Angular distribution of high-order harmonic generation from aligned molecules

Peng Liu [1], Zhinan Zeng, Pengfei Yu, Ruxin Li [2], Zhizhan Xu

State Key Laboratory of High Field Laser Physics, Shanghai Institute of Optics and Fine Mechanics, Chinese Academy of Sciences, Shanghai 201800, China

Synopsis: We experimentally investigate the high-order harmonic generation (HHG) from aligned CO_2 molecules and demonstrate that the modulation inversion of harmonic yield with respect to molecular alignment can be altered dramatically by the intensity of the driving laser pulse. The laser field dependent inversion can be explained by the shift angular distribution of harmonic emission calculated with the strong field approximation (SFA) model including a ground state depletion factor. The observed angular distributions of harmonics are discussed according to the available theoretical models.

The HHG from aligned molecules attract intensive studies due to its potential applications in the imaging of molecular orbital and controlling of the harmonic generation. The HHG from CO_2 represents an interesting feature of molecular alignment dependence - quantum interference effect. The two-center interference model has the limit in explaining the variation of inverted harmonic orders in different reports [1,2]. By taking into account the ground state depletion effect on HHG, A.-T. Le et al. [3,4] proposed theoretically that the harmonic yield inversion can be varied by changing driving laser intensity. The experimental verification of this model is strongly desired.

Our result [5,6] indicates that as the driving laser field increases from 1.6×10^{14} W/cm^2 to 2.4×10^{14} W/cm^2, the 25th order harmonic intensity evolution becomes inverted with the molecular alignment modulation (See Fig. 1, in which the dashed line is the alignment degree as a function of delay time, and the dot + solid line is the harmonic intensities of aligned molecules at the delay times). This is the direct confirmation of the laser field dependence of the HHG angular distribution.

Using the extended Lewenstein's strong field approximation (SFA) model, the harmonic emissions are calculated for the angles θ between the molecular axis and the laser polarization direction. By taking into account the alignment distribution of molecules, the harmonic intensities of the aligned and anti-aligned molecules are calculated, showing that the inversion of harmonic emission happens at the laser field of 1.4×10^{14} W/cm^2, which is close to the observed 1.6×10^{14} W/cm^2. The calculation by neglecting the ground ionization factor indicates no harmonic intensity inversion in the laser field range discussed, showing that the ground state depletion influences the harmonic emissions.

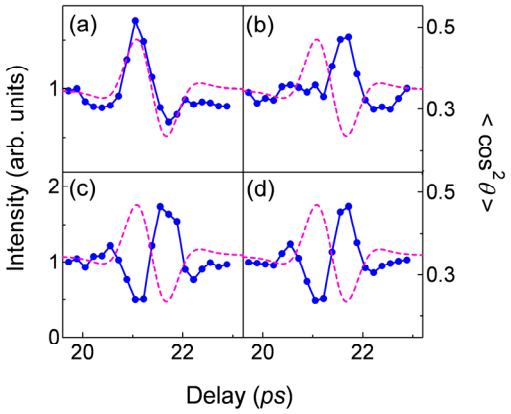

Fig. 1. Time evolution of the 25th order harmonic intensity from aligned CO_2 at around half revival for the laser intensities of (a) 1.6×10^{14} W/cm^2, (b) 1.9×10^{14} W/cm^2, (c) 2.1×10^{14} W/cm^2, and (d) 2.4×10^{14} W/cm^2.

Angular dependence of the harmonics is also investigated experimentally by adjusting the angle of aligning laser polarization and HHG driving laser polarization. The results are compared with the available theoretical models, and the applicability of SFA model on larger molecules is discussed.

References

[1] T. Kanai, et al., Nature 435, 470 (2005).
[2] C. Vozzi, et al., PRL 95, 153902 (2005).
[3] A.-T. Le, et al., PRA 73, 041402 (2006).
[4] A.-T. Le, et al., Journal of Modern Optics 54, 967 (2007).
[5] P. Liu, et al., PRA 78, 015802 (2008).
[6] P. Yu, et al., PRA 79, 053814 (2009).

[1] E-mail: peng@siom.ac.cn

[2] E-mail: ruxinli@mail.shcnc.ac.cn