LETTER

High energy photoelectron emission from gases using plasmonic enhanced near-fields

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Received 28 May 2013, in final form 22 August 2013
Accepted for publication 23 August 2013
Published 13 September 2013
Online at stacks.iop.org/LPL/10/105302

Abstract
We study theoretically photoelectron emission in noble gases using plasmonic enhanced near-fields. We demonstrate that these fields have a great potential to generate high energy electrons by direct excitation from mid-infrared laser pulses of current femtosecond oscillators. Typically, these fields appear in the surroundings of plasmonic nanostructures with various geometrical shapes, such as bow-ties, metallic waveguides, metal nanoparticles and nanotips, when illuminated by a short laser pulse. Here, we consider metal nanospheres, in which the spatial decay of the near-field of the isolated nanoparticle can be approximated by an exponential function according to recent attosecond streaking measurements. We establish that the strong spatial inhomogeneous character of the enhanced near-field plays an important role in the above threshold ionization (ATI) process and leads to a significant extension in the photoelectron spectra. In this work, we employ the one-dimensional time-dependent Schrödinger equation to calculate the photoelectron emission of xenon atoms in such enhanced near-fields. Our findings are supported by classical calculations.

(Some figures may appear in colour only in the online journal)
In a few-cycle pulse the laser electric field can be fully characterized by both its duration and the so-called carrier-envelope phase (CEP). In contrast to multicycle pulses, the electric field of a few-cycle pulse, and consequently the laser–matter processes driven by it, are greatly affected by the CEP [6, 7]. The importance of CEP has been experimentally observed in high-harmonic generation (HHG) [8], the direct emission of electrons from atoms [9], and in the nonsequential double ionization yield [10]. In order to have a better control of the system in a sub-femtosecond temporal scale, it is important to find reliable schemes to measure and precisely characterize the absolute phase of few-cycle pulses.

The sensitivity of the energy and angle-resolved photoelectron spectra to the absolute value of the CEP has sparked the investigation of ATI generated by few-cycle driving laser pulses. Consequently, this feature makes the ATI phenomenon a very reliable and robust tool in laser pulse characterization. In order to extract the CEP of a few-cycle laser pulse, the backward–forward asymmetry of the ATI spectrum has to be measured, and from the information collected the absolute CEP can be obtained [11]. It is also established that the absolute CEP can be measured more accurately if the ATI photoelectron kinetic energy spectrum has a higher cutoff.

New experiments in noble gases have demonstrated that the harmonic cutoff could be extended by using plasmonic field enhancement [12, 13]. This field appears when a nanostructure is illuminated by a short laser pulse. As a result, the external low intensity femtosecond pulse couples with the plasmon mode, inducing a collective oscillation of free charges within the localized regions of the nanostructure and forming a spot with a highly enhanced electric field. Due to the strong confinement of the plasmonic spots and the distortion of the electric field by surface plasmons, the locally enhanced field is not spatially homogeneous and strongly influences the subsequent motion of the electron in the continuum. Consequently, important changes will occur to the main features of the strong field phenomena [12, 13], since now the force applied to the active electron depends also on position. Strong field phenomena in such a kind of field are a topic of intense activity nowadays [14–22].

Recently, in the context of strong fields, an alternative process has been employed to produce photoelectron emission using metallic solid state nanostructure targets instead of atoms and molecules in the gas phase. This phenomenon, which is known as above threshold photoemission (ATP), has received special attention due to its novelty and the new physics involved. Due to the plasmonic field enhancement, the electrons emitted by ATP from metallic surfaces or metal nanoparticles present distinct characteristics, namely higher kinetic energies in a non-ponderomotive regime [32]. In comparison to noble gases interacting with the same strong laser field, here the photoelectron kinetic energy spectrum has a much higher cutoff (see e.g. [23–36]).

In this letter we put forward the plausibility to perform ATI experiments by combining plasmonic enhanced near-fields and noble gases. The proposed experiment would take advantage of the plasmonic enhanced near-fields, which present a strong spatial inhomogeneous character and the flexibility to use any atom or molecule in the gas phase. From the theoretical viewpoint the ATI process can be tackled using different approaches (for a summary see e.g. [37–42] and references therein). We employed the numerical solution of the one-dimensional time-dependent Schrödinger equation (TDSE) by including the actual functional form of metal nanoparticles plasmonic near-fields obtained from attosecond streaking measurements [43, 44]. We have chosen this particular nanostructure since its actual enhanced field is known experimentally, while for other nanostructures, such as bow-ties [12], the real plasmonic field is unknown. For most of the plasmonic nanostructures the enhanced field is theoretically calculated using a finite element simulation which is based on an ideal system, not the complex situation we are dealing with. For instance, [12] states an intensity enhancement of four orders of magnitude (calculated theoretically), but the maximum harmonic order measured was 17, which corresponds to an intensity enhancement of two orders of magnitude rather than four (for more details see [17, 45]). However, our numerical tool is developed in such a way as to allow the treatment of a very general set of spatially inhomogeneous fields, such as those present in the vicinity of metal nanostructures [12], dielectric nanoparticles [7, 13], or metal nanotips [32]. The kinetic energy for the electrons, both direct and rescattered are classically calculated and compared to the predictions of our quantum mechanical approach.

The one-dimensional time-dependent Schrödinger equation (1D-TDSE) for a model atom reads:

\[
\frac{i}{\hbar} \frac{\partial \Psi(x, t)}{\partial t} = \mathcal{H}(t) \Psi(x, t)
\]

\[
= \left[ -\frac{1}{2} \frac{\partial^2}{\partial x^2} + V_a(x) + V_l(x, t) \right] \Psi(x, t)
\]

where \( V_l(x, t) \) is the laser–atom interaction. For the atomic \( V_a(x) \) potential, we use the quasi-Coulomb or soft core potential \( V_a(x) = -\frac{1}{\sqrt{x^2+a^2}} \), which was firstly introduced in [46] and has been widely used in the study of laser–matter processes in atoms. The parameter \( a \) allows us to tune the ionization potential of the atom under consideration once the 1D-TDSE is discretized in space. In our studies we consider the field to be linearly polarized along the \( x \)-axis and modify the interaction term \( V_l(x, t) \) in order to treat spatially inhomogeneous near-fields while still maintaining the dipole character. Consequently we write

\[
V_l(x, t) = -E(x, t) x
\]

where \( E(x, t) \) is the laser electric field characterized by

\[
E(x, t) = E_0(t) \exp(-x^2/\chi^2).
\]

In equation (3), we have collected all the temporal information in \( E_0(t) \), i.e. \( E_0(t) = E_p f(t) \sin(\omega t + \phi) \). Here, \( E_p, f(t), \omega \) and \( \phi \) are the peak amplitude of the laser electric field, the pulse envelope, the frequency of the laser pulse and the CEP, respectively. The exponential part of equation (3) corresponds to the spatial dependency of the plasmonic enhanced field, where \( \chi \) is a parameter that characterizes the spatial decay of

\[
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\]

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the plasmonic near-field and depends on both the geometrical and material characteristics [13, 17] (note the units of $\chi$ are in length). Equation (3) is only valid for $x \geq R_0$, where $R_0$ is the radius of the metal nanoparticle, i.e. for values of $x$ that are outside the metal nanoparticle surface. As a first approximation to the problem we put our model atom at $x = 0$. We could justify this choice by considering the very rapid fall in the cutoff energy as the atom is displaced towards larger $x$ values, and consequently the high energy tail of the ATI spectra will be governed by the atoms located at the origin. In addition, the electron motion is in the region $x \geq R_0$, with $(x + R_0) \gg 0$. To model short laser pulses, we use a $\sin^2$ envelope $f(t)$ of the form $f(t) = \sin^2 \left( \frac{\omega t}{2n_p} \right)$, where $n_p$ is the total number of optical cycles. The time duration of the laser pulse will be $T_p = n_p \tau$, where $\tau = 2\pi/\omega$ denotes the laser period. We assume that the atomic target is in the ground state (1s) before we turn on the laser ($t = -\infty$). This particular state can be found by solving an eigenvector and eigenvalue problem once the 1D-TDSE has been discretized. For Xe atoms we chose $a = 1.62$ to match its atomic ionization potential $I_p = 12.199$ eV (0.446 a.u.).

Equation (1) was solved numerically using the Crank–Nicolson scheme with an adequate spatial grid [47]. In our numerical experiment we employed boundary reflection mask functions [48] in order to avoid spurious contributions, and the grid size has been adequately extended in order to treat high energy electrons. It is well known that these electrons can travel far away from their parent nucleus without returning to the core; however, this effect was considered in our numerical scheme. To calculate the energy-resolved photoelectron spectra $P(E)$ we used the window function technique developed by Schafer [49, 50]. This tool represents an improvement with respect to the usual projection methods and has been widely used to calculate angle-resolved and energy-resolved photoelectron spectra [51].

In figure 1, we present a schematic picture of the systems under study, in which a metal nanoparticle such as those used in the experiment of [43] is surrounded by Xe atoms. We also included the time and spatial profile of the laser electric field, where it is possible to observe the usual time dependence and the strong spatial variation at the nanometer scale. In this nanoparticle the spatial variation of the electric field produces strong modifications in the electron motion, and consequently in the associated laser–matter processes, for example above threshold ionization (ATI). Furthermore, each metal nanoparticle acts as a laser nanosource that contributes to the photoelectron emission process.

In our model we have employed the laser field enhanced parameters used in the experiment of [13] ($I = 2 \times 10^{13}$ W cm$^{-2}$ and $\lambda = 720$ nm) to make our proposed experiment perfectly feasible. For this kind of metallic nanoparticle, the enhanced laser field intensity corresponds to initial laser input intensities of $10^{12}$ W cm$^{-2}$, which through the plasmon enhanced amplification process is intensified by more than one order of magnitude (see [36] and references therein). The Gaussian shaped experimental laser pulses with 5 fs FWHM are modeled by employing a $\sin^2$ shaped pulse with five optical cycles (13 fs).

As a first test we maintain the aforementioned laser field enhanced parameters and we varied the $\chi$ parameter in equation (3). In figure 2, we present the photoelectron spectra calculated using 1D-TDSE for Xe atoms. Each curve presents different values of $\chi$: homogeneous case ($\chi \to \infty$), $\chi = 40, 30$ and 29. We have chosen arbitrarily the values of $\chi$ in order to show how the ATI spectra are...
rescatter. Here, \( \text{U} \) electrons that, once ionized, return to the core and elastically well known ATI cutoff at 10\,\text{U} intensity the cutoff at 2\,\text{U} one shown in [13]. On the other hand, for this particular \( \chi \) parameter chosen. As depicted in figure 2, the cutoff is far beyond the classical limit 10\,\text{U} direct ionized electrons is not visible in the spectra. For the homogeneous case there is a visible cutoff at \( \approx 10.5 \,\text{eV} \). This value is in perfect agreement with the one shown in [13]. On the other hand, for this particular intensity the cutoff at 2\,\text{U} (\approx 2.1 \,\text{eV}) developed by the direct ionized electrons is not visible in the spectra. For the inhomogeneous cases the cutoff of the rescattered electron is far beyond the classical limit 10\,\text{U}, depending on the \( \chi \) parameter chosen. As depicted in figure 2, the cutoff is extended as we decrease the value of \( \chi \). For \( \chi = 40 \) the cutoff is at around 14 \,\text{eV}, while for \( \chi = 29 \) it is around 30 \,\text{eV}. The low energy region of the photoelectron spectra is sensitive to the atomic potential of the target and one needs to calculate TDSE in full dimensionality in order to model this region adequately. In this letter we are interested in the high energy region of the photoelectron spectra, which is very convenient because it is not greatly affected by the considered atom. Thus, by employing 1D-TDSE, the conclusions that can be taken from these highly energetic electrons are very reliable.

In order to investigate fully the behavior of the system, i.e. the metallic nanoparticles surrounded with noble gas atoms, we considered a higher laser field intensity of \( I = 5 \times 10^{13} \,\text{W cm}^{-2} \) while keeping the rest of the laser parameters the same. In figure 3, we have plotted the photoelectron spectra for the homogeneous case and for \( \chi = 29 \). From this plot we observe that the inhomogeneous character of the laser enhanced electric field introduces a highly nonlinear behavior in this particular laser–matter phenomenon. For this intensity with \( \chi = 29 \) it is possible to obtain very energetic electrons, reaching values of several hundreds of \,\text{eV}. This is a good indication that the nonlinear behavior of the combined system of the metallic nanoparticles and noble gas atoms could pave the way towards generating \,\text{keV} electrons with tabletop laser sources.

We now concentrate our efforts in order to understand the extension of the energy-resolved photoelectron spectra using classical arguments. From the simple-man model [52] we can describe the physical origin of the ATI process as follows: an electron bound to its parent ion at a position \( x = 0 \) is liberated to the continuum at a given time, called the ionization or birth time \( t_i \), but with zero velocity, i.e. \( \dot{x}(t_i) = 0 \). In the strong field picture this electron now experiences only the influence of the oscillating laser electric field (the residual Coulomb interaction is neglected) and will reach the detector either directly or through the so-called rescattering process. By using the Newton equations it is possible to calculate the maximum kinetic energy of the electron for both the direct and rescattered processes. The classical equation of motion for an electron in the laser field can be written as \( \ddot{x}(t) = -\nabla_x V_l(x, t) \). More specifically, for the electric field of the form (3) it reads \( \ddot{x}(t) = -E(x, t) (1 - \dot{x}(t)/\chi) \). Note that in the limit \( \chi \rightarrow \infty \), we recover the conventional homogeneous case \( \ddot{x}(t) = -E(t) \).

For the direct ionization the electron reaches the detector with a kinetic energy given by

\[
E_d = \frac{\left[\dot{x}(t_i) - \dot{x}(t_i)\right]^2}{2},
\]

where \( t_i \) is the end time of the laser pulse. For the rescattered ionization, in which the electron returns to the core at a time \( t_i \) and reverses its direction, the kinetic energy of the electron yields

\[
E_r = \frac{\left[\dot{x}(t_i) + \dot{x}(t_i) - 2\dot{x}(t_i)\right]^2}{2}.
\]

For homogeneous fields equations (4) and (5) result in

\[
E_d = \frac{\left[A(t) - A(t)\right]^2}{2} \quad \text{and} \quad E_r = \frac{\left[A(t) + A(t) - 2A(t)\right]^2}{2},
\]

with \( A(t) \) being the laser vector potential \( A(t) = -\int E(t') \,dt' \). Furthermore, it

**Figure 2.** Energy-resolved photoelectron spectra for Xe atoms driven by an electric enhanced near-field. The laser intensity after interacting with the metal nanoparticles is \( I = 2 \times 10^{13} \,\text{W cm}^{-2} \). We employ \( \phi = \pi/2 \) (cos-like pulses) and the laser wavelength and number of cycles remain unchanged with respect to the input pulse, i.e. \( \lambda = 720 \,\text{nm} \) and \( n_p = 5 \) (13 fs in total). The arrow indicates the conventional 10\,\text{U} cutoff (10.5 \,\text{eV} for this case).

**Figure 3.** Idem figure 2, but now the output laser intensity is \( I = 5 \times 10^{13} \,\text{W cm}^{-2} \). We include arrows for the two classical limits, 2\,\text{U} (5.24 \,\text{eV}) and 10\,\text{U} (26.2 \,\text{eV}), respectively.
is well known that the maximum values for $E_d$ and $E_r$ are $2U_p$ and $10U_p$, respectively [37]. Although for laser electric fields of the form given by equation (3), the aforementioned limits should be revised.

In figure 4 we present the numerical solutions of the Newton equation for an electron moving in a linearly polarized spatially inhomogeneous electric field of the form (3), in terms of the kinetic energy of the direct and rescattered electrons. We employed the same laser enhanced field parameters as in figures 2 and 3. Panels (a) and (c) show the kinetic energy of the direct (●) and rescattered (○) electrons for the homogeneous case with laser enhanced intensities of $I = 2 \times 10^{13}$ W cm$^{-2}$ and $I = 5 \times 10^{13}$ W cm$^{-2}$, respectively. In panels (b) and (d), we plot the kinetic energy of the direct (●) and rescattered (○) electrons for the extreme inhomogeneous case ($\chi = 29$) and for the mentioned intensities. Furthermore, we observed an unconventional behavior due to the inhomogeneous character of the enhanced near-fields. From the panels (b) and (d), we clearly see that spatially inhomogeneous enhanced fields produce electrons with high kinetic energy, well beyond the usual classical limits of $2U_p$ and $10U_p$ for the direct and rescattered process respectively. These new features are related to the changes in the electron trajectories (for details see e.g. [15–17]). Here, the electron trajectories are substantially modified in such a way that the released electron has a longer time to spend in the continuum, acquiring more energy from the laser electric field. For the homogeneous case, on the other hand, classical simulations show both the $2U_p$ (direct electrons) and $10U_p$ (rescattered electrons) kinetic energy limits and the values are in perfect agreement with the 1D-TDSE simulations. A similar behavior was observed in metal nanotips [32], although in our case both direct and rescattered electrons are considered.

In this letter we propose the generation of high energy photoelectrons using near-enhanced fields by combining metallic nanoparticles and noble gas atoms. Near-enhanced fields present a strong spatial dependence at the nanometer scale; a behavior that introduces substantial changes in the laser–matter processes. We have modified the time-dependent Schrödinger equation to model the ATI phenomenon in noble gases driven by the enhanced near-fields of such a nanostructure. We predict a substantial extension in the cutoff position of the energy-resolved photoelectron spectra, far beyond the conventional $10U_p$ classical limit. These new features are reproduced well by classical simulations. Our predictions would pave the way towards the production of high energy photoelectrons, reaching the keV regime by using a combination of metal nanoparticles and noble gases. In this kind of system, each metal nanoparticle configures a laser nanosource with particular characteristics that allow not only the amplification of the input laser field, but also the modification of the laser–matter phenomena due to the strong spatial dependence of the generated coherent electromagnetic radiation.

Acknowledgments

We acknowledge the financial support of the MICINN projects (FIS2008-00784 TOQATA, FIS2008-06368-C02-01 and FIS2010-12834); ERC Advanced Grant QUAGATUA, the Alexander von Humboldt Foundation and the Hamburg Theory Prize (ML). This research has been partially supported by Fundació Privada Cellex. JAP-H acknowledges support from Spanish MINECO through the Consolider Program SAUUL (CSD2007-00013) and research project FIS2009-09522, from Junta de Castilla y León through the Program...
for Groups of Excellence (GR27) and from the ERC Seventh Framework Programme (LASERLAB-EUROPE, Grant No. 228334). AZ acknowledges support from EPSRC Grant No. EP/J002348/1 and Royal Society International Exchange Scheme 2012 Grant No. IE120539.

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