4.10 Project part P10

Interaction of ultrashort electromagnetic pulses with matter – Theory

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Allocation to technical disciplines (according to code of OeStat*)

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4.10.1 Summary

This project part is concerned with theoretical aspects of the interactions of strong ultrashort pulses with matter. Its goal is to explore fundamental concepts in a new domain that is becoming accessible through experimental advances in the development of ultrashort pulses. Ultrashort pulses refers to both the number of cycles with single and even half-cycle pulses available as well as to absolute pulse duration reaching attoseconds for XUV pulses. The ongoing theoretical work is performed in close collaboration with experimental activities within the SFB016. During the second period, our research was focused on: (A) protocols to generate HCP’s in the sub-fs regime and the interaction of single and trains of unipolar HCP-cycle pulses with atoms, (B) response of matter to ultrashort pulses described by time-dependent density functional theory (TDDFT) with applications to electron emission from surfaces and two-electron quantum dots, (C) time-resolved interferences and Fano resonances in inner-shell processes generated by attosecond XUV pulses.

Highlights in (A) were the development of protocols for steering, shaping and manipulating of wavepackets in atoms by a sequence of half-cycle pulses. Wavepackets can be localized at specific positions and subsequently transported to another preselected location. First experimental tests in Rydberg atoms have confirmed this scenario. Moreover, we developed a first protocol for attosecond-HCP generation, with the goal to extend the applicability of these tools to the electronic ground state.

One highlight in (B) was the calculation of carrier-envelope phase (CEP) sensitive photoemission from a metal surface employing TDDFT in the multi-photon regime. The predicted CEP phase dependence has been meanwhile experimentally verified, however with a much reduced contrast. We have, furthermore, developed a novel read-out functional that allows the direct determination of scattering matrix elements from the time-evolved electron density and have successfully tested it for a harmonically confined two-electron quantum dot.

In (C) we have presented first attosecond-streaking images of time-resolved Fano resonances identifying signatures of both lifetimes and asymmetry parameters of the resonances. We have shown the feasibility of observing coherent dynamics and interferences on an attosecond time scale in inner-shell process, e.g. for super-Coster Kronig transitions and have identified observables not easily accessible in time-integral measurements.

Research of this project during the second funding period resulted in a total of 39 papers, among them 4 Phys. Rev. Lett. Published, in press or submitted.
4.10.2 Results and discussion

(A) Interaction of single and trains of half-cycle and few-cycle pulses with atoms

The response of atoms to ultrashort electromagnetic pulses is the subject of this project part. In the present context the term “ultrashort” takes on different meanings, yet involves the same underlying physics. On an absolute time scale, ultrashort refers to sub-fs (or attosecond) pulse duration. This time scale matches the timescale of the ground-state electronic motion in the atom. The classical orbital period in hydrogen, is

\[ \tau_0 = \frac{2\pi n^3}{\hbar} \text{ (a.u.)} \approx 100 \text{ as.} \]

However, for Rydberg states with \( n \approx 1 \), pulses in the ps to ns regime can likewise explore the physics of ultrashort, impulsive dynamics. Ultrashort refers also to the number of cycles subtended by the pulse. While an optical 5 fs pulse still contains 2 full cycles, it is now possible in the THz and GHz regime to form (sub) single cycle pulses, in particular effective half-cycle pulses (HCP). Clearly, freely propagating pulses require a vanishing time integral over the field amplitude which come about in HCP’s by long-time tails of opposite polarity.

During the second period we have demonstrated that sequences of unipolar HCP’s (u-HCP’s) allow the coherent control, steering and shaping of electronic wavepackets inside atoms. A highly elongated quasi-one dimensional Rydberg state (n=350) can be first transformed into a highly localized wavepacket near the outer turning point, subsequently “kicked” up in energy to broad coherent superposition with \( n \) up to \( n \approx 800 \), and subsequently reshaped to a narrow energy band near \( n \approx 600 \), while maintaining its strongly polarized quasi-1D character. This protocol has proven to be highly efficient (\( \eta \geq 0.5 \)) and robust against noise (Zhao, 2005). Moreover, a sequence of chirped HCP’s allows to literally push and pull on demand a strongly localized electronic wavepacket around in coordinate space inside an atom (Yoshida, 2005). Numerous other HCP driven excitation processes have been investigated (Dunning, 2005). The problem of collisional decoherence was addressed by developing the quantum trajectory Monte-Carlo method (Seligier, 2005, Reinhold, 2005) which is a generalization of the wavefunction Monte Carlo method (Dalibard, 1992) developed in the context of quantum optics.

The protocols of wavepacket control and manipulation makes use of the classical-quantum correspondence in Rydberg states and the coexistence of regular and chaotic phase space dynamics. For example, the narrowing of the energy distribution proceeds via contraction on the stable manifold near unstable hyperbolic fixed points. We therefore developed a semiclassical description of phase space propagation (Yoshida et al, 2004) employing the Herman-Kluck propagator (Herman and Kluck, 1984).
In order to develop tools for shaping and manipulation of ground-state atoms, molecules or solids, we have explored avenues to create u-HCP’s in the attosecond domain. Similar to the production of attosecond bursts (Paul, 2001) we utilize high harmonics to produce HCP’s by superimposing all harmonics, starting from the fundamental one, with phases properly aligned. For u-HCP’s, both odd and even harmonics must be present. We therefore propose to use a two-colour driving field including the second harmonic of the fundamental field to produce either a single u-HCP’s or trains of u-HCP (Persson, 2004). In order to probe the robustness of the pulse under propagation effects, the solution of the Maxwell equations are coupled to the solution of the time-dependent Schrödinger equation. Our first results achieved in collaboration with P11 indicate that it is possible to find optimal conditions for the driving field leading to the production of fairly well-defined u-HCP’s (Persson, 2005). The current upper limit of the number of aligned harmonics is given by the frequency corresponding to a resonant transition to the first excited state in the atom. For example, we find a width of the u-HCP of the order of 500 attoseconds when propagating the two-colour field through hydrogen gas. For the intensity of $I = 10^{14}$ W/cm$^2$ of the driving field and fundamental wavelength 1064 nm, trains of u-HCP’s with an intensity in the order of $I = 6 \times 10^{12}$W/cm$^2$ result.

The propagation code for the time-dependent Schrödinger equation based on the pseudo-spectral discretization method of Tong et al (1997), originally developed with the aim to describe strongly perturbed high Rydberg states, has also been used to study the two-dimensional momentum-differential ionization spectrum of hydrogen by a few-cycle strong-field ($\approx 10^{14}$W/cm$^2$) optical pulse. We have found a remarkable interference pattern (Fig. 1) which closely resembles generalised Ramsauer-Townsend (GRT) interference oscillations in electron-atom scattering (Arbo, 2005). The interference pattern calculated for hydrogen also resemble the reaction microscope image observed by Rudenko (2005) for rare gases. As GRT diffraction oscillations have a semiclassical origin (Egelhoff, 1993, Burgdörfer, 1995) it is not surprising that some of the structures also appear in quasi-classical trajectory Monte
Carlo (CTMC-T) calculations (Dimitriou, 2004). Specifically, the projected momentum perpendicular distribution \( P(p_\perp) \) features a Coulomb-like cusp while the longitudinal distribution \( P(p_\parallel) \), averaged over the rapid oscillation, displays a double peak structure near \( p_\parallel = 0 \). Within an SFA calculation (Faisal and Schlegel, 2005) the latter result from the projection onto \( p_\parallel \) of the first ATI peaks. Only the first-order peak appears to have a classical analogue while signatures of higher-order peaks are absent. Further details of this classical-quantum correspondence remain to be explored.

(B) Time-dependent density functional theory for ultrashort pulses

Calculating the many-electron response to strong ultrashort pulses has remained a challenge. One possible avenue for few-particle dynamics is the multi-configuration time-dependent Hartree-Fock approximation (see P11), another is TDDFT. The latter promises, in principle, a highly efficient approach for large systems (Runge and Gross, 1984). However, several major obstacles have to be overcome before TDDFT can provide reliable predictions for observables in ultrashort pulse-matter interaction. One well-known problem is associated with the lack of reliable exchange-correlation functionals which will be, in general, non-local in space and time. A less appreciated but for laser-matter interaction equally pressing problem is that of unknown read-out functionals. Even if the exact propagated density \( n(\hat{r},t) \) were known, only for few observables such as the time-dependent induced dipole moment the mapping is straight-forward. On the most fundamental level the response of an atom (or molecule or solid) is described by the \( S \) matrix for pulse-induced state-to-state transitions. We have recently developed a read-out functional that allows the direct determination of transition probabilities, \( |S_{ee}|^2 \) from the evolved density \( n(\hat{r},t \to \infty) \). First tests for a harmonically bound two-electron quantum dot for which a comparison with exact transition probabilities can be performed, turned out to be promising (Rohringer, 2005).

Application of TDDFT within the ALDA framework to the interaction of a few-cycle pulse interacting with a metal surface (modelled by a jellium surface) yielded the surprising result (Fig. 2) that the electron emission probability is sensitive to the carrier-envelope phase (CEP) of the pulse, remarkably more so in the multi-photon regime than in the tunnelling regime (Lemell, 2003). We have addressed the physical origin of the CEP dependence within a semiclassical model (Dombi, 2004). We follow the motion of the classical electron subsequent to a vertical transition to the continuum representing multi-photoionization with the minimum number (\( \approx 3 \)) of photons for crossing the ionization threshold assumed to take place at the time of the envelope maximum. For a CEP \( \phi = -\pi/4 \), the electron is most effectively driven away from the surface in the combined fields of the surface potential and
the laser. For phases $\phi; \pi/2$, the electron is likely to be driven into the surface, in qualitative agreement with what is observed. The first experimental tests (Apolonski, 2004) confirm this effect, yet with a considerably lower level of phase contrast than predicted. Before this process can be implemented within a compact solid-state detector, more theoretical and experimental studies are required to identify the origin of this difference. Part of the discrepancy may be due to the roughness and absorbate contamination of the surface. On the theoretical side, the representation of a gold surface by jellium, the use of the local density approximation (LDA), but also the lack of a self-consistent coupling between the Kohn-Sham equations and the Maxwell equations may be at fault. Some of these questions will be addressed in the third period.

![Computed charge emitted from a gold surface exposed to a Gaussian laser pulse](image)

**Fig. 2:** Computed charge emitted from a gold surface exposed to a Gaussian laser pulse ($\lambda_0 = 750 \text{ nm}$, $I_0 = 5 \times 10^{12} \text{ W/cm}^2$) with the electric field oriented along the surface normal as a function of the CE phase $\phi$ and the pulse duration $\tau_s$. (see Dombi, 2004)

(C) **Time-resolved Fano resonances in inner-shell processes generated by XUV attosecond pulses**

The availability of short XUV pulses with duration in the attosecond regime ($\tau_{XUV} \approx 200 – 500 \text{ as}$) opens up the possibility for time-resolved observation of the electronic motion in atoms. Using the XUV pulse as pump and the optical field as probe, attosecond streaking of electronic dynamics can be realised (Drescher, 2002). The goal of this project was to investigate streaking for the motion of a non-stationary coherently excited wavepacket.

A prototype case for the coherent dynamics in a nonstationary system is the excitation of a Fano resonance. Fano line shapes are an ubiquitous feature of resonance scattering when the continuum can be accessed both directly and by way of a quasibound state embedded in the continuum (e.g. autoionization). We have presented first theoretical results for attosecond streaking (Wickenhauser, 2004, 2005a) of a Fano resonance when using an attosecond
pump. The transition frequency from the ground state to a Fano resonance must be in the XUV range and the lifetime of the resonance should be comparable to the period of the optical cycle in order to make use of the sub-fs time resolution afforded by the streaking. Prime candidates are super-Coster Kronig processes where the inner-shell hole and the electrons filling this hole and being ionized are all residing in the same principal shell, e.g. in the lanthanides. Our treatment involves the strong-field approximation (SFA) in the optical streaking field and a first-order approximation in the XUV pump pulse. While the

![Fig. 3](image)

**Fig. 3:** Time-dependent ionization probability $P(E_i,t)$ as a function of energy and time. Inset 1: spectral distribution at short times $t \leq \tau_r$. Inset 2: spectral distribution in the scattering time limit $t \to \infty$ for $q = 1$. (See Wickenhauser, 2005a)

modification of the ionization continuum by the moderately strong optical field ($I_L = 10^{12}$ W/m$^2$) is fully taken into account, its influence on the quasi-bound state embedded in the continuum is neglected. The approximate validity of this assumption was recently demonstrated by Zhao and Lin, (2005). The streaking image (Fig. 4) of a time resolved window resonance with Fano parameter $q=0$ (Fig. 3) reflects the “hole burning” into the spectrum during the characteristic lifetime $\tau_r$ of the resonance due to destructive interference (Wickenhauser, 2005a, 2005b). Moreover, transient “beats” can be observed. Remarkably, we have found similar interference structures in time-resolved quantum transport through a network of quantum wires allowing for Fano resonances (Bärnthaler, 2005).

The significance of time-resolved studies hinges on its potential to extract information not easily accessible spectroscopically or, more generally, by time-integral measurements. We have identified two novel elements that reach beyond time-integral measurements. The phase angle of a complex Fano parameter $q$ for Fano resonances in open quantum systems (Clerk, 2001) and the relative phase of the excitation amplitude of nearby or overlapping Fano resonances (Wickenhauser, 2005b).
Fig. 4: $P_{\Delta}(E)$ as a function of the time delay between pump and probe pulse and observed in the direction of the polarization of the probe laser pulse. (a) Direct ionization with the resonant excitation suppressed ($q = 0$), (b)-(c) autoionizing resonance with both pathways open ($q = 2.2$) for different $\tau_r$. (See Wickenhauser, 2005a)

The latter represents the key to unravelling the complete density matrix for a coherent excitation of an ensemble of overlapping Fano resonances and is expected to be of importance when analyzing experimental streaking images since nearby and overlapping resonances are ubiquitous.

In parallel with atomic systems we have also studied Fano resonances in “artificial atoms”, i.e. open quantum dots. The latter can be realized on a mesoscopic scale in semiconductor heterostructures (see P4) but also in microwave cavities on a macroscopic scale. This follows from the one-to-one mapping between the Schrödinger equation and the scalar Helmholtz equation (Stöckmann, 1999). Several basic properties of Fano resonances can be varied in a controlled manner. The resonance parameters $\tau_r$ and $q$ can be varied by tuning the shutter opening. Moreover, the effect of decoherence and dissipation, in this case caused by the finite finesse of the cavity, can be investigated. Remarkable agreement between theory and experiment has been found (Rotter, 2004).

References

Wickenhauser, M., J. Burgdörfer, F. Krausz, and M. Drescher J. Mod. Optics (accepted) (2005b)
4.10.3 Collaboration within and beyond the SFB

**Collaboration within the SFB during the second period**
- calculation of carrier-envelope phase sensitive photoemission from surfaces, collaboration with Krausz and Lezius, *P2* (4 joint papers, two PRL)
- calculation of adiabatic potential hypersurfaces and conical intersections near insulators surfaces in the presence of strong Coulomb fields, collaboration with Lischka, *P9* (two joint papers)
- calculation of propagation effects for attosecond pulse generation, in particular HCP generation and high-harmonics amplification, collaboration with Scrinzi, *P12* (two joint papers)
- calculation of time-resolved inner-shell Fano resonances, collaboration with Drescher and Krausz, *P17* (3 joint papers, one PRL)

**Collaboration beyond the SFB**
- microwave radiation transport through microcavities, collaboration with Stöckmann, Marburg
- Rydberg atom response to half-cycle pulses (Dunning, Rice University, and Reinhold, Oak Ridge National Laboratory)
- CEP dependent photoemission, collaboration with Hänsch, MPQ Garching
- semiclassical theory for HCP-atom interaction, collaboration with Grossmann, TU Dresden
- theory of non-adiabatic transitions through hidden crossings, collaboration with Krstic, Oak Ridge National Laboratory
- interaction of ultrashort laser pulses with large rare-gas clusters, collaboration with Vernhet, University of Paris VI
- calculation of adiabatic potential hypersurfaces and conical intersections near insulators surfaces in the presence of strong Coulomb fields, collaboration with L. Wirtz, IEMN, Lille, France
- TDDFT, Surface response to strong fields, collaboration with K. Tökesi, ATOMKI, Debrecen, Hungary
- Interaction of atoms with time-dependent electrical fields, collaboration with X. M: Tong, Tsukuba University, Japan