Ultrashort laser oscillator pulses meet nano-structures: from attosecond physics at metal tips to dielectric laser accelerators

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Abstract. We report on two experiments about the interaction of femtosecond laser pulses with nano-scale structures. In the first experiment, we have observed strong-field effects at nanometer sharp metal tips, such as rescattering, the high-energy cut-off and a strong carrier-envelope phase dependence. All these effects are hallmarks of attosecond physics. We have used the new understanding of these processes at a solid tip to build a near-field sensor with record sensitivity of 1 nm. In the second experiment, we have demonstrated the acceleration of non-relativistic electrons with the optical near-field at a photonic grating structure, with an acceleration gradient of 25 MeV/m. The grating serves to generate a near-field mode co-propagating with and efficiently imparting momentum onto the electron, which is impossible in vacuum over a macroscopic distance. For relativistic electrons we expect an acceleration gradient of more than 1 GeV/m with two-sided grating structures.

1. Introduction
Nanooptic or near-field optics opens possibilities that are impossible with common (far-field) optics. In the near-field modes with significant population can exist that are exponentially damped out in the far-field. We take advantage of nanooptics in two different settings. In the first part we will discuss our recent observations of strong-field effects driven by the near-field of and at a nano-scale metal tip. In the second experiment we will show how evanescent (near-field) modes can be utilized to accelerate electrons with the electro-magnetic light-field of femtosecond laser pulses. Whereas in the former case field enhancement at the tip allows us to work with low-power oscillator pulses and still reach the required peak intensities, so enables, in the second case, the grating the generation of the accelerating mode. This mode is co-propagating with the electrons and allows continuous and phase-synchronous momentum transfer from the laser beam to the electron beam.

2. Attosecond physics at metal nano-tips: the driven electron as near-field sensor
The recollision scenario lies at the core of strong-field and attosecond physics [1]. It describes that an electron, liberated by a strong laser pulse from an atom or molecule in the gas phase, can be driven back to the parent ion or molecule inside a single cycle of the laser field once the
laser field has flipped sign. Several different processes can then happen: 1. The electron can recombine, upon which the energy that the electron has gained in the laser field is radiated away in the form of a high-harmonic photon. It is this process that coined the name of the field. 2. The electron can scatter elastically at the parent matter. In that case, the momentum vector just flips around, upon which the electron can gain more energy in the laser field. 3. Other processes, including inelastic scattering can take place.

In this contribution we focus on the second process. We observe and measure the electron, here emitted and re-colliding with a metal nano-tip. The metal tip here replaces the gas phase particle known from high harmonic generation physics.

Our basic observations have been described elsewhere [2, 3, 4, 5, 6], which is why we only briefly discuss the main features here. We have observed the tell-tale plateau in the electron spectra, which gives evidence of the elastic recollision [7]. Because the plateau indicates a process that is driven by the electric field of the laser pulse – as opposed to the pulse envelope – we have investigated the field dependence of the process. We have done this be recording spectra in a carrier-envelope phase (CEP) resolved way. These spectra reveal photon orders for certain settings of the CEP, whereas the photon orders are absent for other settings.

With the tree-step model (also known as simple man’s model) [8] we are able to explain the spectra both qualitatively and quantitatively. The key point is that the photon picture is replaced by the notion of electronic matter wave interference in time and energy. Interference fringes in the energy domain show up if two emission windows in time exist during which the electron can be emitted. On the way to the detector, the two electronic matter wave packets spread out and overlap. This leads to interference fringes, which we detect with a retarding field spectrometer. In essence, in a CEP resolved measurement the photon picture can not explain the observed effects any longer: For certain settings of the CEP the emission process lacks any periodicity if only a single emission window exists. The intuitive process description therefore requires the matter wave picture and interference effects instead of photon orders. For CEP settings for which interference fringes exist, their spacing matches the photon picture because according to Fourier, results in a fringe spacing of \( \Delta E = \frac{h}{T} \approx 1.5 \text{ eV} \).

The experimental data are well-supported by theory models [5, 6, 9, 10]. With the understanding of the dynamics, we can turn the perspective around and employ the re-colliding electron as a probe for the local electro-magnetic field in front of the tip [11]. This is based on the “10-\( U_p \) law” [12], which links the maximum classically allowable kinetic energy with the local intensity via

\[
T_{\text{cutoff}} = 10.007U_p + 0.538\Phi.
\]

Here, \( U_p = e^2\lambda^2E^2/(16\pi^2mc^2) \) is the ponderomotive potential of the electron in the oscillating electric field with field strength \( E \). \( e \) and \( m \) are the electron’s charge and mass, and \( c \) is the speed of light. \( \Phi \) is the workfunction of the tip material.

We measure \( T_{\text{cutoff}} \) and hence \( U_p \) and \( E \). The field enhancement factor is the ratio between the measured peak field and the peak field in the bare laser focus, without the tip present. The results are shown for tungsten and gold tips as a function of tip radius in Fig. 1.

Also shown are FDTD simulation results for tungsten, gold and silver. For all materials, the field enhancement factor rises with decreasing tip radius, as can be expected from geometrical arguments. Interestingly, all materials exhibit a rather similar field enhancement factor, even though gold is considered to be a plasmonic material while tungsten is not. We note that we do observe a plasmon resonance in theory plots, however not for the dielectric function of gold for a wavelength of \( \sim 800 \text{ nm} \). Hence also for gold, the field enhancement effect is mainly geometric and thus broadband in nature. While these data show the efficacy of the method, more research is needed to figure out as to why larger field enhancement factors for gold are reported in the
Figure 1. a) Field enhancement taking place at a nanoscale metal tip when laser pulses are focused at the tip. In the figure, the light is propagating in the y-direction with the polarization in the x-direction. The white line shows the decay of the field away from the tip on its symmetry axis. b) Conceptual picture of the electron dynamics in front of the tip. We utilize the electron wave packet to sample the field. The extension of the electron trajectory is less than 1 nm, hence the field can be approximated as constant. c) Results of the measurement of the electric field: the field enhancement factor is the ratio between the field obtained with the electron rescattering method and the field of the bare focus, without the tip present. Points are measurement data for tungsten (blue balls) and gold (red squares). The lines are theory curves obtained from extensive FDTD simulations. The solid blue line is for tungsten, the red dashed curve is for gold, and the dash-dotted is for silver.

We note that the spatial integration of 1 nm is almost one order of magnitude smaller than in the previously published record resolution work on for near-field sensing [14].

3. Dielectric laser acceleration with the inverse Smith-Purcell effect: efficient momentum transfer from a light field to electrons with the help of a transparent grating structure

In free space, momentum transfer from an oscillating field to a charged particle is not possible over extended time and length scales. This does not hold any more close to a boundary. With suitable grating structures, propagating electrons can be efficiently accelerated. This process becomes the more efficient, the closer the electrons’ velocity approaches the speed of light.

Figure 2 shows the concept of the acceleration: The grating generates an evanescent mode that co-propagates with the electron and thus transfers momentum efficiently. Grating period, laser wavelength and electron velocity have to be matched, yielding the synchronicity condition

\[ \lambda_p = m\beta\lambda. \]

Here, \( \lambda_p \) is the grating period, \( m = 1, 2, 3 \ldots \) and \( \lambda \) the laser wavelength. \( m \) represents the order of the spatial harmonics. In Figure 2, \( m = 3 \).

We send a beam of electrons, generated in and controlled with the help of a scanning electron microscope column, along a transparent grating structure. At the grating structure, the electron beam is overlapped with a laser beam. The laser beam is derived from a long-cavity Titanium:sapphire oscillator, with the following parameters: pulse duration of 110 fs, center
Figure 2. a) A plane wave propagates from top to bottom through a transparent grating structure (light blue). Electrons, indicated by the black dots, propagate parallel to the horizontal grating surface. If the grating period is matched to the speed of the electrons and the driving wavelengths, a mode co-propagating with the electrons at the surface at the grating is excited. It can efficiently transfer momentum to the electrons. In the figure, only electron 1 is accelerated. Electron 2 is decelerated, while electron 3 is deflected towards the grating and electron 4 away from the grating. b) The acceleration gradient $G_{acc}$ for a given laser peak field $E_p$ as function of the electron velocity.

wavelength of 787 nm, repetition rate of 2.745 MHz, average power of $\sim 350 \text{ mW}$. The focal parameters are: spot radius of 8.3 $\mu\text{m}$, peak electric field of $2.5 \ldots 3 \text{ GV/m}$. The grating is written into fused silica. The electron beam passes by with a distance down to $\sim 50 \text{ nm}$. Because of these small dimensions, we put the grating on top of a mesa structure such that the electron beam is only close to the glass where necessary.

We measure a maximum acceleration gradient of $\sim 25 \text{ MeV/m}$. The polarization dependence, the dependence on incoming electron beam energy (synchronicity condition) and the dependence on the distance of the electron beam from the grating surface all agree with what is expected from this near-field based scheme of electron acceleration. We are currently in the process of writing up the results in greater detail and expect them to be submitted soon.

In summary, near-field based experiments with femtosecond oscillator pulses at nanoscale tips and at transparent grating structures have allowed the observation of novel effects, such as strong-field physics at solids and electron acceleration right with the electro-magnetic field of laser pulses. The latter work represents the first observation of the inverse Smith-Purcell effect in the optical regime.

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