Supercontinuous high harmonic generation from asymmetric molecules in the presence of a terahertz field*

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We have investigated high-order harmonic generation from asymmetric molecules. It is found that supercontinuous high harmonics, which are produced from asymmetric molecules by significantly steering the ionization, are broken down when the electric field of the 5-fs driving laser pulse is increased to 0.16 a.u. The high harmonic generation from asymmetric molecules with the presence of a terahertz field is also investigated. This reveals that the terahertz controlled laser pulse significantly increases the energy difference between photons, emitted from the ejected electrons, in the first and second halves of the optical cycle at the centre of the driving laser pulse. In this way, a 200-eV broadband supercontinuum can be produced in the plateau, from which a 60-as pulse with a bandwidth of 60 eV can be directly obtained with a minor post-pulse.

Keywords: high-order harmonic, attosecond pulse, supercontinuum

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1. Introduction

In the past decade, attosecond science[1] has emerged as a new field to study the behaviour of electrons in atoms and molecules. Success in this new field requires efficient production of isolated attosecond pulses. Among the available techniques for synthesizing attosecond pulses, high-order harmonic generation (HHG), especially broadband continuum HHG, appears to hold the greatest promise.[2−8] Isolated attosecond pulses were first achieved by band-pass filtering the harmonics, which were generated when few-cycle driving laser pulses interacted with a jet of neon atoms.[2] Very recently, Mashiko *et al.*[9] directly generated an efficient broad-band supercontinuum using a double optical gating (DOG) technology, which supports a pulse duration of 16 as and it is below one atomic unit of time (24 as). Our previous work also generated a broadband supercontinuum using a few-cycle two-colour laser pulse and a high-order pulse, from which an isolated 24-as pulse can be directly obtained.[10]

The mechanism of high harmonic generation can be well understood using the three-step model:[11] ionization, acceleration and recombination. During recombination, a photon is emitted with energy equal to the ionization potential plus the kinetic energy of a recombining electron. Therefore, harmonic generation can be controlled by manipulating these different steps. It has been proposed that the two-colour field can form a broadband supercontinuum near the cutoff by enlarging the energy difference between the highest and the second highest half-cycle photon,[12,13] and it can also control the tunnel ionization and restrict the ionization peak with one half optical cycle.[14] Mashiko *et al.*[15] recently combined the ionization gating and polarization gating techniques to simultaneously gate the ionization and recombination steps, termed as double optical gating (DOG), and they directly produced an efficient broadband supercontinuum in the plateau. Very recently, we also proposed a method to obtain an ultra-broadband supercontinuum combining mid-infrared chirped-pulse modulation and a generalized polarization gating, which supports the isolated sub-100 attosecond pulse generation with a tunable

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wavelength.[16] Hong *et al.*[17] generated isolated sub-100-as pulses with tunable wavelengths using a mid-IR modulated polarization gating technique. Lan *et al.* found that HHG can be controlled with asymmetric molecules by steering the ionization, by which the supercontinuum was also generated in the plateau since the electron prefers to locate at one nucleus.[18,19] During consecutive half optical cycles, the ionization asymmetry in asymmetric molecules is reduced when the electric field of the driving laser pulse increases, therefore it is important to investigate in what electric field conditions for the driving laser pulses the ionization asymmetry will be broken down.

In this paper, we present our investigation in the approximate range of electric field that breaks down the supercontinuous high harmonics, which were produced from asymmetric molecules by significantly steering the ionization. We found that the supercontinuous high harmonics were broken down when the electric field of the 5-fs driving laser pulse was increased to 0.16 a.u., which is also suitable for the driving laser pulse of longer duration. The high harmonic generation from asymmetric molecules was also investigated with the presence of a terahertz (THz) field, as schemes for high power THz pulse generation have been developed with the peak electric field reaching tens of kV/cm and even exceeding MV/cm.[20,21] This revealed that the THz controlled laser pulse significantly increases the energy difference between photons, emitted from ejected electrons, in the first and second halves of the optical cycle at the centre of the driving laser pulse. In this way, a 200-eV broadband supercontinuum was produced in the plateau, from which a 60-as pulse with a bandwidth of 60 eV was directly obtained with a minor post-pulse.

2. Model and method

We simulated the laser-molecular interaction by numerically solving the time-dependent Schrödinger equation (TDSE). In our calculation, we use a soft-core potential model[19,22]

$$V(x) = -Z_1/\sqrt{1+(x+R/2)^2} - Z_2/\sqrt{1+(x-R/2)^2}, \quad (1)$$

where R is the internuclear distance, and Z_1 and Z_2 are the electric charges of the nuclei. Here we have assumed that the nuclei are fixed at $\pm R/2$ since the molecular vibrational period is longer than the laser pulse duration.[23] We adopt $R = 4$ a.u., $Z_1 = 1$ a.u., and $Z_2 = 2$ a.u.[19,24] to simulate the asymmetric molecule HeH^{2+}. The electric field is the superposition of a linearly polarized near-infrared driving laser pulse of 5 fs at 800 nm and a linearly polarized THz field at 100 μm. The total electric field is expressed by

$$E(t) = E_0 f(t)\sin(\omega_0 t) - E_{\rm THz}\sin(\omega_{\rm THz} t), \quad (2)$$

where E_0 and $E_{\rm THz}$ are the amplitudes and ω_0 and $\omega_{\rm THz}$ are the frequencies of the driving pulse and the THz field, respectively. $f(t)$ is the envelope of the driving pulse, given by $f(t) = \cos^2(\pi t/T)$, where $T = 552$ a.u. corresponding to a duration of 5 fs, which is the full width at half maximum. The molecule is aligned along the laser polarization direction, which can be achieved with a prepulse or static electric field.[25] Sakai *et al.* also demonstrated another effective way to orientate asymmetric molecules with combined pulses.[26,27] Then the electron predominantly oscillates in the laser polarization direction, and the one-dimensional model will produce quite reliable results, which have been widely used in previous work.[28]

3. Results and discussion

We first investigated the approximate limit of the electric field that breaks down the supercontinuous high harmonics produced from asymmetric molecules by significantly steering the ionization. In the calculations, $E_{\rm THz} = 0$, and E_0 is either 0.14 a.u. or 0.16 a.u. The spectra of the high harmonics generation from the asymmetric molecules HeH^{2+} are shown in Fig. 1, where the harmonic spectrum with $E_0 = 0.14$ a.u. (bold solid curve) has been shifted 2 units down for clarity. At first glance, the two spectra exhibit a similar overall structure: the plateau covers a broad bandwidth and exhibits an almost constant intensity, and then it drops quickly in the cutoff. However, their fine structures are quite different. When $E_0 = 0.14$ a.u., the plateau is rather regular and continuous for the harmonic order higher than 60. It suggests that the HHG can be controlled by using asymmetric molecules by steering the ionization. However, when the electric field is increased to 0.16 a.u., the harmonic spectrum becomes irregular, which indicates that the supercontinuous high harmonics, produced from asymmetric molecules by significantly steering the ionization, is now broken down.

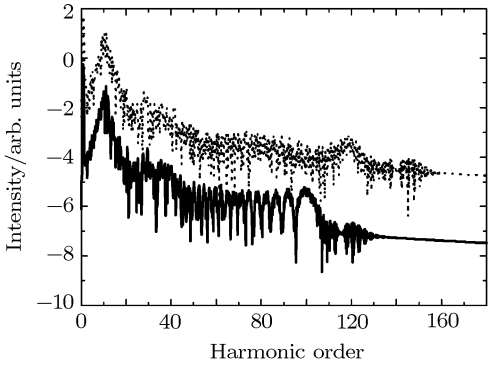


Fig. 1. Spectra of high harmonics generated from asymmetric molecules in a fundamental field alone. The electric fields are 0.14 a.u. (bold solid line) and 0.16 a.u. (thin dotted line), respectively.

In order to show a clear physical picture, we analyse the electron dynamics using a classical three-step model, and the results are shown in Fig. 2. The solid curve and dotted curve in Fig. 2(a) are the waveforms with the electric fields $E_0 = 0.14$ a.u. and $E_0 = 0.16$ a.u., respectively. In Figs. 2(b) and 2(c), we show the classical electron energy and ionized electron fluxes as a function of the ionization time, with the electric fields of 0.14 a.u. and 0.16 a.u., respectively. The ionized electron flux describes the first step and is calculated via $\Gamma(t) = -d_t(\log N(t))$, where $N(t)$ is the norm of wave function in the bound states.[29] For $E_0 = 0.14$ a.u., as shown in Fig. 2(b), the ionization takes place around the electric peaks at $t = 0.0$, 0.5, and $1.0T_0$, where T_0 is the optical cycle of the driving field. Consequently, electrons ionized around $t = 0$ are accelerated by the highest peak, and they accumulate the highest energy of $97\omega_0$. The second highest energy is $64\omega_0$ acquired by electrons ionized around $t = 0.5T_0$. Therefore, the harmonic spectrum is regular and continuous for the harmonics orders higher than 60, as shown by the bold blue curve in Fig. 1. However, when the electric field reaches 0.16 a.u., the ionization takes place around the peaks at $t = -0.5$, 0.0, 0.5, and $1.0T_0$, as shown in Fig. 2(c). Although the electron ionization flux is weak around the peak at $t = -0.5T_0$, it significantly affects supercontinuous high harmonics produced by the asymmetric molecules shown in Fig. 1 (thin dotted curve). To further clarify the above analysis, we also investigated the emission time of the harmonics using a time-frequency analysis method. The results are shown in Fig. 3. where the quantum path P2 in Fig. 3(b) ($E_0 = 0.16$ a.u.) is stronger than that in Fig. 3(a) ($E_0 = 0.14$ a.u.).

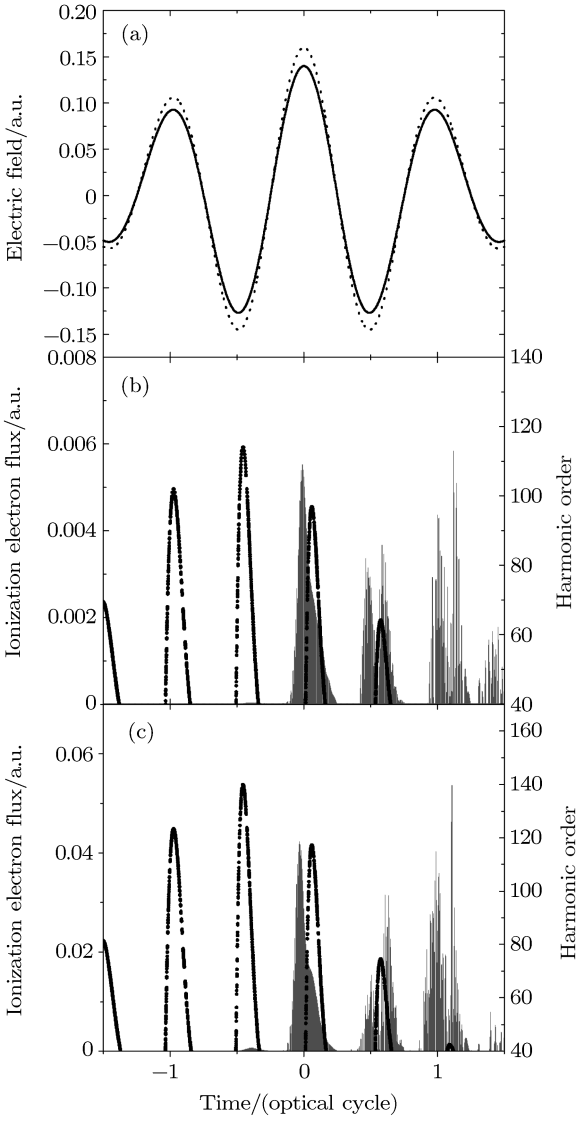


Fig. 2. (a) The waveforms of the fundamental field with $E_0 = 0.14$ a.u. (solid curve) and $E_0 = 0.16$ a.u. (dotted curve); the classical electron energy (dotted curve) and the ionized electron flux (filled grey curve) as a function of the ionization time, with the fundamental field: (b) $E_0 = 0.14$ a.u. and (c) $E_0 = 0.16$ a.u.

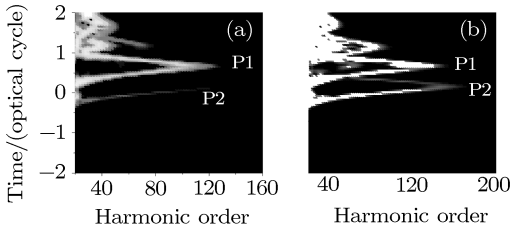


Fig. 3. The time-frequency distribution of HHG with the fundamental field: (a) $E_0 = 0.14$ a.u. and (b) $E_0 = 0.16$ a.u.

We also investigated the high harmonic generation from asymmetric molecules with the presence of a terahertz field. In our scheme, since the wavelength

of the terahertz field is too large compared with that of the near-infrared laser field and the pulse duration of the near-infrared field is too short (only a few cycles), the terahertz field can actually be considered as a static electric field. In our calculations, the electric field of the fundamental pulse and the terahertz field are 0.16 a.u. and 0.04 a.u., respectively. Figure 4 shows the spectrum of the high harmonics generated from asymmetric molecules with the presence of a terahertz field (bold solid curve). As a comparison, the spectrum of the high harmonics with the fundamental field alone is also shown in the figure (thin dotted curve) but has been shifted down 2 units for clarity. One can clearly see that the spectrum is irregular for the low harmonics, but fascinatingly becomes regular and continuous from 95 to 225 harmonics, and it forms a supercontinuum with a bandwidth of about 200 eV with the presence of a terahertz field. It must be noted that the supercontinuum will not be generated when the direction of the terahertz field is reversed.

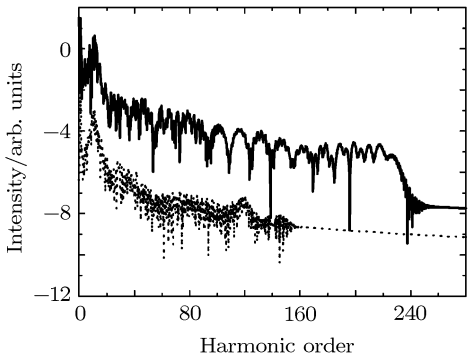


Fig. 4. High harmonics spectra generated from asymmetric molecules with the terahertz field (bold solid curve) and in a fundamental field alone (thin dotted curve).

A physical picture of the above described phenomenon is clarified by studying the HHG process using the semiclassical three-step model. Figure 5(a) shows the waveforms of the fundamental field without (dotted curve) and with (solid curve) the presence of a terahertz field. In Fig. 5(b), we present the kinetic energy and the electron ionization fluxes as a function of the ionization time. As shown in Fig. 5(b), the asymmetric molecule is predominantly ionized around the electric field at $t = -0.5T_0, 0.0T_0$, and $0.5T_0$. Consequently, the electrons ionized around $t = 0.0$ are accelerated by the highest peak, and they accumulate the highest energy of $225\omega_0$. The second highest energy comes from the electron ionized around $t = -0.5T_0$ and $0.5T_0$, and it is $95\omega_0$. Therefore, the harmonic spectrum is regular and continuous for the harmonics order higher than 95, as shown by the bold curve in Fig. 4. Similarly to the case absenting from the terahertz field, we further investigate the emission time of the harmonics using the time-frequency analysis method. The result is shown in Fig. 6, where two main peaks contribute to the harmonics (marked as P1 and P2), but only one dominant radiation peak P1 is visible for the high harmonics above $95\omega_0$. The features are consistent with the results in Fig. 5. We emphasize that the ionization asymmetry in asymmetric molecules plays a key role in the generation of the supercontinuum. Without the ionization asymmetry, the ionization at $t = -0.5T_0$ will not be so weak and the ionization at $t = 0.0$ will not be so strong.

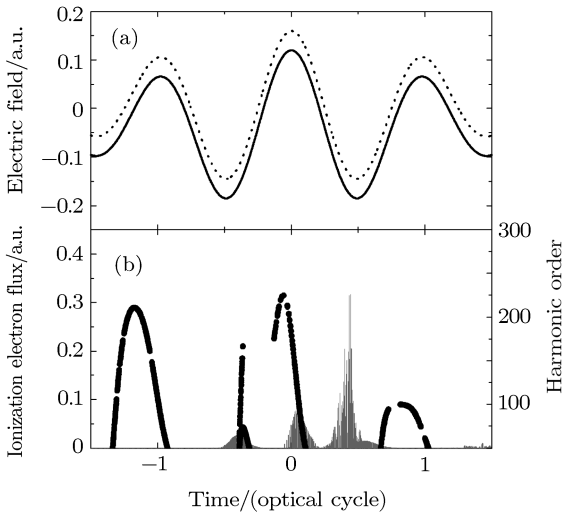


Fig. 5. (a) The waveforms of the fundamental field without (dotted curve) and with (solid curve) the presence of a terahertz field. (b) The classical electron energy (dotted curve) and the ionized electron flux (filled curve) as a function of the ionization time with the presence of a terahertz field.

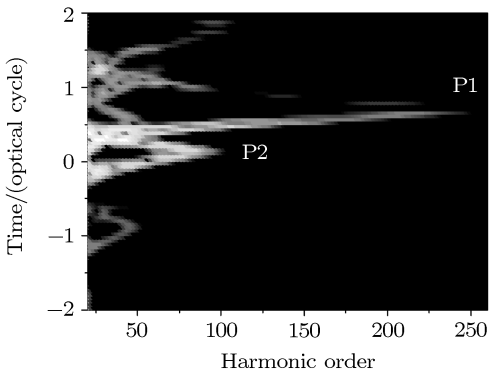


Fig. 6. The time-frequency distribution of HHG with the presence of a terahertz field.

In the following, we further investigate the isolated attosecond pulse generation. Figure 7 shows an isolated 60 as pulse by directly filtering the harmonics from $95\omega_0$ to $135\omega_0$. A satellite pulse with 2% intensity of the main pulse is observed around $t = 0.93T_0$,

which originates from the long path. In order to generate an isolated attosecond pulse, the long trajectory, which leads to spatially divergent radiation,[30] must be eliminated. This can be achieved by adding a small aperture after the HHG cell, and the harmonics and attosecond pulse originating from the long trajectory can then be removed. Alternatively, it has been demonstrated that the short trajectory can also be selected by focusing the laser pulse before the gas jet. These methods are efficient and were also demonstrated in previous studies.[31−33]

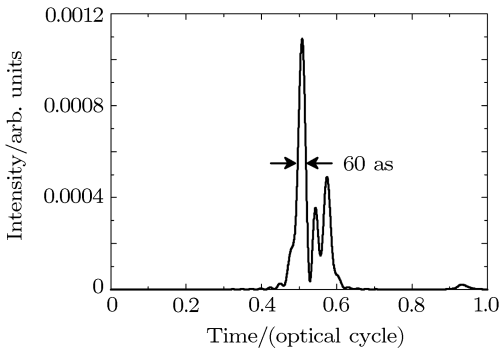


Fig. 7. The temporal profile of the isolated attosecond pulse, which is the superposition of harmonics from $95\omega_0$ to $135\omega_0$.

4. Conclusion

In conclusion, we have investigated the high-order harmonic generation from asymmetric molecules. We found that supercontinuous high harmonics produced from asymmetric molecules by significantly steering the ionization were broken down when the electric field of the 5-fs driving laser pulse was increased to 0.16 a.u. We also presented a scheme to generate broadband extreme ultraviolet supercontinuum harmonics from asymmetric molecules by the combination of the few-cycle laser pulse and terahertz field. The terahertz controlled laser pulse significantly increased the energy difference between photons in the first and second halves of the optical cycle at the centre of the laser pulse, and a 200-eV broadband supercontinuum was produced in the plateau, from which a 60-as pulse can be directly obtained with a minor post-pulse.

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