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Single circularly polarized attosecond pulse generation by spatially inhomogeneous fields from atoms with nonvanishing angular quantum number

Fujun Chen [a,b], Jianghua Luo [c], Fengguang Luo [a,*]

[a] National Engineering Laboratory for Next Generation Internet Access System, School of Optical and Electronic Information, Huazhong University of Science and Technology, Wuhan 430074, China
[b] Department of Electronics and Science Engineering, Huanghuai University, Zhumadian 463000, China
[c] School of Physics and Optoelectronic Engineering, Yangtze University, Jingzhou 434023, China

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ABSTRACT

We address an efficient scheme to generate single circularly polarized attosecond pulse using spatially inhomogeneous fields in the vicinity of metallic nanostructures from atom media with nonvanishing angular quantum number. Based on the numerical solution of a two-dimensional time-dependent Schrödinger equation, it is shown that using linearly polarized plasmonic fields, the harmonic intensity of two orthogonal polarization components, *i.e.*, harmonics polarized parallel and perpendicular to the driving fields polarization direction, is comparable. Moreover, the relative phase between the two components is about $\pi/2$. As a result, near-circularly polarized isolated attosecond extreme ultraviolet or X-ray pulses can be directly produced.

1. Introduction

The fast development of the attosecond science in the past two decades opens the way for monitoring and controlling electron dynamics with attosecond and Ångstörm resolutions [1–3]. Nowadays, attosecond pulses based on high harmonic generation (HHG) have been successfully produced in experiment [4]. Generally, the harmonic signal appears per half cycle of the driving laser, resulting in attosecond pulse train. Technical methods such as polarization gating [5,6] and spectral filter [7] have been employed for obtaining an isolated attosecond pulse. Recently, it has been demonstrated that isolated 148 as pulses can be produced using a generalized double optical gating technique [8]. Very recently, the shortest single attosecond pulse with a duration of 67 as has been generated with few cycle intense femtosecond pulses [9].

The HHG mechanism can be well understood within the classical three-step model [10]. First, a electron tunnels into the continuum through the potential barrier formed by the Coulomb potential and the laser field. Then it is accelerated in the laser field treated as a free particle. Finally, it may recombine with the parent ion and a high energy XUV photon is emitted. The highest kinetic energy that the electron gains from the driving field is determined by $\varepsilon_{max} = 3.17U_p$ [11], where $U_p = E_0^2/4\omega^2$ is the ponderomotive potential and E_0 is the amplitude of the laser field with angular frequency ω. The highest harmonic frequency that can be gained from the HHG process is defined by the cutoff formula $N_{max}\,\omega = I_p + 3.17U_p$, where I_p is the ionization potential.

It is well known that in a elliptically polarized driving field, the generated harmonics also have ellipticity [12,13]. According to the three-step model, only those electrons which can return to the parent ion core are responsible for HHG. However, in an elliptically polarized field, the transverse component of the driving field will cause transverse displacements of the electron trajectories, preventing the electrons from returning to the ion core and thereby diminishing the generation of high harmonics. In a circularly polarized laser field, the tunneling electron would miss the core thus no harmonics could be emitted [14]. In more accurate quantum mechanical terms, the conservation of the angular momentum suppresses the generation of circularly polarized harmonics. Since circularly polarized attosecond pulses are potential new tools for investigations of electron dynamics in atoms, molecules, and materials, many efforts have been made to generate isolated circularly polarized attosecond pulses. Yuan and Bandrauk [15] have shown that a single circularly polarized attosecond pulse can be produced by an intense elliptically polarized laser pulse in an asymmetric molecular ion due to the asymmetry of the Coulomb potential. They also proposed that an intense elliptically polarized laser pulse in the presence of an intense terahertz field can be

* Corresponding author.
E-mail address: fgluo@mail.hust.edu.cn (F. Luo).

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employed to produce a single circularly polarized attosecond pulse [16]. Using bichromatic circularly polarized laser fields with opposite rotation polarization directions, a method was proposed for circularly polarized HHG [17]. Recently, near circularly polarized high harmonics and attosecond pulses have been generated using atom states with nonvanishing angular quantum number [18]. Liu and Nisoli [19] pointed out that the ellipticity, the major axis direction and the helicity of the elliptically polarized attosecond pulses can be easily controlled in an initial $2p_0$ state of He^+.

Very recently, nanoplasmonic field enhancement in the vicinity of nanoantennas [20,21], nanotips [22] and metal waveguides [23] has successfully realized the localization of femtosecond radiation on a nanometer scale and resulted in strong local electric fields. Due to surface plasmon resonance, the plasmonic field is spatially inhomogeneous [24]. Recently, HHG in a spatially inhomogeneous field has been extensively investigated for its potential in extending the cutoff of high harmonics and generating more intense attosecond pulse with shorter duration [25–41]. The experiment of Kim et al. [22] has recently under an intense discussion [42,22] lead to a hot debate that whether the high harmonic radiation is in fact coherent (HHG) or merely an incoherent atomic line emission. Theoretical works done by Ciappina et al. using the 3D finite element simulations have shown that the coherent HHG process can indeed happen in the vicinity of metallic nanostructures when are illuminated by a laser pulse.

In this communication, we employ the spatially inhomogeneous field to stimulate the atom media which are prepared in an initial state with nonvanishing angular quantum number. Based on the numerical solution of a two-dimensional time-dependent Schrödinger equation (2D-TDSE) from the model hydrogenlike system Ne prepared in an initial $2p_0$ state, we find out that at proper angle of the driving field polarization with respect to the quantization axis of the atom state, the two orthogonal harmonic components which are polarized parallel and perpendicular to the polarization direction of the driving fields have comparable intensities. Besides, It is also shown that the two harmonic components are out of phase by about $\pi/2$ over a broad spectral range. As a result, those harmonics can support near-circularly polarized attosecond pulse generation with a duration of about 50 as. Such an isolated circularly polarized laser pulse can serve as a new tool for the research of nonlinear response of matter to intense fields.

2. Theoretical model

We perform simulations of the 2D-TDSE for the HHG process of a valence $2p_0$ state of a model neon atom in a linearly polarized spatially inhomogeneous laser field. The $2p_0$ state can be prepared by exciting neon with linearly polarized π pulses, which can be designed to obtain a complete population transfer from the 1*s* to the 2*p* atomic orbital [43]. $2p_0$ state is the simplest non-spherical state, which allows the generation of elliptically polarized harmonic generation using linearly polarized driving fields with proper alignment angles. In fact, the $2p_0$ state, in particular, can serve as models for a molecular orbital with π symmetry. Atomic units (a.u.) are used throughout this paper. The one electron wavefunction in the laboratory frame, $\psi(\mathbf{r}, t) = \psi(x, z, t)$, is given by

$$i\frac{\partial}{\partial t}\psi(\mathbf{r}, t) = \left[-\frac{\nabla^2}{2} + V(r) + \mathbf{E}(z, t)\mathbf{r}\right]\psi(\mathbf{r}, t). \quad (1)$$

The atomic 2D potential, $V(r)$, is written as

$$V(r) = \frac{Z_{eff}(r)}{\sqrt{r^2 + \zeta}}, \quad (2)$$

where $r = \sqrt{x^2 + z^2}$ and the effective charge of the core is $Z_{eff}(r) = 1 + 9\exp(-r^2)$ to account for the screening of the nuclear charge by the inner electrons. The soft core parameter ζ is chosen to be 2.88 a.u. to obtain the correct energy value of the valence $2p_0$ state. The numerically calculated energy of the $2p_0$ state is $E_{2p_0} = 0.79$ a.u. which matches the negative first ionization potential of Ne. The spatially inhomogeneous driving field is defined as

$$E(z, t) = E_0 f(t)(1 + \epsilon z)\cos(\omega t + \phi), \quad (3)$$

where E_0, ω and ϕ are the amplitude, angular frequency and carrier envelop phase (CEP) of the driving field, respectively. We use a linear functional form $(1 + \epsilon z)$ to approximatively characterize the spatial inhomogeneity of the laser field, which is widely used in previous works of plasmon-assisted HHG [24–27,44,45]. The parameter ϵ determines the degree of spatial inhomogeneity in units of reciprocal length. We would like to mention that a better and more realistic way to model the spatially inhomogeneous is introduced in Ref. [28], in which they used electric fields in the vicinity of metallic nanostructures obtained from 3D finite element simulations. The envelope is chosen to be trigonometric pulse envelope, *i.e.*, $f(t) = \sin^2(\pi t/T)$. Here, $T = 3T_0$ is the pulse duration corresponding to 8 fs full-width at half-maximum (FWHM), and T_0 is one optical cycle of the laser pulse. The wavelength of the laser field is set to be 2000 nm. The reason why we use laser field with wavelength of 2000 nm is that it needs lower absorbency of the nanoantennas and requires lower electric field enhancements to observe the same harmonics for longer wavelength in comparison with 800 nm laser field [28].

The 2D-TDSE is solved using the second-order split-operator method on a Cartesian grid [46]. The spatial grid in each direction ranges from -220 a.u. to 220 a.u. with step sizes 0.1 a.u. The time step is chosen as $\Delta t = 0.01$ a.u. To avoid unphysical reflections at the grid boundaries, the electron wavefunction is applied by an absorbing potential at each time step. The initial wavefunction $\phi_{2p}(\mathbf{r})$ is obtained by imaginary-time propagation and orthogonalization under symmetry conditions. The high harmonic field of the two orthogonal components can be calculated by the Fourier transform of the dipole acceleration expectation value:

$$|A_{j=x,z}(\omega)|^2 = \left|\frac{1}{\tau}\frac{1}{\omega^2}\int - dt\ \exp(-i\omega t)\langle\psi(t)|[H(t), [H(t), j]]|\psi(t)\rangle\right|^2, \quad (4)$$

where $H(t)$ is the Hamiltonian and $\psi(x, z, t)$ is the electron wavefunction.

3. Results and discussions

Fig. 1 shows the 2D distribution of the $2p_0$ state of Ne in the atomic frame (x', z'). The polarization direction of the driving field represented by the *z*-axis forms an angle α with z'. The coordinate system (x, z) forms the laboratory frame. We first calculate the high harmonic spectra produced in the $2p_0$ state by the spatially inhomogeneous laser pulse, with polarization direction forming an angle $\alpha = 30^\circ$ with the z'-axis. We change the angle α from 0° to 90° and find that 30° satisfies the best condition for the generation of circularly polarized harmonics radiation. The peak intensity is set to be $I = 3.0 \times 10^{14}$ W/cm^2. It should be emphasized that this intensity is the laser intensity after the plasmonic enhancement, not the incident intensity. The incident laser intensity could be several orders of magnitude smaller which allows the nanoplasmonic target to withstand thermal damage in the experiment.