Characteristics of High-order Harmonic Generation in Carbon Dioxide Gas Jet

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Angular characteristics of high-order harmonic generation from aligned CO₂ molecules have been investigated. The modulation inversion of harmonic yield with respect to molecular alignment can be altered dramatically by fine tuning the intensity of driving laser pulse for harmonic generation. The result is beyond the two-center quantum interference model and can be modeled by employing the strong field approximation including ground state depletion effect. Laser intensity is found to be a new parameter to control the high harmonic emission from aligned molecules.

Keywords: High-order harmonic generation, Alignment, Molecule, Laser intensity.

1. Introduction

High-order harmonic generation (HHG) from atoms and molecules is proven to be an effective means of producing ultrafast coherent extreme ultraviolet (XUV) radiation and even attosecond pulses.  In the strong field limit, HHG is well understood by a three step model: electrons tunnelling through potential barriers are accelerated by intense laser fields, and finally they recombine with parent ions to emit high energy photons. The fast moving electrons in HHG process provides opportunities to probe electronic and nuclear dynamics of atoms and molecules with unprecedented spatial and temporal resolution, e.g. recent experimental studies succeeded in not only taking tomographic images of the highest occupied molecular orbital (HOMO) of nitrogen molecule but also probing proton motions of hydrogen and methane molecules in sub-femtosecond time resolution. On the other hand, owing to the additional degrees of freedom, i.e. rotational and vibrational motions, molecules offer extra parameters as useful tools to control the HHG characteristics. It has been shown by J. Itatani et al. that HHG from impulsively aligned molecules (O₂, N₂) is dependant on the angle between the aligned molecular axis and the polarization of driving laser field, e.g. the harmonics generated from O₂ are enhanced when aligned near 45° with respect to the laser polarization due to the π symmetry of its HOMO, while for N₂ (with σg symmetry of HOMO), harmonic emission is the strongest when aligned along the polarization direction. This angle dependence of harmonics with respect to molecular alignment offers a means of shaping HHG and attosecond pulses.

HHG from CO₂ represents an interesting new feature of molecular alignment dependence. As shown by Kanai et al., the harmonic yields for the 19th - 29th orders from aligned CO₂ molecules exhibit inverted modulation versus the field ionization and alignment parameter . That is to say, when the ionization yield of aligned molecules is maximum / minimum, the harmonic emission intensity is minimum / maximum. This modulation inversion was attributed to the interference of recombination electrons from the two oxygen atoms in CO₂ molecule. C. Vozi reported further that the two center interference effect can be controlled by changing the ellipticity of the driving laser pulse. However, they showed that the inversion of harmonic signal appears for the 21st - 39th harmonic orders, different from the observation by Kanai et al. This is surprising since the interference should result in the harmonic yield inversion for the same harmonic orders according to the two-center interference model. By taking into account the ground state depletion effect on HHG, Anh-Thu Le et al. recently proposed theoretically that the harmonic yield inversion can be varied by changing the driving laser intensity. Therefore experimental verification of this model and further investigation of angular properties of HHG from aligned molecules are strongly desired.

In order to explore the underlying physics in the angular dependence of high harmonic emission from aligned molecules, we investigate the HHG from aligned CO₂ molecules as a function of the driving laser intensity in this work. We observed that by changing the laser intensity, the modulation of harmonic yield can be varied and even inverted with respect to molecular alignment. The two-center interference model has limitations in explaining the modulation inversion of different harmonic orders and the observed laser intensity dependence. Our experimental results are compared with the numerical calculations employing the strong field approximation model including ground state depletion effect. The role of laser intensity as a new parameter to control the angular dependence of HHG in aligned molecules is revealed.

2. Experiments

Our experiments were carried out by using a Ti:sapphire based chirped pulse amplification laser system (Spectral-Physics, TSA-25), which provides laser pulses of 50 fs duration and pulse energy of ~ 10 mJ at 800 nm. The output laser pulses were split into two beams, one used as the pump...
3. Results and Discussion

3.1 Non-adiabatic alignment of CO₂

The experimentally measured the 23rd harmonic intensity as a function of pump-probe delay time is represented in Fig. 1 (solid line, left axis). The probe laser intensity was 2.4 × 10¹⁴ W/cm². Our calculated molecular alignment parameter <cos²θ> is also plotted in Fig. 1 (dashed line, right axis). In our calculation the initial rotational temperature was taken to be 80 K and the time evolution of the wavepacket is evaluated through solving the time-dependent Schrödinger equation (TDSE). It is evident that the modulation of harmonic signal is reversely matched with that of the molecular alignment parameters <cos²θ> exactly. This is consistent with the experimental results of Kanai et al. where the harmonic signal reversely matches with the alignment parameter and ionization yield. The phenomena were regarded as the evidence of interference of the recombining electrons originated from two oxygen atoms. For CO₂, the constructive interference happens when Rcosθ = (n−1/2)λ, and the destructive interference happens when Rcosθ = nλ, where R is the distance of two oxygen atoms in CO₂ and λ is the deBroglie wavelength of recombining electrons.

3.2 Intensity dependence of high harmonic emission

Our further measurement of the alignment dependent harmonic emission for the 25th orders shows counterintuitive results. In Figs. 2(a)-2(d), we show the 25th harmonic yield at around half revival of molecular alignment under four different probe laser intensities: 1.6 × 10¹⁴ W/cm², 1.9 × 10¹⁴ W/cm², 2.1 × 10¹⁴ W/cm², and 2.4 × 10¹⁴ W/cm². Also shown is the calculated alignment parameter <cos²θ> (dashed line).

![Fig. 2 Time evolution of the 25th order harmonic intensity from aligned CO₂ at around half revival for the probe laser intensities of (a) 1.6 × 10¹⁴ W/cm², (b) 1.9 × 10¹⁴ W/cm², (c) 2.1 × 10¹⁴ W/cm², and (d) 2.4 × 10¹⁴ W/cm². Also shown is the calculated alignment parameter <cos²θ> (dashed line).](image-url)
results 9,10 - as the laser intensity increases from $1.6 \times 10^{14}$ W/cm$^2$ to $2.4 \times 10^{14}$ W/cm$^2$, the temporal evolution of the 25th order harmonic becomes inverted with respect to molecular alignment parameter $\cos^2 \theta$.

3.3 Angular dependence of high harmonic generation

In order to understand further our experimental results, we performed numerical simulation using an extended Lewenstein’s strong field approximation model 12 including the ground state ionization effect. In this model, the harmonic spectrum from molecular dipole moment in the time domain is calculated by using

$$x(t) = \int_{-\infty}^{\infty} d\tau (\frac{\pi}{i\omega})^{1/2} d^* [p_{\omega} - A(t)]e^{-i\omega(\tau - \tau_c)}$$

$$\times \mathbf{E}(t - \tau) \cdot d[p_{\omega} - A(t - \tau)]a^*(t)a(t) + \text{c.c.}. \quad (1)$$

where $d[p_{\omega} - A(t - \tau)]$ are the transition dipole moments between the ground state and the continuum state, $p_{\omega}$ is the canonical momentum at the stationary points with vector potential $A$ and denoted as

$$p_{\omega} = \int_{\theta_0} A(t') dt' \tau' .$$

The exponential term takes account of the action of the electron trajectories within the laser field. We consider the depletion of ground state molecules by a factor of $a(t)$, which is approximated by 9

$$a(t) = \exp[-\int_{-\infty}^t dW(t')/2],$$

and $W(t')$ is the tunneling ionization rate obtained by MO-ADK theory. 13)

To calculate the transition dipole moments the ground state ionization effect is approximated by atomic $2p_y$ orbital $\Phi_{2p_y}$ as 14

$$\Psi_{t_0}(x) = C[\Phi_{2p_y}(x + R/2) - \Phi_{2p_y}(x - R/2)], \quad (2)$$

where $R$ is the distance between oxygen atoms, and the time dependent dipole transition moment is given by

$$d_{t_0}(p) = (2i/\sin\mathbf{p} \cdot \mathbf{R}/2)d_{t_0, y}(p) \cos(\mathbf{p} \cdot \mathbf{R}/2)\tilde{\phi}_{2p_y}(p) \mathbf{R}. \quad (3)$$

in which the $d_{t_0, y}(p)$ is the atomic dipole moment from the $2p_y$ orbital and $\tilde{\phi}_{2p_y}(p)$ is the $2p_y$ wavefunction in the momentum space. Finally the harmonic spectra $S(\theta)$ as a function of the angle $\theta$ between molecular axis and laser polarization are obtained through the Fourier transform of the transition dipole moment.

In order to calculate the harmonics intensities from aligned CO$_2$ molecules, the time-dependent alignment distribution of molecules has to be taken into account. At field-free evolving time $t$, the distribution of aligned molecular axis is expressed as $P(\theta, t) = \left| \psi(t) \right|^2$, in which $\psi(t)$ is obtained by solving the TDSE. The harmonic intensities $f(\theta)$ are given by integrating the product of $S(\theta)$ and $P(\theta, t)$.

$$f(\theta) = C \int S(\theta)P(\theta, t) \sin \theta d\theta . \quad (4)$$

We first calculate the angular distribution $S(\theta)$ of the 25th harmonics from Eq. (1) at the laser field of $2.4 \times 10^{14}$ W/cm$^2$. As shown in Fig. 3(a) (solid line), the $S(\theta)$ is peaked at 67$^\circ$. As a comparison, the $S(\theta)$ at the laser field of $1.6 \times 10^{14}$ W/cm$^2$ is also plot in Fig. 3a (dashed line). It is evident that with the laser field decreased to $1.6 \times 10^{14}$ W/cm$^2$, the peak of $S(\theta)$ shift to 55$^\circ$. The molecular alignment distributions $P(\theta, t)$ at $f(t) = 21.1$ ps and $P(\theta, t = 21.9$ ps) are then obtained from $\psi(t)$ by solving the TDSE using the experimental conditions, and the calculated $P(\theta, t)$ at the intensity of $2.1 \times 10^{13}$ W/cm$^2$ are also plot in Fig. 3b. CO$_2$ molecules are mostly aligned along the laser polarization direction ($\theta = 0^\circ$) at 21.1 ps and perpendicular to laser polarization ($\theta = 90^\circ$) at 21.9 ps, as reported previously. 9) From Eq. (4) the harmonic emissions of $f(t) = 21.1$ ps and $f(t = 21.9$ ps) can then be obtained by integrating the product of $S(\theta)$ and $P(\theta, t)$.

To compare with the experimental results, we calculate the
harmonic emissions of \( f(t = 21.1 \text{ ps}) \) and \( f(t = 21.9 \text{ ps}) \) at the laser field from \( 1.2 \times 10^{14} \text{ W/cm}^2 \) to \( 2.4 \times 10^{14} \text{ W/cm}^2 \). As shown in Fig. 4a, the normalized HHG intensities from aligned molecules \( (t = 21.1 \text{ ps}) \) and antialigned molecules \( (t = 21.9 \text{ ps}) \) reversed at laser intensity of \( 1.4 \times 10^{14} \text{ W/cm}^2 \). The calculated results reproduce well the experimental results shown in Fig. 2, except a small difference in the absolute value of laser intensity where the inversion starts to occur. A more rigid model for molecular HHG in CO\(_2\) may help to reduce this quantitative difference between the experimental and calculated results.

In order to clarify the effect of ground state depletion, we also calculate the harmonic intensities \( f(t = 21.1 \text{ ps}) \) and \( f(t = 21.9 \text{ ps}) \) at the laser fields without the depletion factor of \( a(t) \), and the result is shown in Fig. 4b. Clearly the calculation does not reproduce experimental observation of the inversion of 25th harmonic intensity, which indicates the necessity of including the ground depletion effect into the numerical model.

From Eq. (4) we know that to control the angular distribution of HHG one could also adjust the molecular alignment distribution \( P(\theta) \). We would like to mention that the alignment distribution of \( P(\theta) \) is not dependent on the laser intensity, which only modify the contrast of harmonic signal of aligned molecules to that of isotropic molecules. The recent progress in 3D alignment of molecules \(^{15}\) is expected to play a role in controlling HHG. Experiments on higher order harmonics with finely adjusted driving laser intensity may lead to the more comprehensive understanding of angular dependence of harmonics and therefore better control of HHG.

4. Conclusion

In conclusion, we experimentally demonstrate that the harmonic emission from aligned CO\(_2\) molecules as a function of alignment angle can be altered dramatically by fine tuning the intensity of driving laser pulses. The previously proposed simple two-center quantum interference model cannot account for the laser intensity dependence of harmonic yield inversion with respect to molecular alignment and the inversion for different harmonic orders in different experiments. The experimental results can be modeled by the strong field approximation model including ground state depletion effect. Laser intensity is proved to be a new parameter to control the high harmonic emission from aligned molecules.

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