Extreme-ultraviolet frequency comb generation by polarization-gated surface-enhanced optical fields

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We show that two synchronized counter-propagating femtosecond pulses could be controlled by adjusting their relative delay to excite surface-enhanced optical fields of time-gated linear polarization in contrast with intrinsic elliptic polarization excited by the conventional one-pulse incidence scheme. Such surface-enhanced optical fields can be used to generate efficient high harmonics and thus extreme-ultraviolet frequency combs. The energy conversion efficiency as well as the frequency span of the generated extreme-ultraviolet frequency combs can be significantly increased with controllable emission probabilities along different directions relative to the surface. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2967732]

Extreme-ultraviolet (XUV) frequency combs can be produced through high harmonic generation (HHG) in rare gas based on enhancement cavities,[1–4] which can be used for ultrahigh-precision spectroscopic measurement and optical frequency metrology in the XUV region.[5] The near-infrared high-repetition femtosecond (fs) pulses from oscillators are coherently accumulated and amplified in external cavities to ensure efficient HHG, where the cavity length and dispersions are well controlled to match the source cavities.[1,2] A typical intensity enhancement ranging from several tens to hundreds can be also achieved for the incident optical field as it resonates with the surface plasmon (SP),[6] where the energy stored in the metal film during the lifetime of SP is hence released into a subwavelength space. It has been extensively investigated to enhance the nonlinear optical responses,[7] such as second harmonic generation, stimulated Raman scattering, and so forth. The recent study[8] showed that a peak intensity of 10^{15}–10^{16} W/cm^2 can be obtained close to the surface of the thin metal film by SP resonance. As an alternative efficient and straightforward approach,[9] such SP resonance-enhanced optical fields can be used to produce XUV frequency combs.

Due to the evanescent feature,[10] the SP resonance-enhanced optical field has two orthogonally polarized components of unequal amplitudes and a constant phase delay ($\pi/2$), which analogizes to an elliptically polarized field. As a result, the harmonic emission probability or equivalently the energy conversion efficiency of XUV frequency combs in such optical fields is relatively low, since high harmonics are emitted by the recollision process of the preliberated electron with its ionic core under the driving of external optical fields.[11] Therefore, intense linearly polarized optical field rather than elliptically polarized one is preferred[12] to produce XUV frequency combs by HHG.

In this paper, we numerically demonstrate that SP resonance-enhanced optical fields can be gated to linear polarization by using counterpropagating incident pulses of *p* polarizations, which establish counterpropagating surface-enhanced optical fields of counter-rotating elliptic polarizations and, accordingly, could be controlled with a proper relative delay. The time-gated linearly polarized optical field has a dramatic enhancement in intensity and, thus, benefits the generation of XUV frequency combs through HHG processes.

The so-called Kretschmann configuration[6] is used to enhance the intensity of the incident optical field through SP resonance. A 56-nm-thick model metal film with a plasma frequency of 5.73×10^{15} Hz and a damping rate of 1.3×10^{14} Hz is considered to be coated on a prism with a refractive index of 1.6, which launches SP resonance for incident field with the central wavelength of 800 nm at an incident angle of 45°. The incident optical field is a *p*-polarized 12 fs pulse at 800 nm. The full distribution of the enhanced optical field close to the surface is obtained by solving the Maxwell equations for the optical field evolution using the finite-difference time-domain method.[13] As shown in Fig. 1(a), the output pulse train from a fs oscillator is firstly split into two parts, which are then individually steered from different directions to obtain enhanced optical fields as they resonate with the SP. An intensity enhancement factor of 80 (or equivalent 40 relative to the sum incident intensity of the two incident pulses) can be achieved by using the polarization-gated scheme, which is almost doubled as compared with the conventional one-pulse incident case with an enhancement factor of 22 for the case considered here. Here, the relative time delay between the incident two fs pulses is adjusted to be zero. As shown in Fig. 1(b), another important feature of the polarization-gated surface-enhanced optical field is that the polarization state is gated to be linear,[14] which periodically evolves along the surface. As we will discuss in what follows, this is very important to efficiently produce XUV frequency combs through HHG processes.

The output harmonic spectrum is obtained by Fourier transforming the field induced dipole moment, which is calculated by numerically integrating the time-dependent Schrödinger equation for a single electron in an effective nuclear potential $V(x,y)=-Z/(x^2+y^2+\varepsilon^2)^{1/2}$ and a time-dependent potential of electron-laser interaction.[15] Here, Z is the nuclear charge and ε is the soft well parameter.[16] A model atom with a single-ionization potential of 12.0 eV is

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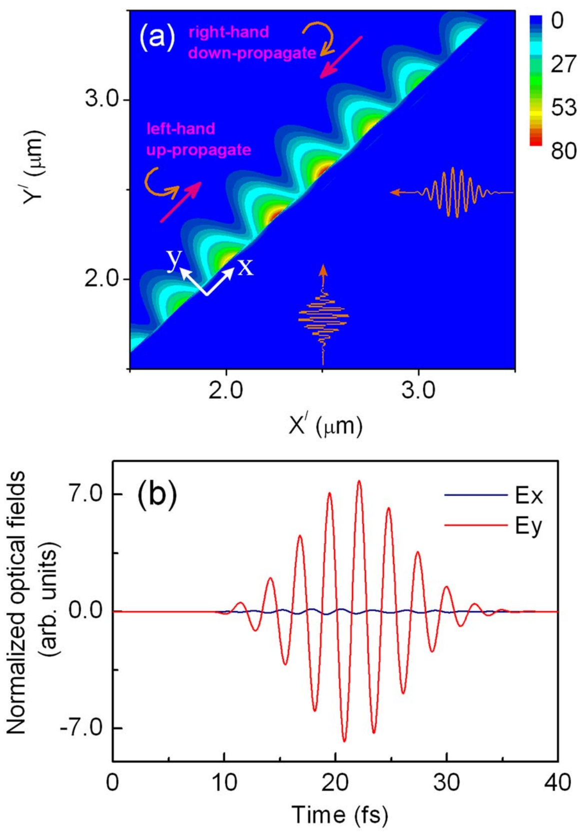

FIG. 1. (Color online) (a) The intensity distribution and (b) the normalized time-dependent field components of the polarization-gated SP resonance-enhanced optical field by two synchronized bottom-incident and right-incident fs pulses with a time delay of zero.

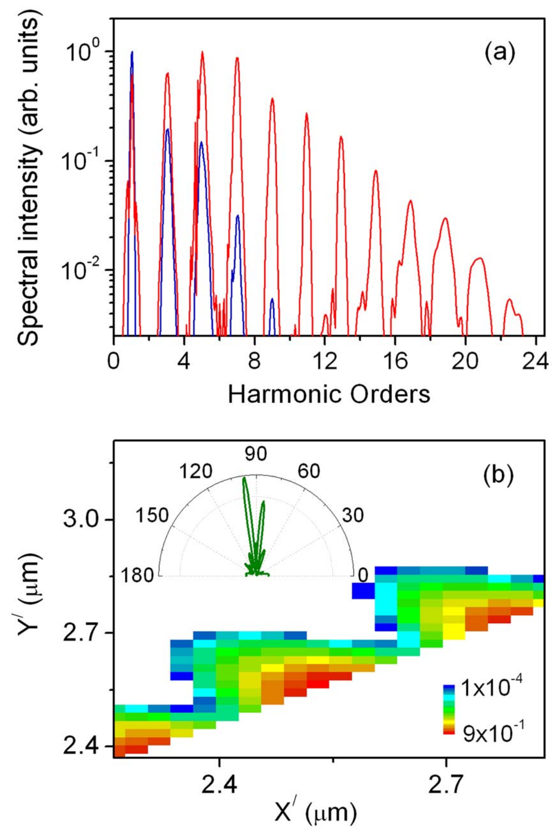

FIG. 2. (Color online) (a) The generated high harmonic spectrum as the atom is exposed to the surface-enhanced optical field. The navy and red lines stand for the results when there is only one incident pulse with an incident peak intensity of 3.0×10^{12} W/cm^2, and there are two incident pulses (zero delayed) when each one has an incident peak intensity of 1.5 $\times10^{12}$ W/cm^2, respectively. (b) The intensity distribution of the generated 11th harmonic as the atoms are exposed to different positions close to the surface by polarization-gated SP resonance scheme with a delay of zero. The corresponding emission probability of the generated 11th harmonic as a function of emission angle is plot as the inset.

considered in our simulations. As plotted with navy line in Fig. 2(a), harmonics up to the ninth order can be generated as the atom is exposed to the SP resonance-enhanced optical field close to the surface when only one fs pulse with an incident peak intensity of 3×10^{12} W/cm^2 is used. These limited harmonics are mainly due to the elliptically polarized state and the restricted intensity enhancement factor of the surface optical field. However, the red line in Fig. 2(a) clearly indicates that the harmonic spectrum, including both the spectral intensity and frequency span, is significantly enhanced with our polarization-gated scheme shown in Fig. 1(a). Harmonics up to the 23rd order are generated even the total intensity of the incident pulses is the same. Such dramatic enhancement in the output harmonic spectrum is resulted from the improved polarization state as well as the strength of the SP resonance launched optical fields by polarization-gated scheme.

The polarization-gated surface-enhanced optical field is periodically modulated along the x axis and attenuates exponentially along the y axis.[6] Consequently, as shown in Fig. 2(b), XUV frequency combs can be produced only when atoms are exposed to the intense optical fields in a subwavelength spatial region close to the surface, which is also periodically modulated as the optical field. This makes the XUV frequency combs to be emitted from a train of periodically distributed "nanoparticles," which may result in a relative low harmonic intensity due to the limited atoms in response. It is possible to observe the generated XUV frequency combs along different directions in the x-y plane, since the enhanced optical fields counterpropagate along the surface with two orthogonally polarized components. The emission directions of the generated harmonics are calculated by considering the superposition of the generated harmonics from all the atoms exposed to different positions close to the surface, where the harmonic emission probabilities along various directions for each single atom are obtained by Fourier transforming the projected dipole accelerations.[17] Not only the polarization state, but also the spatial intensity distribution of the polarization-gated SP resonance-enhanced optical field contributes to the final emissions. The inset of Fig. 2(b) shows emission probabilities along various angles with peak emission probabilities at 84° and 96° relative to the surface.

By varying the time delay between the two incident pulses, the resulted relative oscillating phase and strength between the orthogonally polarized components of the enhanced optical fields can be changed. For example, as shown in Fig. 3(a), the polarization state at the location with maximum harmonic emission probability and its evaluation is changed when the time delay is adjusted to be π/ω_0, where ω_0 stands for the carrier frequency of the incident pulses. As a result, as shown in Fig. 3(c), a noticeable emission probability perpendicular to the surface is observed. Figure 3(e) shows the harmonic spectrum with the driving field as shown in Fig. 3(a). Correspondingly, Figs. 3(b), 3(d), and 3(e) show the resulted polarization state, emission directions and har-

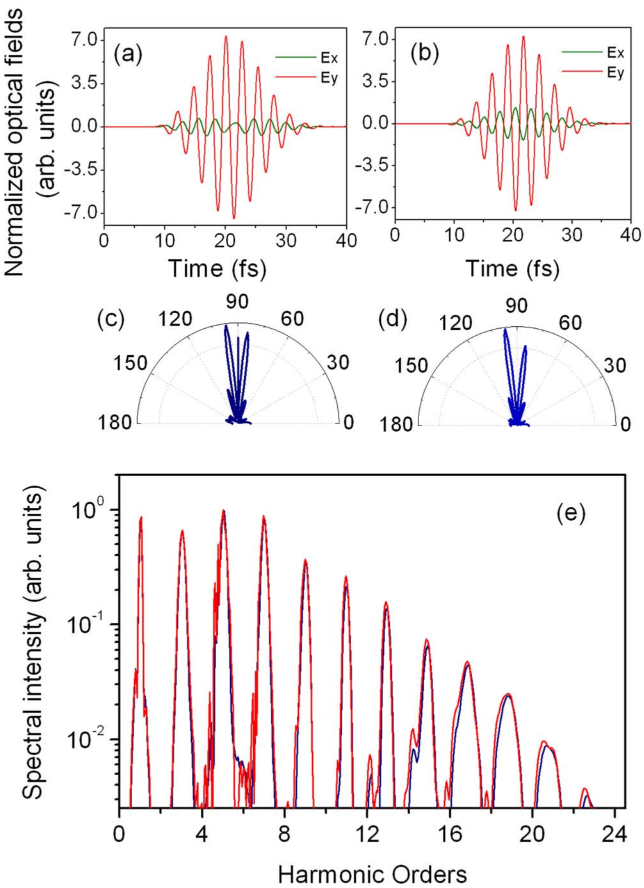

FIG. 3. (Color online) The normalized time-dependent surface-enhanced optical fields [(a) and (b)], the emission probabilities of the generated 11th harmonic as a function of emission angle [(c) and (d)], and the generated harmonic spectra (e) when the delay between two incident fs pulses is adjusted to be, respectively, π/ω_0 [(a), (c), and navy line in (e)] and $\pi/2\omega_0$ [(b), (d), and red line in (e)].

monic spectrum as the time delay is set to be $\pi/2\omega_0$. The periodically spatial evaluation of the polarization state and the standing-wave like surface-enhanced optical field by polarization-gated scheme play significant roles for the observed harmonic emission.

In summary, we numerically demonstrate that XUV frequency comb generation by surface-enhanced optical fields through HHG in atomic gas can be dramatically enhanced by using the polarization-gated scheme with properly controlled time delay, which not only doubles the effective field intensity but also leads to a time-gated linear polarization state. The polarization-gated scheme provides us an efficient way to control the polarization state of the surface-enhanced optical field. This is significant for polarization sensitive processes, such as molecular alignment,[18] electron emission,[8] and other nonlinear optical responses. By using this approach, XUV frequency combs could be generated with a fairly compact or even microstructure device with enhanced optical fields close to the surface, which could be further integrated for practical uses and lead to more opportunities in these fields.

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