

Abstract Submitted
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Multiple-orbital effects on high-harmonic generation of aligned CO₂ molecules¹ CHENG JIN, ANH-THU LE, C.D. LIN, Physics Department, Kansas State University — We calculated the modulated high-harmonic generation (HHG) spectra of transiently aligned CO₂ molecules in an intense infrared laser field. The effect of interference between HOMO and HOMO-2 orbitals and the effect of macroscopic propagation of the harmonics in the gas medium are both included. The calculated results show that the minimum in the HHG spectra shifts with laser intensity which is consistent with experimental measurements. The exact position of the minimum, however, is easily influenced by the degree of molecular alignment and, through macroscopic propagation, by other detailed experimental conditions. From the HHG spectra the relative ionization probability between HOMO and HOMO-2 in different intensity region can also be estimated. In contrast to N₂ molecules, HOMO interferes with HOMO-2 only at small pump-probe angles for CO₂ molecules, and HOMO is dominant at large pump-probe angles. This study provides the needed theoretical basis for understanding how the HHG spectra can be influenced by the molecular structure.

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