Surface-enhanced high-harmonic generation: a promising approach for extreme ultraviolet frequency combs

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We propose a promising scheme to produce extreme ultraviolet frequency combs through high-harmonic generation with a surface-enhanced optical field. High harmonics are generated in a wavelength-scale spatial region and show a noticeable emission probability at an angle around 10° relative to the surface. This can be further controlled with an additional static electric field or polarization-gated scheme, leading to enhanced energy conversion efficiency and extra even-order harmonic generation. © 2008 Optical Society of America

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As a powerful tool for ultrahigh precision spectroscopy and optical frequency metrology, extreme ultraviolet (XUV) frequency combs have attracted much attention for their unique properties [1]. Pumping with near-infrared (IR) high-repetition femtosecond pulses from oscillators, XUV frequency combs could be produced through cavity-enhanced high-harmonic generation (HHG) processes in noble gases based on enhancement cavities [2,3] with well-controlled cavity lengths and dispersions. The generated XUV frequency combs could be further coupled out of the external cavities by inserting a thin Brewster plate [2,3], drilling a small hole in one of the ending concave mirrors [4], or using noncollinear cavity-enhanced HHG processes [5]. On the other hand, the incident optical field can also be enhanced typically in intensity of several tens to hundreds of times by surface plasmon (SP) resonance [6], which has been extensively used for surface-enhanced nonlinear processes and ultrahigh sensitive measurements [7–9]. In this Letter, we numerically demonstrate that the sufficiently enhanced high-repetition near-IR femotosecond pulses by SP resonance can be used to drive HHG processes in noble gases, which may act as an alternative and promising approach for XUV frequency comb generation.

By using the Kretschmann configuration, SP resonance can be readily attained close to the metal–vacuum interface at the resonance angle of p-polarized incident field [6], leading to a significant enhancement of the field intensity. We consider here a 56 nm thick model noble metal with plasma frequency of 5.73×10^{15} Hz and damping rate of 1.3×10^{14} Hz coated on a prism with a refractive index of 1.6, which launches SP resonance for the incident field at 800 nm with an incident angle of 45°. The Maxwell equations governing the field evolution are integrated by using the finite-difference time domain method to obtain the spatiotemporal distribution of the enhanced optical field. As shown in Fig. 1(a), a surface-enhanced optical field with a peak intensity enhancement of 22 is attained, which propagates along the positive direction of the x axis.

The enhanced optical field is analogous to an elliptically polarized one rather than the incident linear polarization owing to its evanescent feature [10]. As plotted in Fig. 1(b), there is a constant phase delay ($\pi/2$) between the field components perpendicular (E_y) and parallel (E_x) to the surface. The ellipticity is estimated as $E_x/E_y=0.45$. HHG could be observed for the elliptically polarized driving field owing to the quantum extension and diffusion of the electron wave packet in laser fields [11] with relatively low energy conversion efficiency as compared with the linearly

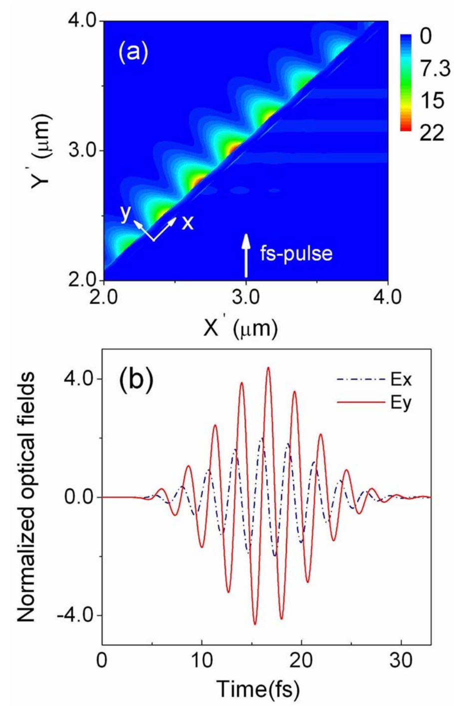

Fig. 1. (Color online) (a) Normalized intensity distribution and (b) temporal profile of the SP resonance-enhanced optical field.

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polarized one. The harmonic spectrum is obtained by Fourier transforming the field-induced atomic dipole moment, which is calculated by numerically integrating the time-dependent Schrödinger equation for a single electron [12]. As shown in Fig. 2(a), high harmonics with orders up to 13 can be generated in atomic Xe exposed to the enhanced optical field. Here, a p-polarized 12 fs pulse with central wavelength at 800 nm and peak intensity of 1.0×10^{13} W/cm^2 is considered as the incident. By using SP resonance, a recent study [13] shows that a peak intensity of $10^{15} \sim 10^{16}$ W/cm^2 can be obtained close to the surface of the thin metal film, with an intensity enhancement factor of more than 10^5. Enhanced HHG based on multielectron effects was discussed in [14], which differs from this Letter by using a surface-enhanced optical field based on SP resonance.

The intensity of the surface-enhanced optical field decreases exponentially along the direction perpendicular to the surface [6]. As shown in Fig. 1(a), this damping length in vacuum is estimated to be 248 nm, which agrees well with the feature length characterized by $[(\omega_0/c)^2 - k_p^2]^{-1/2} \approx 252$ nm, where ω_0 and k_p are, respectively, the carrier frequency of the incident field and the wave vector of the excited plasmon parallel to the surface. Consequently, HHG can occur only for atoms exposed to a thin spatial region close to the surface with a sufficiently driving intensity. For example, as shown in Fig. 2(b), the seventh

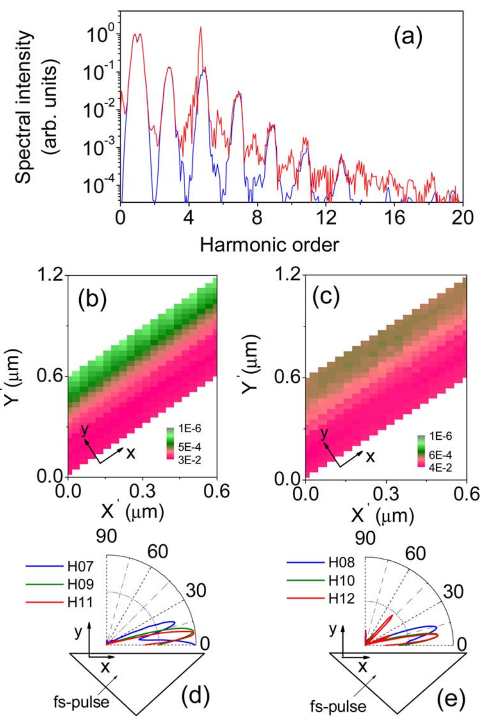

Fig. 2. (Color online) (a) Generated high-harmonic spectra as the atom is exposed to the surface-enhanced optical field with [red (upper) curve] and without [blue (lower) curve] an external static electric field. (b),(c) Intensity distributions of the generated harmonics as the atoms are exposed to various positions close to the surface. (d),(e) Emission probabilities of the generated harmonics as a function of emission angle. (b),(d) and (c),(e) represent, respectively, the results without and with an external static electric field.

harmonic can be generated only as the atoms are exposed less than 264 nm away from the surface. From the experimental point of view, as the losses of the evanescent wave owing to the absorption and heating of the lattices in the metal are considered, the enhanced optical field decays along the surface as it propagates with a typical length of micronmeters. Hence, harmonics can be generated only in a wavelength-scaled spatial region, and the propagation effect is weak and not included in our current simulations.

As the enhanced optical field propagates in the incident plane with a rotating polarization state, the generated harmonics can be detected along various directions in the $x-y$ plane. 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 [15]. As plotted in Fig. 2(d), there is a noticeable emission probability at an angle of 10° relative to the surface owing to the fact that the enhanced optical field is elliptically polarized with a larger field component perpendicular rather than parallel to the surface. Since the generated harmonics do not always propagate along the same direction as the fundamental wave, here, the conventional phase-matching condition by considering the phase lag between the fundamental and the harmonic waves is included in the superposition of the generated harmonics from atoms at different locations.

The nuclear potential seen by the outmost electron can be modified as an additional static electric field is applied together with the optical field, which breaks up the system symmetry and depresses the ionization potential combined with the field component in the same direction. For instance, by applying a static electric field parallel to the surface of the electric amplitude equivalent to 0.2 of the incident peak field, even-order harmonics are observed and the conversion efficiency is increased for the emission spectrum shown in Fig. 2(a). The intensities of the even-order harmonics can be increased by more than 2 orders of magnitude with a static electric field. Figure 2(c) shows that the harmonic generation in this case is also mainly concentrated in a wavelength-scaled spatial region. In addition, the static electric field parallel to the surface suppresses the nuclear potential as the optical field component in the same direction does and thereby increases the normal harmonic emission probability. This can be seen more clearly for the even-order harmonics as shown in Fig. 2(e) because their appearance directly originated from the additional static electric field.

It has been demonstrated that a linearly polarized ultrashort pulse can be obtained by using the so-called polarization gating technique [16] with two properly delayed counterrotating elliptically polarized pulses, which has been successfully used to enhance HHG. Similarly, a linearly polarized intense optical pulse close to the surface can be obtained by synchronizing two counterpropagating surface-enhanced pulses with counterrotating polarization states. For example, by launching an additional surface-enhanced optical field propagating along the negative direction of the *x* axis with an identical right-side incident femtosecond pulse, as plotted in Fig. 3(a), a time-gated linearly polarized field with a peak intensity enhancement factor of 40 (relative to the total incident intensity) is obtained resulting from its interference with the bottom-incident launched surface-enhanced field. Consequently, as shown in Fig. 3(b), the energy conversion efficiency as well as the frequency span of the generated harmonics is significantly enhanced by this polarization-gated surface-enhanced optical field with much lower incident intensity. The corresponding emission directions of the generated harmonics are plotted in Fig. 3(c), where a periodic standing wave is formed for the SP waves and contributes to the final emissions.

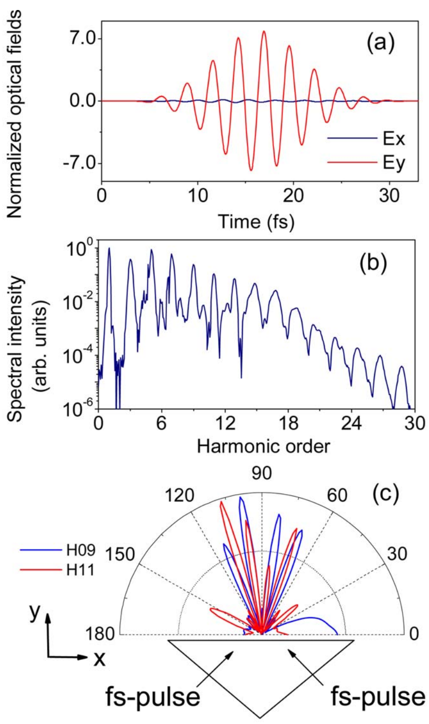

Fig. 3. (Color online) (a) Normalized surface-enhanced optical fields under the driving of two identical femtosecond pulses incident from the bottom and right with a zero time delay. (b) Generated harmonic spectrum as the atom is exposed to the polarization-gated surface-enhanced optical field with the peak intensity of each identical pulse as 1.0 $\times 10^{12}$ W/cm^2. (c) Emission directions of the generated harmonics with polarization-gated surface-enhanced optical field.

In summary, in addition to enhancement cavities [2–5], our numerical results show that high harmonics can be straightforward generated in noble gases exposed to the SP resonance-enhanced optical field, which can be used for XUV frequency comb generation and extends the applications of plasmonics. Another potential feature of this alternative approach is to generate XUV frequency combs with a fairly compact or even microstructured device with enhanced optical fields close to the surface, which can be further integrated for practical uses. At the same time, the generated harmonics separate directly from the driving field without any filter. Experimental implementation of this approach may lead to new opportunities in these fields.

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