

Quantum path selection in high-order harmonic generation from aligned molecules

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Abstract: We theoretically investigate high-order harmonic generation (HHG) from aligned N₂ molecules with a driving field composed of two-color circularly polarized laser pulses. It is shown that the combination of N₂ molecules and the waveform-controlled laser field allows us to select either long or short quantum path, depending on molecular alignment angles, while in atom Ar, two paths show comparable contribution to HHG. The selection of single quantum path in aligned N₂ molecules leads to an ultrabroad and smooth XUV supercontinuum, giving rise to isolated attosecond pulses generation. Moreover, we can control the intensity ratio of two attosecond pulses by adjusting the molecular alignment angles, providing an opportunity for attosecond pump-probe technique.

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In recent decades, with the development of ultrafast laser technology, high-order harmonic generation (HHG) in a strong field regime has attracted tremendous attention due to its important application in molecular tomography, the generation of attosecond pulses and the probe of electron wavepacket dynamics with attosecond and angstrom resolution [1–4]. Generally, HHG inherently produces attosecond pulse train separated by about half optical period of the driving laser field. Therefore, to achieve single attosecond pulse desired for a lot of applications, it is necessary to gate (1) the driving pulses [5–8] or (2) the harmonic emission itself [9–13] based on three-step model. With the approach (1), one has obtained the shortest attosecond pulse in the world [6]. However, up to now, the achievement of few-cycle, high-energy laser pulse is still a challenge for most laboratories, which hampers the widespread use of attosecond technology. The commercial availability of multi-cycle, high-energy laser system motivates us to explore other optical schemes, such as multi-color field synthesis [7, 8], polarization gating [5, 11], double optical gating [14], phase matching gating [15], ionization gating [16], etc., which are capable to confine HHG within a sub-femtosecond timescale [i.e., approach (2)]. All methods mentioned above, which have been successfully performed in the atomic targets, can be extended to molecular gases. As compared to HHG from atoms, HHG from molecules strongly depends on the molecular orientations, which not only enable us to unlock molecular structures and dynamics [17–22], but also provides an addition parameter to control electron motion and thus attosecond pulse generation [23].

In this paper, we theoretically investigated HHG from N₂ molecules and found that as compared to its reference atom Ar, the strong dependence of HHG from N₂ molecules on molecular orientation allows us to select long or short quantum path with a waveform-controlled laser field constructing by two-color circularly polarized lights, giving rise to the generation of isolated attosecond pulses.

In the present work, we performed numerical simulations of HHG in the aligned N₂ molecules using the extended Lewenstein model [24–26] and MO-ADK theory [27]. The

driving laser field, which is composed of 800-nm left circularly polarized pulses and 1600-nm right circularly polarized pulses, can be expressed as [28]:

$$E_x(t) = E_0 e^{-2\ln 2 \cdot (t-T_d/2)^2 / \tau_0^2} \cos[\omega_1(t-T_d/2) + \phi_1] + E_0 e^{-2\ln 2 \cdot (t+T_d/2)^2 / \tau_0^2} \cos[\omega_2(t+T_d/2) + \phi_2], \quad (1)$$

$$E_y(t) = E_0 e^{-2\ln 2 \cdot (t-T_d/2)^2 / \tau_0^2} \sin[\omega_1(t-T_d/2) + \phi_1] - E_0 e^{-2\ln 2 \cdot (t+T_d/2)^2 / \tau_0^2} \sin[\omega_2(t+T_d/2) + \phi_2]. \quad (2)$$

Here ω_i and ϕ_i ($i = 1, 2$) denote angular frequencies and carrier envelope phases of two circularly polarized laser pulses. We used the pulse duration $\tau_0 = 8$ fs and the laser intensity $I_0 = 5 \times 10^{14}$ W/cm². T_d is the delay between two circularly polarized laser pulses. In our numerical simulation, we choose the following parameters as follows: $\phi_1 = 0$, $\phi_2 = -\pi/4$ and $T_d = 4$ fs. As shown in previous works [29, 30], because HHG is determined by the quantum-mechanical expectation value of dipole acceleration, the intensity of the n th harmonic can be obtained by calculating $\omega_n \cdot |\bar{d}_n|^2$, where ω_n is the frequency of the generated n th harmonic photon, and \bar{d}_n is the dipole matrix element for bound-free transitions [7–9, 29, 30].

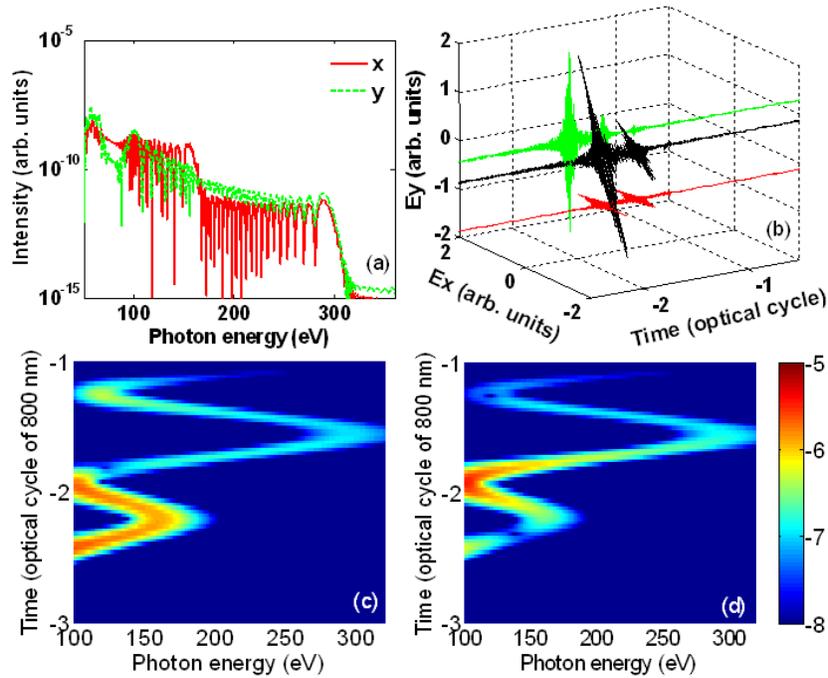


Fig. 1. (a) x (solid line) and y (dashed line) components of the Ar atom medium. (b) 3D plot of electric fields of attosecond pulses generated from spectral range of 180–240 eV. Time frequency analyses of (c) x component and (d) y component of Ar atom HHG spectra.

First, we investigated HHG in Ar atom with the synthesized laser field indicated in Eqs. (1) and (2). Figure 1(a) shows the x and y component of HHG spectra. Apparently, the y -component HHG spectrum shows a smooth supercontinuum in the spectral range of 180–240 eV, while x component presents a significant modulation due to the existence of two or multiple attosecond bursts. From the three-dimensional (3D) electric field of the synthesized attosecond pulses, we can clearly see that the HHG spectrum in the range corresponds to two attosecond pulses, namely, a main pulse emitted at about ~ 1.45 T and a weak one at ~ 1.8 T, where T is an optical period of 800-nm laser pulses. Their amplitude ratio is ~ 2.3 . It is

noteworthy that the two isolated attosecond pulses present different polarization direction. The electric field of the strong one is at an angle of $\sim 70^\circ$ with respect to x axis, while the electric field of the weak one is at an angle of $\sim 40^\circ$ with respect to x axis. Correspondingly, the time-frequency analyses in Figs. 1(c) and 1(d) illustrated that in the spectral range of 180-240 eV, two quantum paths with comparable intensity are involved, referred to as long path and short path. The main attosecond pulse and the satellite pulse, shown in Fig. 1(b), come from the short and long quantum paths, respectively.

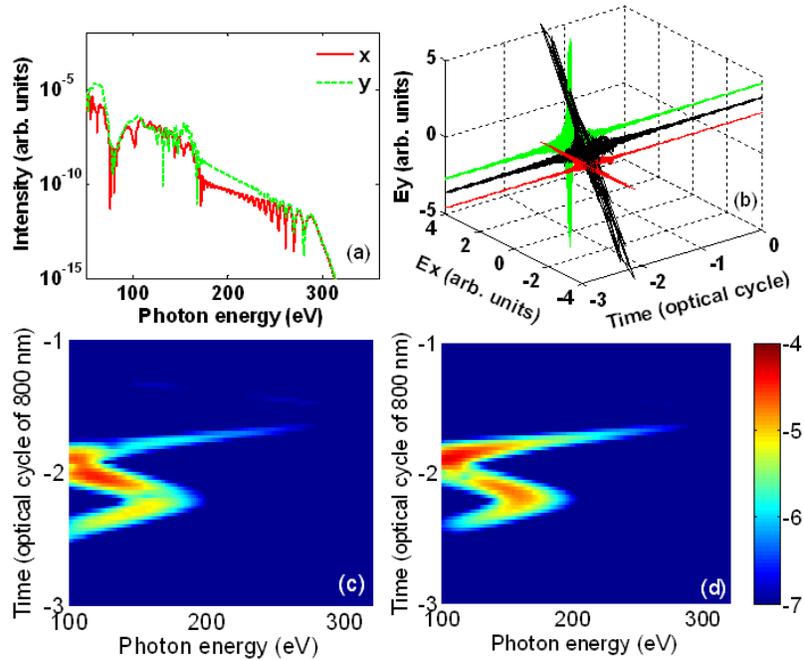


Fig. 2. (a) x (solid line) and y (dashed line) components of the N_2 molecule. (b) 3D plot of electric fields of attosecond pulses generated from spectral range of 180-240 eV. Time frequency analyses of (c) x component and (d) y component of N_2 molecule HHG spectra.

When the same laser field interacts with N_2 molecules which molecular axis is aligned at 70° with respect to the x axis, both x and y component of HHG spectra show smooth supercontinuum structures covering an extremely broad spectral range from 80 eV to 300 eV, as illustrated in Fig. 2(a). For comparison with Ar atoms, we also depict 3D electric field of attosecond pulses synthesized by the supercontinuum from 180 eV to 240 eV in Fig. 2(b). As we expected, the smooth supercontinuum corresponds to an isolated attosecond pulse, which is the same as the main pulse in Fig. 1(b) (i.e., the same polarization and the same emission time). Meanwhile, the weak satellite pulse at ~ 1.45 T disappears. Surprisingly, the corresponding time-frequency analyses in Figs. 2(c) and 2(d) clearly show that in this case only single quantum path (i.e., short path) contributes to HHG in the range of 180-240 eV, while HHG from the long path is completely suppressed. Apparently, molecular gases show significant advantages for creating an isolated attosecond pulse.

When the molecular axis is parallel to x axis, smooth XUV supercontinuum still appear in both x and y component of HHG spectra, as shown in Fig. 3(a), by which we can synthesize an isolated attosecond pulse. The isolated pulse in Fig. 3(b) has the same polarization and release time as the weak satellite pulse in Fig. 1(b). As shown in Figs. 3(c) and 3(d), the time-frequency analyses indicate that in this case, HHG from the long quantum path plays a dominating role, while HHG from the short path is very weak and thus can be neglected.

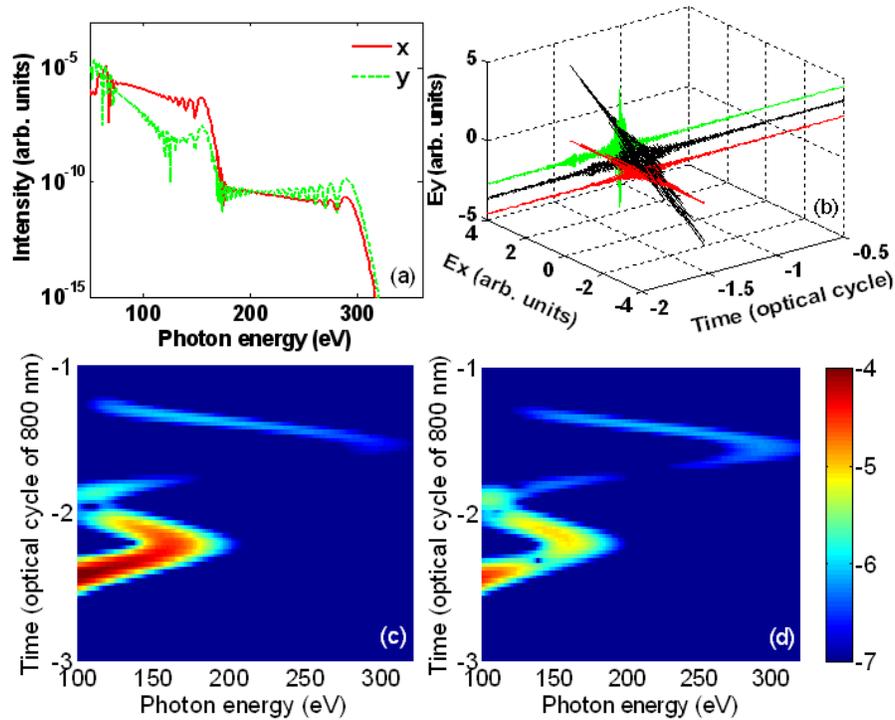


Fig. 3. Same as in Fig. 2 but for molecular alignment angle 0° .

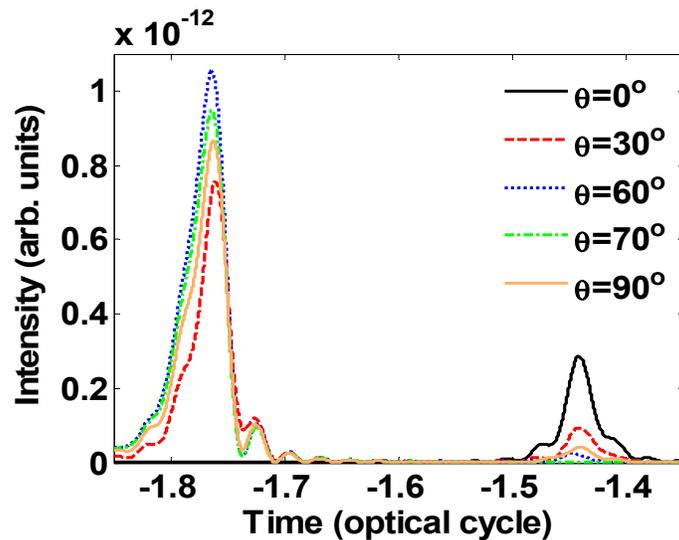


Fig. 4. The temporal profiles of attosecond pulses generated from spectral range of 180-240 eV as a function of molecular alignment angle.

At last, we investigate the dependence of attosecond pulse generation on the molecular orientation. In Fig. 4, it can be clearly seen that when the molecular axis is arranged along the x direction (i.e., $\theta = 0^\circ$), the isolated attosecond pulse released at ~ 1.45 T is the strongest and the attosecond pulse released at ~ 1.8 T is effectively suppressed, which exactly is the case in Fig. 3(b). For comparison, when the molecular axis is orientated at an angle of 70° with respect to x axis (i.e., $\theta = 70^\circ$), the strong attosecond pulse at ~ 1.8 T appears and the weak

satellite pulse disappears, which corresponds to the case of Fig. 2(b). However, the isolated attosecond pulse reaches its maximum at $\theta = 60^\circ$ rather than $\theta = 70^\circ$. When the molecules are aligned at other directions, two attosecond pulses simultaneously exist and their intensity ratio changes as the alignment angle θ of N_2 molecules, which provides a promising way for the performance of attosecond pump-probe techniques.

The calculated results above clearly imply that the combination of the alignment dependence of molecular HHG and the driving laser field with a specific waveform allows us to select a single quantum path and to create isolated attosecond pulses. In order to obtain a clear insight, here we give a detailed discussion. It is well known that HHG yield is the highest when N_2 molecules are aligned at about 0° from the polarization axis of driving laser field (i.e., parallel alignment) [25]. The reason is that, both ionization and recombination probabilities reach the maximum in the molecular ensemble of parallel alignment, which is determined by the σ_g symmetry of highest occupied molecular orbital (HOMO) of N_2 . On the contrary, in the case of perpendicular alignment, the HHG yield is the lowest due to both low ionization and recombination probabilities. The waveform of our constructed driving laser field (i.e., amplitude, phase and polarization) rapidly changes in sub-cycle time scale, leading to the rapid variation of motion trajectories of recolliding electron. Generally speaking, the motion direction of the electron recombined with parent ions determines the polarization direction of the emitted attosecond bursts. Since electrons born at different times revisit their parent ions along different directions, the released attosecond pulses show different polarization. Furthermore, taking advantage of the sensitivity of HHG in molecules on molecular orientation, we can select the attosecond pulse polarized along a certain direction and suppress attosecond pulse polarized along other directions by optimizing the direction of molecular axis with respect to the driving laser field. Our calculated results further prove this true. Two attosecond pulses appeared in Ar atom, which are from long and short quantum paths respectively, can be selected by using aligned N_2 molecules. When the molecular axis is parallel to the motion direction of the recombined electron (i. e., the polarization direction of the main attosecond pulse in Fig. 1(b), $\theta = 70^\circ$), the recombination probability of electron traveling along the short quantum path is the highest, while the recombination probability of electron along the long quantum path is largely reduced, giving rise to an isolated attosecond pulse. As indicated in Fig. 4, the attosecond pulse is not the strongest at $\theta = 70^\circ$, which could be due to that the direction of electron tunneling from the laser field is different from the direction of electron recolliding with parent ions. Therefore, ionization probability could not be the highest at $\theta = 70^\circ$. Similarly, at $\theta = 0^\circ$ rather than the polarization direction of the weak attosecond pulse in Fig. 1(b) (i.e., $\theta = 40^\circ$), the long path is selected, leading to an isolated attosecond pulse at ~ 1.45 T. At $\theta = 0^\circ$, not only the strong attosecond pulse at ~ 1.8 T is effectively suppressed, but also the selected attosecond pulse is the strongest.

To be concluded, taking advantage of the alignment dependence of HHG in N_2 molecules, we theoretically realized the selection of single quantum path by constructing a driving laser field with a rapidly varying waveform. With the scheme, we not only can obtain isolated attosecond pulses, but also can control the relative intensity of two attosecond pulses only by adjusting the direction of molecular axis. As compared to atomic species, molecular orientation provides us the additional parameters to control HHG as well as attosecond pulse generations.

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