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Extreme-ultraviolet frequency combs by surface-enhanced optical fields with diatomic molecules

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An efficient and straightforward approach is proposed to produce extreme ultraviolet frequency combs by using high-order harmonic generation from diatomic molecules driven by surface-enhanced high-repetition-rate femtosecond pulses. For a nonpolar molecule, the maximum (minimum) energy conversion efficiency is observed as the molecule is oriented at an angle of 27° (108°) relative to the surface. For a polar molecule, the frequency components of the generated ultraviolet frequency combs are doubled as both odd- and even-order harmonics are generated, which shows different orientation dependence and shifts from the nonpolar molecule. The generated ultraviolet frequency combs are mainly emitted with angles smaller than 30° close to the surface, while there is a noticeable emission probability around the directions perpendicular to the surface when the molecules are oriented with large angles.

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I. INTRODUCTION

Extreme ultraviolet (xuv) frequency combs can be generated through enhancement-cavity-based high harmonic generation (HHG) processes [1,2] with rare gas atoms, which can be used as a powerful tool for ultrahigh precision spectroscopy in the xuv spectral region [3] and optical frequency metrology [4]. Well-controlled external cavities with extremely high finesse are required to obtain intense high-repetition femtosecond (fs) pulses to drive HHG [1,2], where a high-precision electronic phase-loop-locking system and broadband high-reflection chirped mirrors are needed to strictly compensate for the involved dispersions and losses. The generated xuv frequency combs are coupled out of the external cavity by inserting a thin Brewster plate [1,2] or drilling a small hole in one of the ending concave mirrors [5] or using noncollinear cavity-enhanced HHG processes [6]. Actually, except for resonant cavity enhancement, high-repetition-rate pulse intensity can be enhanced by a factor ranging from tens to hundreds as the incident field is resonant with the surface plasmons (SP) [7], which has been demonstrated to enhance nonlinear processes, such as stimulated Raman scattering [8], second harmonic generation [9], and so forth. Such an intensity enhancement factor is comparable to the typical value obtained on the basis of the traditional enhancement cavity [1,2] to ensure high order nonlinear processes (e.g., HHG), which is fairly reliable, efficient, and straightforward.

In this paper, we demonstrate that the enhanced optical fields by SP resonance (SPR) instead of enhancement cavity can be used to produce xuv frequency combs through HHG processes with diatomic molecules in a scheme as shown in Fig. 1. The fs pulses from the oscillator are prechirped or reshaped to compensate for the ineluctable dispersions introduced by the optical components, and to manipulate the generated xuv frequency combs. For example, in Kretschmann configuration [7], the optical field intensity is significantly enhanced close to the surface as the incident pulse resonates with the SP, where xuv frequency combs can be generated through HHG processes as atoms or molecules are exposed to such intense fields. The energy conversion efficiency, emission probability as a function of emission angle, and spectral profile of the generated xuv frequency combs are studied in detail, which are found to be sensitive to the molecular orientation relative to the surface and to the molecular polarity, i.e., polar or nonpolar molecules.

II. MODEL

In the present work, we consider molecules rather than atoms as the nonlinear medium to produce xuv frequency combs. Molecules with additional degrees of freedom and complicated symmetries provide us much richer physical phenomena under strong optical fields, such as the orientation- [10] and orbital symmetry- [11,12] dependent harmonic emission probabilities as well as the generation of the even-order harmonics with polar molecules [13]. Here, in atomic units, we consider a model diatomic molecule with the potential for the outmost single electron as $V(x,y)=-\Sigma_{k=1,2}\ Z_k/[(x-x_k)^2+(y-y_k)^2+\varepsilon_k^2]^{1/2}$, where $(x_1,y_1)=(R/2\cos\theta,R/2\sin\theta)$ and $(x_2,y_2)=(-x_1,-y_1)$ are the nuclei positions with $R=2.5$ and θ stands for the internuclear distance and the angle between the molecular axis and the x axis, respectively. $Z_1=Z_2=1$ are the nuclear charges,

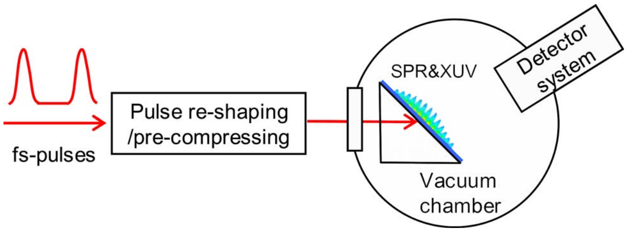

FIG. 1. (Color online) Schematic configuration of xuv frequency comb generation based on SPR-enhanced optical fields, where the incident fs pulses from the oscillator are prechirped or reshaped to manipulate the output spectra.

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and $\varepsilon_{k=1,2}$ are the soft well parameters, which are used to tune the initial state energy. The repulsion between the electrons is not included [14]. The initial ground state of the diatomic molecule is obtained by field-free propagating a random state along negative imaginary time. The nonpolar molecules are constructed by setting $\varepsilon_1^2=\varepsilon_2^2=8.8$, which corresponds to an initial state energy of 12.0 eV. Whereas, the polar molecule with different depths of the nuclei wells is constructed by setting $\varepsilon_1^2=1/6\varepsilon_2^2=4.0$, so that it has the same ground state energy as the nonpolar molecule.

The Kretschmann configuration [7] is considered to enhance the intensity of the incident optical fields through SPR. 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 SPR for incident field with a central wavelength of 800 nm at an incident angle of 45°. The full distribution of the localized 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. Throughout this paper, we define the direction parallel and perpendicular to the surface, respectively, as the x and y axes.

III. RESULTS AND DISCUSSIONS

When the incident p-polarized optical field resonates with the SP, the energy of the incident field is first stored in the metal film during the lifetime of the SP, which is then released into a subwavelength space and leads to a significant enhancement of the optical field intensity with preserved pulse duration close to the surface [7]. As an evanescent wave, the SPR-enhanced optical field decreases exponentially in the y direction and propagates for a certain distance along the x direction. For example, an intensity enhancement factor of 22 close to the surface is observed for the incident fs pulse with a central wavelength at 800 nm at an incident angle of 45°. The detailed time-dependent evolution of the enhanced optical field shows that the electric field component along the y axis is $\pi/2$ delayed as compared with the component along the x axis, and the ellipticity is estimated to be 0.45 (E_{x0}/E_{y0}). Therefore the SPR-enhanced optical field is an elliptically polarized field rather than a linearly polarized one [15].

Due to the quantum extension and diffusion of the electron wave packet, it is demonstrated that high harmonics can be generated even with an elliptically polarized optical field [16]. Furthermore, for molecules with large internuclear distance, it is possible to enhance HHG by using an elliptically polarized optical field with a small ellipticity [16], which may enhance the collision probability of the returning electron with the neighbor nucleus. For the counterclockwise elliptically polarized SPR-enhanced optical field with a larger field component along the y direction, our numerical simulation shows that the electron wave packet is first stretched to the negative direction of the y axis and then rotates clockwise. This leads to a higher probability for the electron wave packet to collide with the nuclei core when it is localized in the third quadrant as compared with the others. Therefore as shown in the inset of Fig. 2(a), the generated harmonic intensity shows a clear orientation dependence, where the maximum conversion efficiency is reached at a angle of 27° and the minimum one at an angle of 108°. The corresponding harmonic spectra when the molecule orients at these two specific angles are also plotted in Fig. 2(a). Obviously, orientation dependence is more evident for the higher harmonic orders, where the collision mechanism is more essential as compared with the lower orders. Here, the orientation angle of the molecule (θ) is defined as the angle between the molecular axis and the x axis or equivalently the surface. The incident optical field is a p-polarized 12-fs pulse with a central wavelength of 800 nm and an intensity of 6×10^{12} W/cm^2. As we can see from the harmonic spectrum, xuv frequency combs centered at 47 nm can be produced.

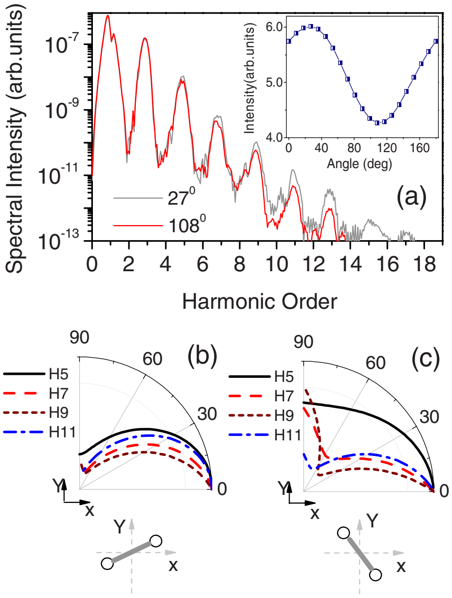

FIG. 2. (Color online) (a) The generated harmonic spectra from nonpolar molecules with SPR-enhanced optical fields. The inset is the conversion efficiency of the generated even-order harmonics as a function of the orientation angle of the nonpolar molecule. (b) and (c) plot the emission probabilities of the generated harmonics as a function of emission angle when the nonpolar molecule is oriented at angles of 27° and 108° relative to the surface, respectively.

Differing from nonpolar molecules, even-order harmonics can be generated for polar molecules resulting from the oscillation of the electron wave packet between the nuclei with asymmetric Coulomb potentials [13]. Consequently, as shown in Fig. 3, both odd- and even-order harmonics are observed when the polar molecules are exposed to our SPR-enhanced optical fields, which may significantly expand the frequency components of the produced xuv frequency combs and lead to supercontinuum frequency combs. Notice, as shown in the inset of Fig. 3(a), even-order harmonics can be generated only when an optical field with polarization parallel to the molecular axis is applied instead of the perpendicular one. Therefore as plotted in the inset of Fig. 3(b), different orientation dependences of the generated odd- and even-order harmonics are observed. The maximum (minimum)

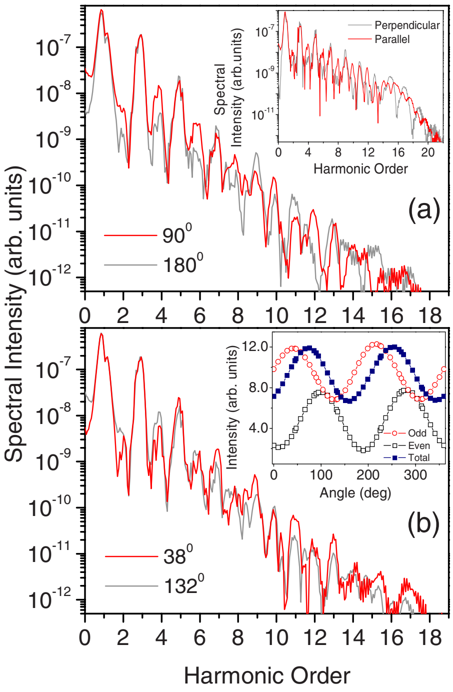

FIG. 3. (Color online) The generated harmonic spectra from a polar molecule with SPR-enhanced optical fields when it is orientated at angles of (a) 90° and 180° and (b) 38° and 132°, respectively. The inset of (a) shows the generated harmonic spectra from the polar molecule when a linear polarized optical field with polarization parallel or perpendicular to the molecular axis is applied. The inset of (b) plots the conversion efficiency of the generated odd-order, even-order, and total harmonics from a polar molecule as a function of the orientation angle with surface-enhanced optical fields.

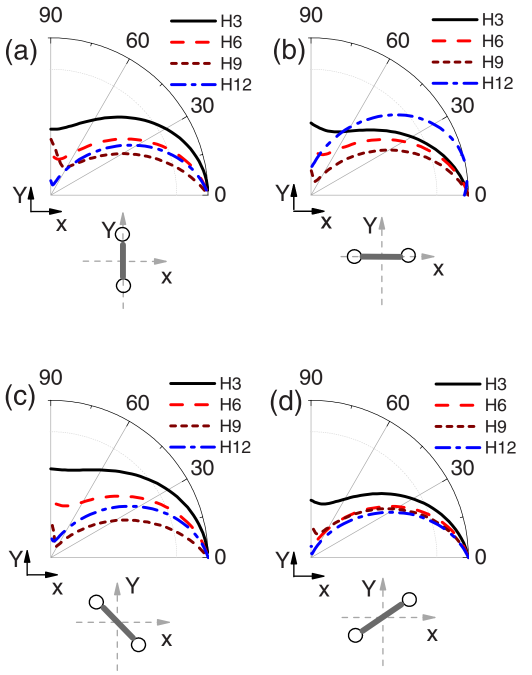

FIG. 4. (Color online) The emission probabilities of the generated harmonic orders from a polar molecule as a function of emission angle when it is oriented at angles of (a) 180°, (b) 0°, (c) 132°, and (d) 38°, respectively.

conversion efficiency for the even-order harmonics is observed when the polar molecule is oriented along the $y(x)$ axis, where the projected optical field along the molecular axis is the maximum (minimum). The orientation dependence of the odd-order harmonics for the polar molecule is similar to the nonpolar molecule with a small shift of the angle for the maximum (minimum) conversion efficiency from 27° (108°) to 38° (132°) due to the asymmetric potential wells, which drags the electron wave-packet partial toward the deeper-well nuclear. The detailed harmonic spectra when the polar molecule orients at these specific angles are plotted in Figs. 3(a) and 3(b), where the different orientation-dependent conversion efficiencies between the odd and even order harmonics can be seen clearly. The difference of the conversion efficiency between the generated odd and even order harmonics originates from the fact that they are produced through different approaches, as shown in the inset of Fig. 3(a). This provides us a possible way to manipulate the intensity profile of the produced xuv frequency combs at different frequencies by properly controlling the orientation of the molecules. This is expected to be observed experimentally as the distribution of the molecular axis angle to the laser polarization is narrow enough.

Since the enhanced optical field is composed of two electric field components with orthogonal polarizations, the generated xuv frequency combs will emit along different directions in the propagation plan. The emission probability of the generated frequency combs along a direction $\hat{e}=(\cos\alpha,\sin\alpha)$ is obtained by Fourier transforming the projected dipole acceleration $\langle\psi|\sin\alpha\,\partial F(x,y)/\partial x-\cos\alpha\,\partial F(x,y)/\partial y|\psi\rangle$ into the frequency domain, where $F(x,y)=V(x,y)+xE_x+yE_y$ and α is the crossing angle between the x axis and $\hat{e}$ [14]. As shown in Figs. 2(b) and 2(c), the generated frequency combs are mainly emitted with angles smaller than 30° for the 27° oriented nonpolar molecules, while there is a noticeable emission probability close to the y axis for the 108° oriented nonpolar molecule. The corresponding emission probabilities as a function of emission angle of the generated harmonics from polar molecules are shown in Fig. 4. Similar to the nonpolar molecules, the emission is mainly constricted within an angle smaller than 30°, while the emission probability along the y axis is somewhat higher as the polar molecule is oriented in the second and fourth quadrants.

It is demonstrated that the interference effect [14,17] between the emitted harmonics from the neighbor nuclei has a remarkable influence on the final harmonic spectrum when molecules with several nuclei are considered, which depends on the internuclear distance and molecular orientation. Whereas the frequency of the generated harmonics in this work with high-repetition SPR-enhanced optical pulses is relatively low and out of the interference region, the constructive or destructive interference between the emitted xuv frequency combs from neighbor nuclei does not occur and therefore is negligible. Here, we consider aligned molecules rather than randomly oriented ones, which may be an average result in the experiment. While, on the other hand, it is also possible to align molecules, especially polar molecules, with a static electric field or additional laser fields [18]. Experimentally, the SPR-enhanced optical fields propagating along the x direction are typically limited to a length of several microns, and exponentially decay along the y direction in a subwavelength range. Consequently, xuv frequency combs can be generated only in a wavelength-scaled spatial region. Therefore we expect the influences from phase mismatching as well as the interference between the molecules in the macroscopic gas target are not as significant as the conventional HHG processes with a large interaction region, where such effects always return to the final output spectra. Additionally, the destructive effect may be further weakened as prealigned molecules [18] are considered.

IV. CONCLUSIONS

In summary, as the incident near-ir high-repetition rate fs pulses from the oscillator resonate with the SP, the resulting enhanced optical fields close to the surface can be used to produce xuv frequency combs through HHG processes. Molecules provide us an additional way to control the generated xuv frequency combs, including the conversion efficiency, emission probabilities along different directions, and a detailed spectral profile. Furthermore, even-order harmonics can be generated by using polar molecules, which directly doubles the frequency components of the generated xuv frequency combs. Rather than enhancement external cavities, the results presented in this paper provide us an alternative approach to generate xuv frequency combs with unique features, and extensively practical applications are expected.

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