Ultrafast monoenergetic electron source by optical waveform control of surface plasmons

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Abstract: We propose coherent control of photoelectron acceleration at metal surfaces mediated by surface plasmon polaritons. A high degree of spectral and spatial control of the emission process can be exercised by amplitude and phase controlling the optical waveform (including the carrier-envelope phase) of the plasmon generating few-cycle laser pulse. Numerical results show that the emitted electron beam is highly directional and monoenergetic suggesting applications in contemporary ultrafast methods where ultrashort, well-behaved electron pulses are required.

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References and links

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1. Introduction

Optical waveform control of recollision processes of atomic electrons has brought a deeper insight into atomic physics since the reproducible generation of isolated attosecond light pulses in atomic gas targets was enabled by carrier-envelope (CE) phase control of few-cycle light pulses (see [1] and references therein). Similarly, in solids, first pioneering coherent control experiments in nanoplasmonic systems underpin the possibility of controlling electronic motion on the nanoscale with visible laser pulses [2]. However, most methods of femtochemistry and attophysics are still waiting to be applied to nanoscale objects. As first steps in this direction, promising studies pave the way toward nanometer and attosecond resolution surface charaterization schemes [3] and existing ultrafast electron diffraction and microscopy setups already offer a form of ultrahigh 4D resolution, too [4,5]. Therefore, studying the properties of ultrafast photoelectron sources optimized for the above applications has become of central interest in many research groups [6-8]. Prompted by these developments, we propose a method where spatial and spectral emission properties of a surface plasmon-based ultrafast photoelectron source can be directly controlled by the waveform of a few-cycle laser pulse. We also provide new insight into the spatiotemporal dynamics of surface plasmon (SP) enhanced electron acceleration [8]. Similarly to a field emission tip [6,7], a plasmonic photoelectron source can also play a key role in the further development of the above-mentioned novel methods. Moreover, studying the properties of such an electron beam has the potential to reveal ultrafast excitation dynamics in solids.

SP enhanced electron acceleration is a recently discovered acceleration method in the evanescent field of surface electromagnetic waves (SPs) [9,10]. More recent, conspicuous results include electron acceleration in SP fields up to keV energies (without external dc fields) with simple Ti:sapphire laser oscillators [8] and predictions about the carrier-envelope phase dependence of this phenomenon [11]. The process is particularly interesting for the development of ultrafast methods where a well-controlled electron source is needed with high repetition rate. We carried out numerical investigations of the relevant properties of such a source when the interacting laser pulse is composed of only few optical cycles, in addition, we consider photoelectron emission in the tunnelling regime, which is, as opposed to other simulations in this field, a much more realistic approach considering inherent field enhancement effects appearing upon SP coupling. We propose the usage of this electron source for the generation of well-behaved particle beams by spectral and spatial filtering of photoaccelerated electrons, easily realizable on a flat metal surface or on a surface-integrated nanostructure. By spatially resolved examination of the emission properties on the nanoscale we come to valuable conclusions about the parameter regime ideally suited for ultrafast, high spatial resolution applications.

2. Simulation methods

We modelled surface plasmon enhanced electron acceleration in a computationally more efficient way than previous approaches by using a semiclassical model analogous to the 3-step model of high harmonic generation [12]. SP enhanced electron acceleration involves distinct physical processes such as the couping of the free-space and plasmonic electromagnetic fields, photoelectron emission from the metal layer, and the subsequent acceleration of free electrons by the decaying plasmon field on the vacuum side of the surface. Therefore the steps of our model correspond to these processes. First we gained analytic formulae for SP fields instead of the computationally intensive complete numerical solution of Maxwell's equations in the Kretschmann-Raether SP coupling configuration [8]. Based on the well-known fact that SP fields decay exponentially away from the surface [13], we used

$$\mathrm{E}_y^{SP}(x,y,\mathrm{t}) = \eta E_0 \,\mathrm{E}_{env}(x,t)\cos\left(k_{SP}x - \omega_0 t + \varphi_0\right)\exp(-\alpha y) \tag{1a}$$

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$$\mathrm{E}_x^{SP}(x,y,\mathrm{t}) = \eta a E_0\, \mathrm{E}_{env}(x,t)\cos\left(k_{SP}x - \omega_0 t - \frac{\pi}{2} + \varphi_0\right)\exp(-\alpha y) \qquad (1b)$$

for the field components on the vacuum side of the metal layer, where E_0 is the field amplitude, $\mathrm{E}_{env}(x,t)$ is an envelope function determined by the temporal and spatial beam profiles of the incoming Gaussian pulse, η is the inherent field enhancement factor upon plasmon coupling [13], k_{SP} is the plasmon wave vector, ω_0 is the carrier frequency, φ_0 is the carrier-envelope phase of the laser pulse and α is the decay length of the plasmonic field in vacuum. For the accurate determination of the field we used the evanescent decay parameter $\alpha = 247\ \mathrm{nm}^{-1}$ from previous, non-approximative studies carried out for the same input parameters for laser pulses having a central wavelength of 800 nm [14]. We used the value of $a = 0.3$ according to the notion that the amplitudes of the *x*- and *y*-components of the plasmonic field have this ratio according to the numerical solution of Maxwell's equations [14]. This way both the distribution of the field amplitude in the vicinity of the surface in Fig. 1 (false colour representation) and the vector representation of the field (not shown here) demonstrate very good agreement with rigorous calculations (see Fig. 3 in Ref. [14]).

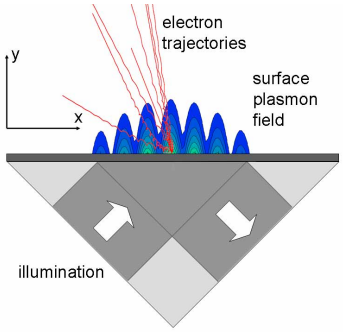


Fig. 1. The generation of electron beams by SP enhanced electron acceleration. Field amplitudes (false colour plot) and electron trajectories (red lines) illustrate the model used. For further details see text.

As a second step, we placed a point array along the prism surface and examined the spatial and temporal distribution of the instantaneous tunneling current (induced by the plasmon field) by applying the Fowler-Nordheim equation, routinely used in studies involving electron emission from metal nanotips [6,7]. Tunneling emission is a more realistic starting point for such a simulation than previously used multiphoton assumptions [8, 11]. This is supported by experimental evidence demonstrating efficient electron acceleration for SP fields of 1.8×10^9 V/cm (Keldysh-gamma of 0.01) [8]. Physically, tunneling emission sets in in this scheme even for lower driving intensities because of the inherent field enhancement phenomenon of surface plasmonic fields (with an *E*-field enhancement factor of ×3-4 for flat and up to ×100 for rough surfaces [13]). This way we end up with a spatially and temporally resolved map of tunneling probabilities as determined by the SP field. We have to mention here that our model slightly overestimates the beneficial effect of tunneling emission by assuming that the quoted field enhancement appears uniformly over the whole illuminated surface. However, experimental observation of highly efficient electron acceleration up to almost keV energies with low-energy (1.5 nJ) laser oscillator pulses [8] suggests that field enhancement (and the resulting tunneling emission) is a key element in this process and can not be neglected if we want to investigate the most interesting properties of the electron beam (such as the maximum energies, monoenergeticity etc.). Further refinement of the model from this point-of-view can take place after systematic comparison with targeted experiments.

Thirdly, we scrutinized vacuum electron trajectories in the plasmon field for each point in the above-mentioned array and for several emission instants. Some representatively chosen trajectories originating in the central point can be seen in Fig. 1 (red curves). In some cases

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the electron trajectories involved a recollision with the metal surface and when this happened, no electron emission was assumed. In all other cases the final kinetic energies and directions of the photoemitted and accelerated electrons were placed in a matrix for each emission point in space. This way we ended up with novel acceleration maps carrying both nanometer-scale spatial and high-resolution spectral information of the process. With these steps both microscopic and (by integrating the emission map along the spatial coordinate) macroscopic electron spectra and emission distributions can be generated. We checked that macroscopic spectra generated this way reproduce former measurement and simulation results (published in Refs. [8, 11]) extremely well, includig basic scaling laws, such as the linear scaling of the highest electron energy with the field intensity [14]. Thereby the applicability of our analytic field expression (1a-b) was confirmed supporting the robustness of our simplified model.

3. Monoenergetic, highly directional electron beams with few-cycle pulses

We present several emission maps using realistic parameters to reveal the fine structure of the acceleration process and to arrive to conclusions about macroscopically observable properties of the electron beams generated. We examined the final kinetic energy distribution of plasmon-accelerated electrons along the plasmon propagation direction (*x*-axis, representing emission locations along the surface) for a few-cycle interacting pulse with a Gaussian pulse shape, 15 fs and 5 fs intensity full width at half maximum (FWHM), $\varphi_0 = 0$ carrier-envelope phase (with envelope and field maxima coinciding) and 800 nm central wavelength. The pulse was assumed to be focussed to a spot with a diameter of 4 μm on the prism surface so that a peak plasmon field strength of 5.8×10^8 V/cm (Keldysh-gamma of 0.31) was reached. With this effective intensity value we have already taken into account that substantial field enhancement factors (up to ×100) can be achieved with respect to the plasmon generating field.

With these simulation parameters, we calculated the spatial and spectral distribution of the emitted electrons along the plasmon propagation direction [in false colour representation in Figs. 2(a), 2(d)] for two different pulse lengths to illustrate few-cycle effects. Whereas in the multi-cycle regime (15 fs pulse length) in Fig. 2(a) a much more structured distribution can be observed, in Fig. 2(d) (5 fs pulse length) the emission is mostly concentrated to a single structure on the emission map providing a better behaved electron beam. We can also see that the emission of high-energy electrons is localized to the centre of the illuminated spot and that the number of distinct structures on the emission maps roughly correspond to the number of optical cycles in the generating pulse. This is because the "birth" interval of those electrons in the continuum that can leave the vicinity of the surface is limited to about 1/4 of every laser cycle, which is due to the breaking of the symmetry by the surface such that positive and negative half-cycles are not identical from this point-of-view. During every laser cycle there is one such favored interval and electrons emitted in each of these intervals spend different amounts of time in the field, hence they undergo different accelertion.

There is an even more conspicuous property that seems crucially important form the point-of-view of the applications of this electron source. Figs. 2(b) and 2(e) depict the angular-kinetic energy distributions of the emitted electron beams, showing in which direction the energetic electrons leave the surface. We can see that the emission is confined to a small range of angles supporting a directionally emitted electron beam ideally suited for novel ultrafast techniques. Provided that the pulse length is in the few-cycle range [Fig. 2(e)], the angular emission map is reduced to a single distinct structure corresponding to a highly directional, quasi-monoenergetic electron beam representing the most favourable regime of SP enhanced electron acceleration. By integrating any of the distributions along the *x*-axis we end up with the macroscopically observable electron spectra depicted in Figs. 2(c) and 2(f). The spectrum in Fig. 2(f) has a FWHM $\Delta E_{kin}/E_{kin}$ value of 0.22 (with E_{kin} denoting the electron kinetic energy) corresponding to a quasi-monoenergetic spectrum. Under experimental circumstances the spectral properties of this electron beam can be further enhanced by applying a retarding potential to suppress the low-energy wing of the spectrum.

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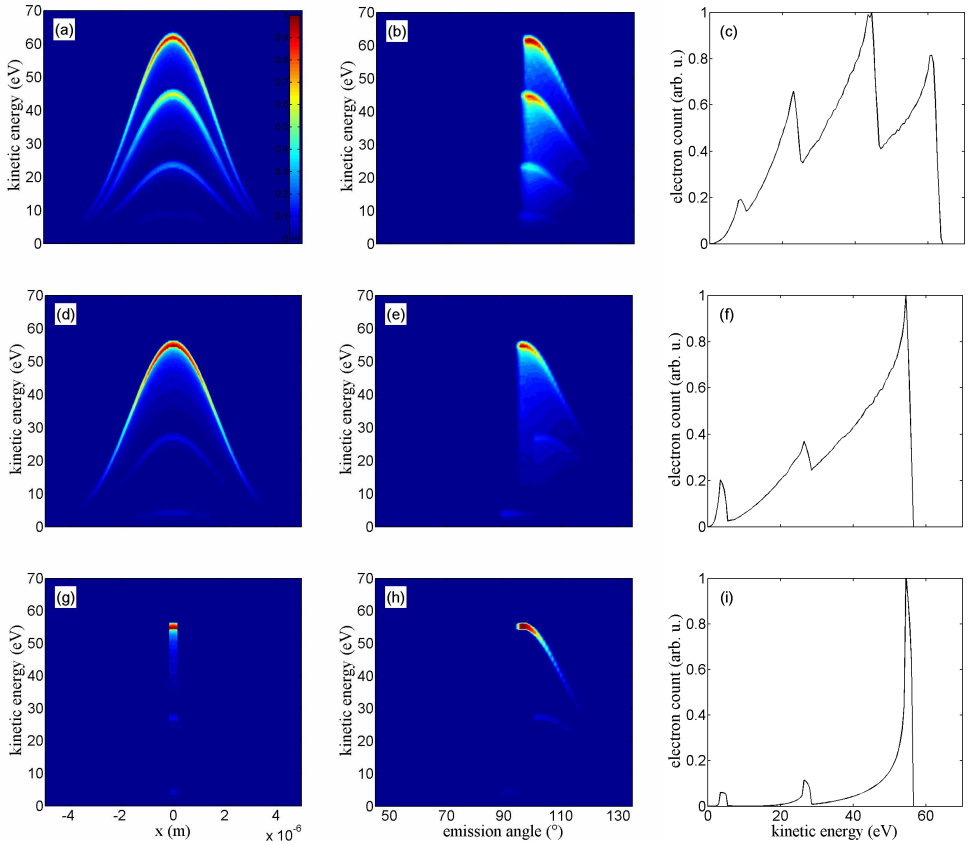


Fig. 2. Normalized photoacceleration maps (kinetic energy distribution of electrons emitted at different points of the surface: (a), (d) and (g), in false colour representation), angular and kinetic energy distribution ((b), (e) and (h)) and macroscopic electron spectra ((c), (f) and (i)) of surface plasmon accelerated electrons for three example parameter sets. (a)-(c) are for 15 fs, (d)-(i) are for 5 fs laser pulses. In (g)-(i) we considered emission only from a 300 nm wide strip, as illustrated in (g) corresponding to a nanolocalized emission region. For further parameters, see text.

The fact that the appearance of high energy electrons is confined to the central portion of the surface, prompted us to investigate what happens to the angular distribution if one restricts the emission on the prism surface to a limited, nanoscopic range. This is illustrated in Figs. 2(g) – 2(i) where the same emission maps and spectra are given as in Figs. 2(d) – 2(f) with the only difference that electrons coming only from a 300 nm wide central strip of the surface were considered. It is possible to realize such a situation with experimental nanofabrication tools, for example, by depositing a dielectric layer on top of the metal-coated prism, which has an opening with such a width to let electrons escape. By confining the emission area this way, it can be seen that the distribution in Fig. 2(h) shows a highly enhanced contrast. This means that even more monoenergetic spectra and even more directional beams can be generated from this spatially confined source. This way the $\Delta E_{kin}/E_{kin}$ value of the integrated spectrum can be enhanced by almost an order of magnitude to 0.033, as depicted in Fig. 2(i). Thereby our results suggest that SP electron acceleration offers a robust and powerful technique for the generation of ultrafast, monoenergetic, highly directional electron beams.

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4. Carrier-envelope phase effects

It is known that the carrier-envelope phase of few-cycle pulses has a measurable effect on laser-solid interaction processes even in the perturbative regime of nonlinear optics [15-18]. Motivated by these findings we also examined the effect of the optical waveform on the electron beam. The angle-energy distributions in Figs. 3(a) and 3(b) (CE phases of $\varphi_0 = \pi/2$ and $\varphi_0 = \pi$, respectively) can be directly compared to that of Fig. 2(e) (CE phase $\varphi_0 = 0$); the only difference in the input parameters is that we varied the CE phase of the interacting pulses but otherwise left other parameters unchanged.

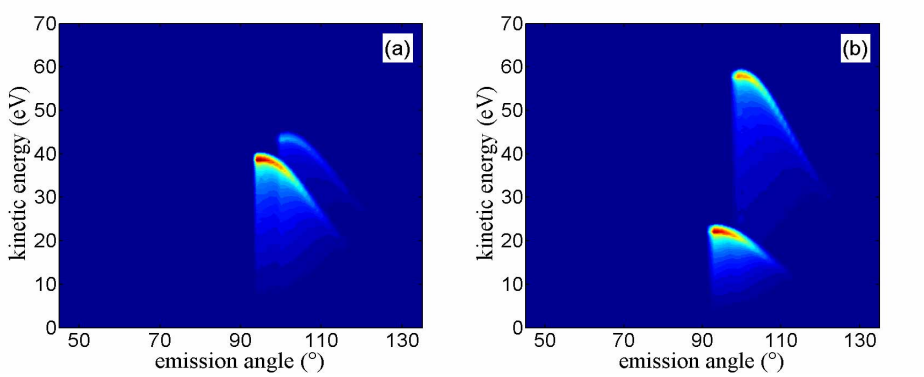


Fig. 3. Normalized photoacceleration maps (the kinetic energy distribution of electrons emitted at different angles from the surface) in false colour representation for two different carrier-envelope phase values ($\varphi_0 = \pi/2$ for (a) and $\varphi_0 = \pi$ for (b)). The accompanying animation shows the same distributions for more CE phase values together with integrated electron spectra and the waveform of the interacting laser pulse (file size 1.08 MB).

We can see that the position of the spectral cut-offs determined by the acceleration process are highly dependent on the CE phase of the pulses in accordance with previous results [11]. In our case, however, by having taken tunneling emission into account (instead of multiphoton emission) the influence of the CE phase becomes more pronounced. The number of structures observable on the emission maps (and in the integrated spectra, see accompanying animation) corresponds to the number of optical cycles in the laser pulse (2 in our case). However, for CE phase values between 1.75π and $\pi/4$ these distinct structures merge serving as a basis for an ideal photoelectron source. Therefore the generation of electron beams with the desired features requires state-of-the-art laser sources with CE phase stabilization.

5. Summary

We proposed a possibility of unprecedented plasmonic control of free-space electron beams by cutting edge photonic technology in optical waveform (or, in other words, carrier-envelope phase) control of few-cycle pulses. We have also proven for the first time that the simulation of this process is possible with very simple computational tools thanks to a model analogous to the three-step model of high harmonic generation. Our spatially and spectrally resolved numerical results demonstrate that the generation of highly directional, quasi-monoenergetic electron beams (with a FWHM $\Delta E_{kin}/E_{kin}$ value exceeding 0.03) is possible in nanoscopic environments. These suggestions pave the way toward the further development of ultrafast and ultrahigh spatial resolution charazterization methods. This is becoming a central effort in many leading research groups in the world and it will hopefully bring about real-time observation of electron motion on the natural spatial scale of these charge transfer processes on surfaces, surface-bound chemical and biological samples and in electron diffraction and microscopy schemes.

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