzer, et al. *Science* 341, 1096 (2013).
[78] M.C.H. Wong, J.-P. Brichta, M. Spanner, S. Patchkovskii, V.R. Bhardwaj. *Phys. Rev. A* 84, 051403(R) (2011).
[79] M. Shapiro, E. Frishman, P. Brumer. *Phys. Rev. Lett.* 84, 1669 (2000).
[80] Y. Fujimura, L. Gonzalez, K. Hoki, D. Kröner, J. Manz, Y. Ohtsuki. *Angew. Chem. Int. Ed.* 39, 4586 (2000).
[81] C. Vozzi, M. Negro, S. Stagira. *J. Mod. Opt.* 59, 1283 (2012).
[82] A.D. Shiner, B.E. Schmidt, C. Trallero-Herrero, P.B. Corkum, J.-C. Kieffer, F. Légaré, D.M. Villeneuve. *J. Phys. B* 45, 074010 (2012).
[83] A.D. Shiner, et al. *Nature Phys.* 7, 464 (2011).
[84] C. Vozzi, et al. *Appl. Phys. Lett.* 97, 241103 (2010).
[85] Y. Huismans, ..., **M.J.J. Vrakking**. *Science* 331, 61 (2011).
[86] C.I. Blaga, F. Catoire, P. Colosimo, **G.G. Paulus**, H.G. Muller, P. Agostini, L.F. DiMauro. *Nature Phys.* 5, 335 (2009).

[87] T.-M. Yan, S.V. Popruzhenko, **M.J.J. Vrakking**, D. Bauer. *Phys. Rev. Lett.* 105, 253002 (2010).
[88] C. Lemell, K.I. Dimitriou, X.-M. Tong, S. Nagele, D.V. Kartashov, J. Burgdörfer, S. Gräfe. *Phys. Rev. A* 85, 011403(R) (2012).
[89] A. Kästner, U. Saalmann, **J.M. Rost**. *Phys. Rev. Lett.* 108, 033201 (2012).
[90] **M. Lein**, E. K. U. Gross, V. Engel. *Phys. Rev. Lett.* 85, 4707 (2000).
[91] C. Ott, A. Kaldun, P. Raith, K. Meyer, M. Laux, J. Evers, C.H. Keitel, C.H. Greene, T. Pfeifer. *Science* 340, 716 (2013).
[92] A.N. Pfeiffer, C. Cirelli, M. Smolarski, X. Wang, J.H. Eberly, R. Dörner, U. Keller. *New J. Phys.* 13, 093008 (2011).
[93] K.C. Kulander, K.J. Schafer, J.L. Krause. *Proc. SILAP (*1993).
[94] **G.G. Paulus**, W. Becker, W. Nicklich, H. Walther. *J. Phys. B* 27, L703 (1994).
[95] I. Barth, O. Smirnova. *Phys. Rev. A* 87, 065401 (2013).
[96] R. Boge, C. Cirelli, A.S. Landsman, S. Heuser, A. Ludwig, J. Maurer, M. Weger, L. Gallmann, U. Keller. *Phys. Rev. Lett.* 111, 103003 (2013).
[97] C. Liu, M. Reduzzi, A. Trabattoni, A. Sunilkumar, A. Dubrouil, F. Calegari, M. Nisoli, G. Sansone. *Phys. Rev. Lett.* 111, 123901 (2013).
[98] E. Goulielmakis, Z.-H. Loh, A. Wirth, R. Santra, N. Rohringer, V.S. Yakovlev, S. Zherebtsov, T. Pfeifer, A.M. Azzeer, M.F. Kling, S.R. Leone, F. Krausz. *Nature* 466, 739 (2010).
[99] I. Barth, O. Smirnova. *Phys. Rev. A* 88, 013401 (2013).
[100] X. Xie, S. Roither, D. Kartashov, E. Persson, D.G. Arbó, L. Zhang, S. Gräfe, M.S. Schöffler, J. Burgdörfer, A. Baltuška, M. Kitzler. *Phys. Rev.Lett.* 108, 193004 (2012).
[101] J. Breidbach, L. Cederbaum. *Phys. Rev. Lett.* 94, 033901 (2005).
[102] H. Stapelfeldt, T. Seideman. *Rev. Mod. Phys.* 75, 543 (2003).
[103] O. Ghafur, A. Rouzée, A. Gijsbertsen, W.K. Siu, S. Stolte, **M.J.J. Vrakking**. *Nature Phys.* 5, 289 (2009).
[104] S. De, I. Znakovskaya, D. Ray, F. Anis, N. Johnson, I.A. Bocharova, M. Magrakvelidze, B.D. Esry, C.L. Cocke, I.V. Litvinyuk, M.F. Kling. *Phys. Rev. Lett.* 103, 153002 (2009).
[105] P.M. Kraus, A. Rupenyan, H.J. Wörner. *Phys. Rev. Lett.* 109, 233903 (2012).
[106] A. Chacon, **M. Lein**, C. Ruiz. *Phys. Rev. A* 87, 023408 (2013).
[107] A.E. Boguslavskiy, J. Mikosch, A. Gijsbertsen, M. Spanner, S. Patchkovskii, N. Gador, **M.J.J. Vrakking**, A. Stolow. *Science* 335, 1336 (2012).
[108] Z. Ansari, M. Böttcher, B. Manschwetus, H. Rottke, W. Sandner, A. Verhoef, M. Lezius, **G.G. Paulus**, A. Saenz, D.B. Milošević. *New J. Phys.* 10, 093027 (2008).
[109] M.F. Kling, C. Siedschlag, A.J. Verhoef, J.I. Khan, M. Schultze, T. Uphues, Y. Ni, M. Uiberacker, M. Drescher, F. Krausz, **M.J.J. Vrakking**. *Science* 312, 246 (2006).
[110] X. Xie, K. Doblhoff-Dier, S. Roither, M.S. Schöffler, D. Kartashov, H. Xu, T. Rathje, **G.G. Paulus**, A. Baltuška, S. Gräfe, M. Kitzler. *Phys. Rev. Lett.* 109, 243001 (2012).
[111] T. Bayer, H. Braun, C. Sarpe, R. Siemering, P. von den Hoff, **R. de Vivie-Riedle**, T. Baumert, M. Wollenhaupt. *Phys. Rev. Lett.* 110, 123003 (2013).
[112] A. Flettner, T. Pfeifer, D. Walter, C. Winterfeld, C. Spielmann, G. Gerber. *Appl. Phys. B* 77, 747 (2003).
[113] S.R. Krishnan, L. Fechner, M. Kremer, V. Sharma, B. Fischer, N. Camus, J. Jha, M. Krishnamurthy, T. Pfeifer, R. Moshammer, J. Ullrich, F. Stienkemeier, M. Mudrich, A. Mikaberidze, U. Saalmann, **J.M. Rost**. *Phys. Rev. Lett.* 107, 173402 (2011).
[114] A. Mikaberidze, U. Saalmann, **J.M. Rost**. *Phys. Rev. Lett.* 102, 128102 (2009).
[115] M. Richter, S. Patchkovskii, F. Morales, O. Smirnova, M. Ivanov. *New. J. Phys.* 15, 083012 (2013).
[116] L. Englert, M. Wollenhaupt, C. Sarpe, D. Otto, T. Baumert. *J. Laser Appl.* 24, 042002 (2012).
[117] A.L. Cavalieri, et al. *Nature* 449, 1029 (2007).
[118] S. Zherebstov, et al. *Nature Phys.* 7, 656 (2011).
[119] M. Krüger, M. Schenk, P. Hommelhoff. *Nature* 475, 78 (2011).
[120] U. Saalmann, C. Siedschlag, **J.M. Rost**. *J. Phys. B* 39, R39 (2006).
[121] U. Saalmann, **J.M. Rost**. *Phys. Rev. Lett.* 100, 133006 (2008).
[122] H. Ruf, et al. *Phys. Rev. Lett.* 110, 083902 (2013).
[123] T. Rathje, A.M. Sayler, S. Zeng, P. Wustelt, H. Figger, B.D. Esry, **G.G. Paulus**. *Phys. Rev. Lett.* 111, 093002 (2013).
[124] **M. Lein**. *J. Phys. B* 40, R135 (2007).
[125] I. Barth, O. Smirnova. *Phys. Rev. A* 87, 013433 (2013).
[126] A. Becker, F.H.M. Faisal. *J. Phys. B* 38, R1 (2005).
[127] A.M. Perelomov, V.S. Popov, M.V. Terent'ev. *Sov. Phys. JETP* 23, 924 (1966).