Focus conditioning effects on molecular field-free alignment observed with high-order harmonic generation

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Abstract We investigate the focus conditioning effects on molecular field-free alignment observed with high-order harmonic generation (HHG) from CO_2 molecules. We also experimentally demonstrate that both the spectral shape and alignment signal of HHG significantly vary with changing focus position. A maximal alignment signal is achieved at a given focus position because of the optimal intensity of the driving laser. This intensity is related to the ionization potential of the molecules. These results indicate that a unique focus position provides an optimal alignment signal for practical applications.

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1 Introduction

High-order harmonic generation (HHG), which results from the interaction between femtosecond lasers and molecules, has been extensively studied as a unique technique for probing orbital structures, electron dynamics, and other ultrafast processes[1-6]. Unlike atoms, however, molecules are non-isotropic systems, and processes such as tunneling ionization or HHG can be influenced by the angle between the laser electric field vector and molecular axis[7]. Therefore, molecules should be aligned before ultrafast processes are probed; field-free alignment is advantageous because it does not interfere with subsequent applications. Guo *et al.*[8] investigated the ionization effects on molecular field-free alignment, and presented the optimal intensity of the aligning laser, thereby enabling maximal molecular alignment. In this paper, we demonstrate the focus conditioning effects on molecular field-free alignment observed with HHG, and provide the optimal intensity of the driving laser to obtain maximal alignment signals at a given focus position.

In this paper, we experimentally observe HHG from aligned CO_2 molecules, and find that both the spectral shape and alignment signal of HHG significantly vary under changing focus position. We then obtain the maximal alignment signal at the focus position that results from the optimal driving laser intensity. This intensity is related to the ionization potential of the molecules. A simple ionization model is introduced to explain the experimental results.

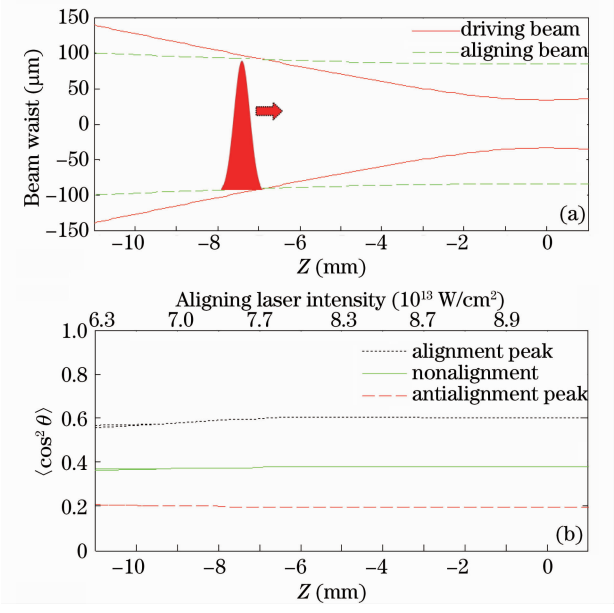

Fig. 1. (a) Gaussian beams of aligning and driving pulses in the interaction region; (b) the calculated alignment parameter $\langle\cos^2\theta\rangle$ at the non-alignment moment (19.0 ps), alignment peak (21.1 ps), and anti-alignment peak (21.9 ps) as functions of focus position (bottom axis) and as functions of aligning laser intensity (top axis).

2 Experimental Setup

The experiments were conducted using a Ti:sapphire-based chirped pulse amplification laser system, which produces 50-fs laser pulses at a center wavelength of 800 nm. The laser pulse is split into two beams: one is used as the pump pulse (for aligning molecules) and the other is used as the probe pulse (for driving HHG from

the aligned molecules). Using a lens with a 300-mm focal length, we collinearly focused the two beams onto a pulsed supersonic molecular beam located in a high-vacuum interaction chamber. The aligning pulse is propagated through a soft iris aperture ($d = 4$ mm) with a Rayleigh length of 17 mm and a beam waist of 84.3 μm; the driving pulse is propagated through another soft iris aperture ($d = 10$ mm) with a Rayleigh length of 2.75 mm and a beam waist of 33.8 μm (Fig. 1(a)). Z is the scalar axis of the lens position. If $Z = 0$ mm, the focus position is located under the gas jet; if $Z < 0$ mm, the gas jet is located before the focus position. The intensity of the aligning laser in the interaction region is varied from 6.3×10^{13} to 9.0×10^{13} W/cm^2, and the intensity of the driving laser in the interaction region is varied from 1.6×10^{14} to 28.0×10^{14} W/cm^2. The laser focus is about 1-mm downstream of a 0.25-mm-diameter nozzle orifice. The stagnation pressure of CO_2 gas is around 2 bars, leading to a rotational temperature of tens of Kelvin. The generated high-order harmonics are detected by a flat-field grating spectrometer equipped with a soft X-ray charge-coupled device (CCD) camera.

3 Result and Discussion

When a CO_2 molecule is irradiated by a short laser pulse at 42.7 ps (a duration that is considerably shorter than the molecular rotational period, 50 fs), nonadiabatic field-free alignment is achieved by the excitation of a rotational wave packet $\psi(t) = \sum_{J,M} A_{J,M}(t) |J, M\rangle$[9, 10]. The temporal evolution of alignment parameter $\langle \cos^2 \theta \rangle$ can be calculated by solving the time-dependent Schrödinger equation[9, 11]. In our calculation, the initial rotational temperature is 80 K. The results are shown in Fig. 2 (dashed line, right axis). The experimentally measured the 23rd harmonic intensity as a function of pump-probe delay is also presented in Fig. 2 (solid line, left axis). The experimental focus position is $Z = -9$ mm, the intensity of the aligning laser is about 7×10^{13} W/cm^2, and the intensity of the driving laser is about 2.4×10^{14} W/cm^2. The calculated values of alignment parameter $\langle \cos^2 \theta \rangle$ at the nonalignment moment (19.0 ps), alignment peak (21.1 ps), and the anti-alignment peak (21.9 ps) as functions of focus position (and as functions of aligning laser intensity) are shown in Fig. 1(b).

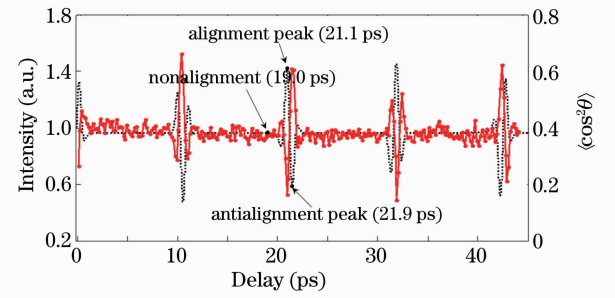

Fig. 2. Experimentally measured the 23rd harmonic intensity from the aligned CO_2 molecules (dotted red curve, left axis) and the calculated alignment parameter $\langle \cos^2 \theta \rangle$ (dashed black curve, right axis) as functions of pump-probe delay. The focus position is $Z = -9$ mm, the aligning laser intensity is approximately 7×10^{13} W/cm^2, and the driving laser intensity is about 2.4×10^{14} W/cm^2.

Figure 3 shows the HHG spectra from the aligned CO_2 molecules at different focus positions; the spectra are recorded at the non-alignment moment (19.0 ps). HHG intensity initially increases and then decreases from $Z = -11$ to -5 mm, before slightly increasing and then decreasing from $Z = -5$ to 0 mm. The spectral shapes before and after $Z = -5$ mm significantly differ; the former are of broadband type and the latter are of narrowband type. Shining filamentation is observed in the interaction region at the observation window of $Z = -5 - 0$ mm, and weak fluorescence is observed from $Z = -11$ to 5 mm.

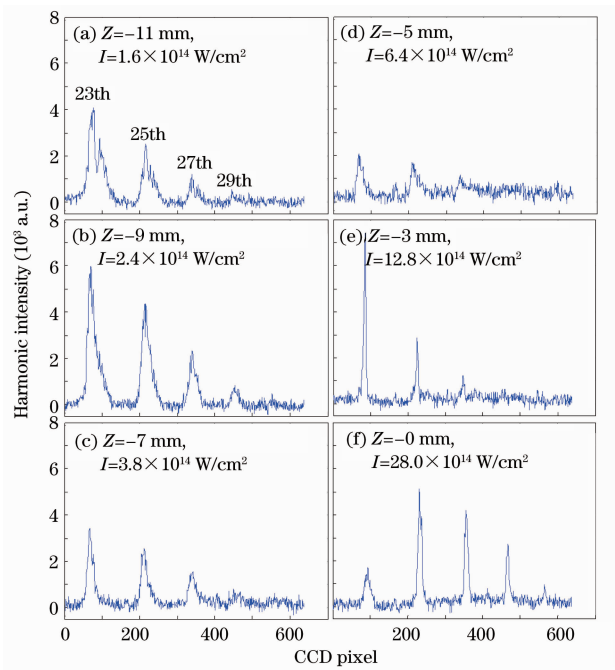

Fig. 3. HHG spectra from the aligned CO_2 molecules at different focus positions. $Z < 0$ mm means that the gas jet is located before the focus position.

Figure 4(a) shows the integral intensity of the 23rd harmonic spectral at the non-alignment moment (19.0 ps), alignment peak (21.1 ps), and anti-alignment peak (21.9 ps) as functions of focus

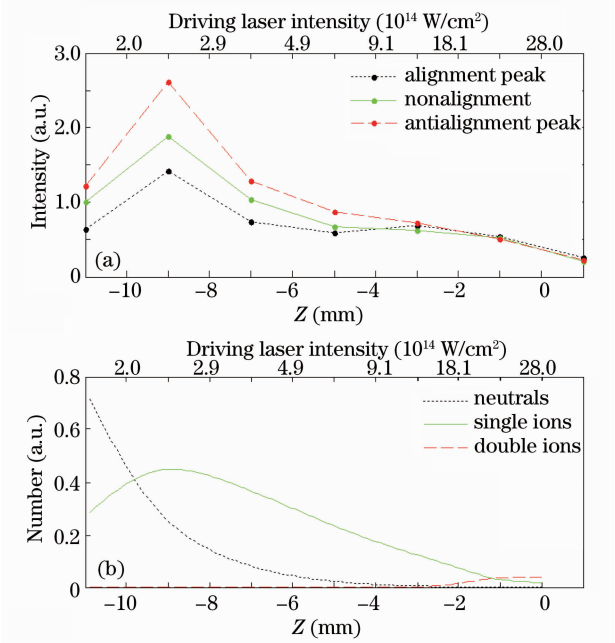

Fig. 4. (a) Integral intensity of the 23rd harmonic spectral at the non-alignment moment (19.0 ps), alignment peak (21.1 ps), and anti-alignment peak (21.9 ps) functions of focus position. The top axis corresponds to the driving laser intensity; (b) neutrals ions and ions in the interaction region (as functions of focus position) simulated using a simple ionization model.

position. The contrast of the alignment signal initially increases and then decreases from $Z=-11$ to -5 mm, and finally disappears from -5 to 0 mm. The results are consistent with the law of spectral variation depicted in Fig. 3. That is, the broadband signal from -11 to -5 mm contains alignment information, while the narrowband signal from -5 to 0 mm does not contain alignment information. Such phenomena can be attributed to the laser field distortion of the molecular highest occupied molecular orbital (HOMO)[12] and multiple HOMO effects[2, 13-15]. The aforementioned effects can destroy the alignment signal by enhancing the intensity of the driving laser. We introduce a simple ionization model[8,16] to explain our experimental results. This model relies only on the molecular ionization potential. In numerically calculating the ionization rate, we use the Ammosov-Delone-Krainov and Keldysh theories. Our simulation results, in which the interaction region of the Gaussian beam has been considered, are shown in Fig. 4(b). Using the first (13.77 eV) and second (37.2 eV) ionization potentials of the CO_2 molecules, we obtain the number of neutrals, single ions, and double ions in the interaction region at different focus positions. The laser intensity that corresponds to the threshold of the first ionization is 8.95×10^{13} W/cm^2, and the laser intensity that corresponds to the 2nd ionization is 1.2×10^{15} W/cm^2.

From $Z=-11$ to -5 mm, therefore, single ionization is the dominant process; the harmonic signal is generated from the outermost HOMO; the alignment signal is clearly observable. From $Z=-5$ to 0 mm, double ionization gradually occurs, and the harmonic signal is generated from multiple HOMOs, causing the alignment signal to worsen and disappear. An optimal focus position ($Z=-9$ mm in our experiment) exists for the maximal signal because of the maximal single ionization rate. When the optimal focus position is close to $Z=-5$ mm, the intensity of the driving laser will be too large to deplete all neutral ions and produce double ions. Numerous electrons and ions then form plasma through filamentation. Plasma can destroy the rotational wavepacket, causing the alignment signal to disappear. The slight enhancement in HHG intensity after $Z=-5$ mm, can be attributed to the formation of double ions. Filamentation phenomena and spectral variety can support the aforementioned observations. The laser beam shape (or the laser intensity) before and after the focus position ($Z<0$ mm and $Z>0$ mm) are almost invariably symmetrical; the experimental results also show that the harmonic shape and molecular alignment are almost invariably symmetrical. Thus, only one side ($Z<0$ mm) of the focus position is discussed in this letter.

4 Conclusion

The experiment and simulation demonstrate that focus conditions are key in molecular field-free alignment. The spectral shape and alignment signal of HHG from aligned CO_2 molecules significantly vary under changing focus position. A maximal alignment signal is achieved at a given focus position because of the optimal driving laser intensity, which is related to the ionization potential of the molecules. These results indicate that an optimal focus condition provides the best alignment signal for practical applications.

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