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Attosecond control of spin polarization in electron-ion recollision driven by intense tailored fields

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Ionization of noble gases by strong infrared circularly-polarized laser pulses can produce electron currents with a controllable degree of spin polarization [1-4]. Spin polarization arises as a consequence of (1) entanglement between the liberated electron and the parent ion, and (2) sensitivity of ionization to the sense of electron rotation in the initial state. In this context, the use of two-color counter-rotating bicircular fields [5] opens new opportunities for introducing the spin degree of freedom into attosecond science [6], since the liberated electrons can be driven back towards
the ionic core within one optical cycle.

In this conference we will present theoretical evidence of spin polarization of the electrons recolliding with xenon atoms upon tunnel ionization by a strong circularly polarized laser field in combination with a counter-rotating second harmonic. We have found that degree of polarization depends strongly on the electron's excursion time and therefore on the recollision energy [7]. Furthermore, we will show that a high degree of control can be achieved by varying the intensities of the counter-rotating
fields.

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Brunel Harmonics: Signatures of Electron Ionization Dynamics and Propagation in the Continuum

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Laser driven nonadiabatic electron dynamics in molecules

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We discuss theoretical analysis of nonadiabatic electron dynamics emerging from molecular systems driven by intense femtosecond laser fields [1]. These nonadiabatic dynamics have been seen to modify ionization and fragmentation behavior in large molecular systems driven by mid-IR laser fields. The hydrogen molecular ion has proven an exceptional model system for simulating the nonadiabatic features found in larger molecules. When dissociating, H$_2$^+$ passes through a regime of internuclear distances which are accompanied by near-degeneracy of the ground and first-excited state. The strong coupling of these states to the driving laser field enhances ionization rates and induces transient

localization of the electron wavepacket, mimicking the charge density modulation theoretically observed in larger molecules and the increased ionization experimentally reported [2,3]. The capacity for H$_2$^$ to reproduce nonadiabatic dynamics was further explored as a means of testing the influence of nonadiabatic electron dynamics upon common signatures of strong-field physics. In particular, transient electron localization was shown to strongly alter fragmentation behavior upon molecular dissociation [4], modify photoelectron distributions after laser-induced ionization [5], and to modulate the high energy plateau of HHG, introducing spectral minima reflective of transiently suppressed ionization[6,7].

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Strong-Field Approximation with Twisted Light Beams

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The strong-field approximation has successfully been used to theoretically describe a variety of phenomena related to atoms in strong laser fields, e. g. above-threshold ionization and high-harmonic generation. Usually these studies are performed for the case of plane wave fields. On the other hand, the interaction of light beams with a more complex spatial structure (e. g. Bessel beams) with atomic systems has only been treated perturbatively and has, for example, been shown to lead to a modification of the selection rules of atomic transitions and photoelectron momentum spectra. We discuss the possibility to study the photoionization of atoms by intense Bessel beams using the strong-field approximation.

Soft X-ray shines on the new attosecond horizon through the water window

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Strong-Field Ionisation with Tailored Two-Color Laser Fields

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We plan to report on our results on strong-field ionization in tailored two-colour (780 nm & 390 nm) laser fields. The 3D-momenta of the fragments are recorded using Cold Target Recoil Ion Momentum Spectroscopy (COLTRIMS) as experimental technique.

Modelling strong-field ionization of a rare-gas dimer: the effect of electron correlation

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We present a model for strong-field ionization of neutral diatomic molecules in terms of a two-level system describing the molecular ion, coupled to an outermost electron that is subject to the laser field and an effective potential depending on the ion state. The model is applied to ionization of Neon-2 molecules and compared with recent measurements by the Dörner-Group from Frankfurt. It is shown that the effects of electron correlation, or more precisely the correlation between outgoing electron and residual ion, have to be taken into account in order not to overemphasize scattering effects in the momentum distribution.

Ultrafast spin currents at ferromagnet/metal interfaces analyzed with femtosecond time-resolved non-linear optics

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Femtosecond (fs) laser excitation of ferromagnetic solids leads to a non-equilibrium state in which the magnetization is strongly reduced within a few tens of fs [1]. Recently, ultrafast spin currents have in particular received attention both as one contribution to this fs demagnetization [2] and a new opportunity to manipulate magnetic materials via spin injection or spin transfer torque on ultrafast time scales. With regards to a better understanding of the fundamental quantum processes in the interaction of fs laser pulses with spins as well as potential future applications, it is furthermore desirable to analyze how magnetization in a ferromagnetic solid can be directly manipulated by tailoring laser pulses.
As a first step, we identify laser-induced spin currents in epitaxial Co/Cu(001), which constitutes a model system for spin transport across ferromagnet/metal interfaces, via interface-sensititve magnetization-induced second harmonic generation (mSHG). We relate the transient, Co thickness dependent behavior of the mSHG signal in a 0.4-10 nm thickness range to a transient spatial profile of the Co magnetization, via the interference of the contributions to mSHG from the vacuum/Co and Co/Cu interfaces. From this we conclude that the fs spin current into the Cu substrate has an effective mean free path of about 3 nm [3]. We discuss these results in comparison with ab-initio time-dependent density functional theory calculations on the very same material system, which take into account both spin currents as well as the direct interaction of the spins with the laser pulse mediated by spin-orbit coupling.
We acknowledge funding by the DFG through SPP 1840 QUTIF.
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Controlling photoemission with taylored near-fields

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Strong-field ionization of two-dimensional molecular model systems by circularly polarized laser fields

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We have developed a numerical model system for a single active electron on a two-dimensional spatial grid in an asymmetric environment of nuclei with tunable charges. This enables mimicking of both

symmetric and asymmetric molecular systems, while keeping the ionization potential fixed. We investigate the interaction of these model systems with intense, few-cycle, circularly polarized infrared laser pulses and examine the asymmetries between the photoelectron spectra induced by the left- and right-polarized fields. We show that these asymmetries are influenced by the initial symmetry of the model systems and demonstrate that few-cycle laser pulses can induce an additional asymmetry in the photoelectron spectra, which can weaken or enhance the asymmetry imprinted by the initial symmetry properties of the model system.

Ellipticity Controlled Sideband Generation in Solids

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High harmonic generation in solids is fundamentally different from its atomic counterpart, which is highlighted by the emergence of even and odd harmonics, as well as by electronic quantum interferences in multi-band systems [1]. At the same time, one can realize harmonic sideband generation in solids by modulating an optically excited transition with a strong terahertz (THz) driving field to generate sidebands around it [2]. We will discuss why this scenario is in many ways analogous to atomic high harmonic generation. For example, it leads to generation of sidebands of even orders of the THz field around the optical photon energy. The unique combination of sources available at DESY provide a testbed for understanding these physical phenomena, by opening up intriguing new avenues for experimentally realizing a 0.3THz source with 0.3 GV/m field strength [3] and petahertz/terahertz electronics [4]. We present developments in experiment-theory collaboration, and discuss the effect of elliptically polarized driving THz fields.

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Generation and control of elliptically polarized attosecond pulses with bicircular fields

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Circular or highly elliptical femtosecond and attosecond pulses in the extreme ultraviolet spectral range present numerous applications in chiral-sensitive light-matter interactions^${1,2}. Until recently, such radiation has only been available at large-scale facilities, where the time resolution is above 100 femtoseconds. Generation of coherent light sources with attosecond duration and controllable ellipticity will enable complementary studies on ultrafast time-scales.

An elegant approach to the generation of such pulses consists of combining a circularly polarized fundamental field with a counter-rotating second harmonic^3. This scheme leads to harmonic peaks at (3N+1) and (3N+2) lines, with the helicity of the fundamental field and the second harmonic, respectively, while 3N harmonics are absent due to symmetry. Recent theoretical studies^{4,5} have shown that when neon is ionized by such bicircular fields, a considerable amount of suppression of the (3N+2)-lines is observed in the high harmonic spectrum. The reason behind this effect is still not well understood and thus, while these works opened the way to the generation of elliptically polarized attosecond pulses, control over such ellipticity is still to be achieved. Moreover, recent experiments have reported the appearance of forbidden 3N harmonics even for long and perfectly three-fold-symmetric pulses, implying a breaking of the symmetry that is yet to be explained.

In this work, we provide an in-depth analytical analysis of the high harmonic generation process in two-color counter-rotating circular fields. We do so using an analytical model based on the strong field approximation and by solving the time-dependent Schrödinger equation in the single-active electron approximation for both helium and neon, comparing our predictions to experiment. In particular, we outline the reasons behind the appearance of forbidden harmonics and derive three propensity rules which are responsible for the suppression of the (3N+2)-lines in the high harmonic spectrum of neon. By changing the relative intensity and time delay between the two pulses, we show that we can coherently control the symmetry breaking of the system and the ellipticity of the generated attosecond bursts.

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Quantum control with smoothly varying pulses: Application to charge migration

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By using direct search algorithms for solving the quantum optimization problem, we demonstrate on model systems that with specifically tailored Gaussian-form laser pulses one can achieve a very good control over the dynamics in complicated quantum systems. We show that by manipulating a very limited number of laser-pulse parameters, one is able to control the charge migration process in molecules. In particular, by combining two identical Gaussian laser pulses with an appropriate delay between them, one can stop the pure electronic, few-femtosecond oscillation of the charge, redistributing it along the molecule of propiolic acid.

Laser-induced electron diffraction of controlled molecules

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Two-color excitation of image potential states and above threshold ionization at gold nanotips

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In atomic and molecular gases, excitation with high-intensity femtosecond laser pulses results in spectacular phenomena like above-threshold ionization and high harmonic generation. In solid state media, metallic nanotapers in principle also give access to strong field phenomena [1-3]. However, in

contrast to atomic systems, electrons in solid state systems are emitted from a continuum of states near the Fermi level, such that the observation of above-threshold ionization is obscured. A possible improvement to such experiments is offered by surface states, which have been established by two-photon photoemission experiments on bulk solid state systems [4].

Here we use femtosecond two-color photoemission to study the excitation and the dynamics of image potential states on a metallic nanotip. We measure kinetic energy spectra of the photoemitted electrons as a function between the visible pump pulse and the near-infrared probe pulse. Atomic-like above-threshold ionization spectra are observed, which indicate the excitation of long-lived, weakly bound states at the surface of the nanostructure. With this work we take a step towards the emission of cold electrons as a promising source for ultrafast electron microscopy.

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Ionization dynamics of current-carrying orbitals by strong circularly polarized laser fields

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We will report the recent progress of our research regarding the ionization of the current-carrying orbitals $p_\pm$ (argon), $\pi_\pm$ (nitric oxide), and $e_\pm$ (benzene) by strong circularly polarized laser fields. Our work is based on the numerical solution of the 3D TDSE and the calculation of 3D photoelectron momentum distribution (PMD).

Molecular processes in a bicircular laser field

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We consider various atomic and molecular processes induced or assisted by a bicircular laser field. Bicircular field, which consists of two coplanar counter-rotating circularly polarized fields of frequency rω and sω (r and s are integers), possesses particular dynamical symmetry. In the case of polyatomic molecules, this symmetry can be adjusted to the symmetry of the molecular Hamiltonian and used to investigate the molecular symmetry. For example, in high-order harmonic generation process, for polyatomic molecules having the Cr+s symmetry, only the harmonics n = q(r + s)±r, q = 1,2,..., are emitted having the ellipticity εn =±1. We illustrate this using the example of the planar molecules BH3 and BF3, which obey the C3 symmetry. We will also present results for high-order above-threshold ionization (HATI) of molecular systems. In the case of homonuclear diatomic molecules, we have identified four symmetries which are satisfied in the (H)ATI process. Two of these symmetries are general rotational symmetries valid both for direct and rescattered HATI electrons. The remaining two symmetries are reflection symmetries valid only for the direct ATI electrons.

Tailoring the vacuum fluctuations of the electric field

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I will discuss the time-resolved behavior of the photonic ground state and show that vacuum fluctuations

of its electric field can be directly detected using the linear electro-optic effect. I will sketch the main aspects of a general paraxial theory of electro-optic sampling of quantum fields developed for this purpose. Our calculations and experimental results demonstrate that nonlinear mixing of a femtosecond near-infrared probe pulse with the multi-terahertz vacuum field in a thin electro-optic crystal leads to an increase of the signal variance with respect to the shot noise level. Moreover, with another femtosecond pump pulse interacting with the vacuum in an additional nonlinear optical crystal, we can modify the ground state of the field that leads to the generation of a pulsed squeezed vacuum state. Our findings shall pave the way for a new approach to quantum optics operating with an extreme, subcycle time resolution.

Imaging Excited State Dynamics of Doped He Nanodroplets in Real-Time

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The real-time dynamics of excited alkali metal atoms (Rb) attached to quantum fluid He nanodroplets is investigated using femtosecond imaging spectroscopy and time-dependent density functional theory (TDDFT). We disentangle the competing dynamics of desorption of excited Rb atoms off the He droplet surface and solvation inside the droplet interior as the Rb atom is ionized. For Rb excited to the states 5p and 6p desorption occurs mostly impulsively but on starkly differing time scales (~100 ps versus ~1 ps, respectively). RbHe exciplex formation and desorption as well as He droplet induced electronic relaxation is is discussed.

Ionization and dissociation of HeH^+ in intense laser pulses

Oppermann, Florian
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The ionization and dissociation dynamics of HeH^+ in intense 800 nm laser pulses are studied using TDSE calculations. For intensities where ionization cannot be neglected, frustrated tunnel ionization (FTI), i.e. fragments in highly excited Rydberg states, plays an important role.

In the kinetic-energy-release spectrum for dissociation, two prominent regions can be identified: (i) low energy dissociation into the ground state and (ii) FTI. The latter is very similar in energy to the kinetic-energy-release spectrum for fragmentation into two ions.

The results are compared to experimental data. We also investigate the behaviour of different isotopologues

Laser-induced grating spectroscopy with absolute time information

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The nonlinear response function of transparent dielectrics is studied in time and spectral domain. A laser-induced grating is generated in a thin sample with short laser pulses (~5 fs, 700 nm) and imaged with a 2f-2f imaging system. The individual beams (the two fundamental beams and two orders of self-diffraction) are blocked consecutively in the imaging system while varying the pulse delay. From the absolute phase of the interference gratings, the delay between the fundamental pulses and the nonlinear optical response can be determined with subcycle precision. However, the optical path difference in the optical system is usually not known and inhibits the measurement of the absolute grating phase.

In a first step, the information contained in relative phase measurements (intensity dependence, material dependence) is discussed. In a second step, a method is discussed for retrieving the absolute grating phase by stepwise translation and rotation of each element of the optical system.

Free-electron quantum optics

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Katharina E. Priebe, Christopher Rathje, Armin Feist, Sergey V. Yalunin, Sascha Schäfer, and Claus Ropers

Besides being a powerful tool for time-resolved measurements of nanoscale dynamics, ultrafast transmission electron microscopy (UTEM) serves as an ideal test bench for quantum optical experiments studying the interaction with free-electron beams. Specifically, inelastic scattering between the electrons and strong optical near-fields [1] allows for a coherent manipulation of the electron quantum state [2]. The optical near-field imprints a sinusoidal phase modulation on the electron wavefunction, which is manifest in a comb of sidebands in the electron kinetic energy distribution. In this contribution, we will demonstrate how multiple near-fields can be employed to coherently control the free-electron momentum superposition states [3,4]. Furthermore, dispersive propagation translates the phase modulation into a density modulation: the electron wavefunction is self-compressed into a train of attosecond bursts. This temporal structuring of free-electron beams may find applications in electron microscopy with attosecond resolution.

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Stimulated x-ray Raman Scattering in molecular targets – a critical study of the building block of coherent nonlinear x-ray spectroscopy

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In the soft x-ray range, coherent amplification of spontaneous x-ray emission [1] and stimulated resonant inelastic x-ray scattering (SRIXS) [2] have been demonstrated in Neon, with amplification levels of up to eight orders of magnitude. A next crucial step for advancing time resolved spectroscopy is the transfer of stimulated emission and SRIXS to the hard x-ray domain that would allow single-shot spectroscopy in chemically and biologically relevant samples in the liquid phase. Here, we present results of amplified spontaneous K-α emission in Manganese salts in aqueous solutions [3] with highly focused XFEL beams. Coherent amplification of the Mn K-α emission by four orders of magnitude and saturation of the signal has been demonstrated in MnCl2 one-molar solution. More excitingly, the chemical shifts of MnCl2 and KMnO4 aqueous solution is maintained in the strongly spectrally sharpened stimulated K-α emission spectra and coherent amplification has been shown at lower concentration. A more comprehensive technique for the study of chemical structure is SRIXS. Although demonstrated in Neon [2], the realisation of SRIXS in molecular targets is more difficult to achieve, even in the soft x-ray range [4-6] due to a smaller stimulated gain-cross section, that is distributed over many electronic, vibrational and rotational channels. We present two experimental studies, based on different two-colour XFEL schemes, that were specifically developed to achieve vibrationally resolved SRIXS in CO. In a first attempt [4], the XFEL was operated in a two-colour self-amplified spontaneous emission (SASE) scheme, with one frequency band tuned to the Oxygen π* resonance, and the other band overlapping with the Stokes-shifted

emission frequencies. According to our theory [6], this setting should result in high-resolution SRIXS spectra by covariance analysis. Experimentally, the spectra are, however, contaminated by strong absorption features of molecular ions generated in competing processes. In a second experiment [6], the SASE pump pulse was replaced with a self-seeded, narrow-band pulse at considerably lower pulse energy that resulted in a significant decrease of the background. A comparison with theory shows that the experimental conditions were at the onset of an observable SRIXS signal, but so far, no statistical evidence is seen, to confidently report the demonstration of SXRIS in a molecular target. We however developed an experimental protocol that allows for the detection of relatively small SRIXS signals with highly fluctuating XFEL spectra. The challenges and the necessary experimental parameters to ultimately reach the conditions for stimulated x-ray emission spectroscopy in chemically relevant targets will be critically assessed.

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Recovery of dynamic interference

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Dynamic interference has been predicted to occur for the photo-effect in intense high-frequency laser pulses. Thereby, non-linear behavior does not emerge as higher-order processes (a.k.a. above-threshold ionization), but rather dynamical Stark shifts that induce a double-slit-like interference in time. In contrast to published claims, dynamic interference does not occur for ground state hydrogen. We show, however, that by using hydrogen in an excited state one indeed will be able to observe it. Although the process is of non-perturbative nature, we could fully disentangle system and pulse properties allowing for predictive statements about the visibility of the effect without performing any cumbersome numerical propagation.

Real-time imaging of nuclear motion and tunneling in ammonia molecules

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Exact Single-Electron Description of Many-Electron Dynamics in Strong Laser Fields

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We present an exact single-electron picture that describes correlated many-electron dynamics, with application to the electron dynamics in strong laser fields. Our approach is based on the factorization of the electronic wavefunction as a product of a marginal and a conditional amplitude. The marginal amplitude is a single-electron wavefunction that obeys a time-dependent Schrödinger equation and yields

the one-electron density and current density. From this formulation, it is straightforward to obtain single-active electron (SAE) approximations that are applicable to atoms as well as to molecules.

By studying model systems with two and three electrons, we show that the effective ionization barrier may strongly depend on the instantaneous field strength. In this situations, the usual SAE approximation that uses a static potential to represent the electron-electron interaction needs to be corrected to model the dynamics of the system.

Molecules in Tailored Fields

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The interplay of tailored fields with molecular structure will be investigated in in harmonic and photo-emission spectra of simple few-atomic molecules
such as CO, CO_2, H_2O, and methane. The potential for short-time dynamical imaging will be discussed.

Laser-induced alignment of molecules in helium nanodroplets

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Ultrafast electron dynamics in biomolecules initiated by attosecond pulses

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Purely electron dynamics in large molecules are expected to be at the origin of many biological phenomena. Attosecond pulses provide a unique tool to initiate and observe ultrafast charge migration in complex molecular systems, that is expected to occur on a temporal scale between hundreds of attoseconds up to few femtoseconds, preceding any nuclear rearrangement [1].
Here we present the first experimental demonstration of charge migration in biological relevant molecules. The molecules were photoionized by sub-300-as isolated attosecond pulses and probed by 4-fs VIS/NIR probe pulses. The molecular fragments were collected in a mass spectrometer, and the time-dependent mass spectrum was investigated as a function of the pump-probe delay.
The signal coming from doubly charged fragments [2] presents an ultrafast oscillatory dynamics with a time periodicity that is shorter than the vibrational response of the molecule [3].
Theoretical calculations were performed by using a standard time-dependent density matrix formalism, in order to describe the hole dynamics induced by an attosecond pulse similar to that used in the experiment. The results of the simulations clearly show the production of an ultrafast electron dynamics, characterized by oscillation frequencies in good agreement with the experimental results.

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Plasmon assisted high harmonic generation from silicon

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Plasmonic antennas can enhance the intensity of a nano-Joule laser pulse by localizing the electric field in

their proximity. It has been proposed that the field can become strong enough to convert the fundamental laser frequency into high order harmonics through an extremely nonlinear interaction with gas atoms that occupy the nanoscopic volume surrounding the antennas. However, the small number of gas atoms that can occupy this volume limit the generation of high harmonics. We use monopole nano-antennas to demonstrate plasmon-assisted high harmonic generation directly from the crystalline silicon substrate that hosts an array of antennas. The high density of the substrate compared to the gas allows macroscopic buildup of harmonic emission. Despite the sparse coverage of antennas on the surface, harmonic emission is 10 times brighter than without antennas. Imaging the high harmonic radiation will allow nanometer and attosecond measurement of the plasmonic field8 thereby enabling more sensitive plasmon sensors while opening a new path to extreme ultra-violet frequency combs.

Control of photoelectron momentum distributions by multi-photon ionization with polarization-shaped laser pulses

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We present results of our current experiments on multi-photo ionization of atoms with polarization-shaped femtosecond laser pulses. In the first part, we demonstrate the generation of vortex-shaped photoelectron wave packets from resonance enhanced multi photon ionization (REMPI) of potassium atoms with sequences of two time-delayed, counterrotating circularly polarized femtosecond laser pulses. In the second part, bichromatic polarization-shaped femtosecond laser pulses are used to control photoelectron momentum distributions. These bichromatic light fields, consisting of spectral bands with different ellipticity, are produced by ultrafast polarization shaping [1]. For example, we study REMPI by counterrotating circularly polarized and orthogonally linearly polarized bichromatic femtosecond laser pulses. The tomographically reconstructed three-dimensional photoelectron momentum distributions show different uncommon angular superposition states at different photoelectron energies. The analysis of the photoionization pathways reveals that REMPI by bichromatic polarization-shaped fields relies on intrapulse frequency mixing of spectral bands with different ellipticity.

Attosecond time-resolved spectroscopy of liquid water

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