

Angle dependence of molecular high harmonic emission in three dimensions

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An important goal in laser-matter science is to obtain structure-dependent information about laser-induced strong field processes at the molecular level. Lower symmetry asymmetric top systems comprise the widest and most general class of molecules, but are difficult to explore in strong field physics. This gap is particularly evident in HHG. The dependence of HHG on molecular orientation in simple linear molecules shows how different molecular orbitals participate in this highly nonlinear process [1, 2]. This provides strong motivation for extending these studies to asymmetric tops. Unfortunately, the technical difficulties of molecular orientation or molecular alignment have hampered attempts to study strong field interactions in more complicated molecules. Here we demonstrate a method to extract axis-dependent information from asymmetric tops and apply it to the sulfur dioxide (SO₂) molecule. Furthermore, we find the contributions of lower-lying orbitals to each harmonic, and find strong contributions from the ionization part of the HHG process.

The richest information is obtained from molecular sources that have been aligned in the laboratory, either through static alignment via adiabatic means or by impulsive transient stimulated rotational Raman scattering, which produces periodic transient alignment. Transient impulsive alignment works well for linear molecules with only one relevant polarizability axis. But for asymmetric top molecules, which have three different moments of inertia and a complicated polarizability tensor, a linearly polarized alignment pulse leaves the molecules in free rotation about multiple axes, with up to five different revival constants. To obtain the axis-dependent amplitude and phase of harmonic emission in the molecular frame we use a technique that is conceptually related to picosecond coherence spectroscopy. We fit single-axis alignment patterns to our data to decompose the HHG signal [3]. As depicted in the lower portion of Fig. 1, our analysis allows us to reconstruct the harmonic emissions along each axis of the molecule by decomposing each harmonic revival pattern into three components, one per axis, with complex coefficients that reveal the relative amplitude and phase of harmonic emission from each axis.

Fig. 1 shows the experimental HHG spectrum for all harmonics with a dramatic inversion around 12 ps and 25-29 ps. Using calculations of field ionization and the recombination dipole, we show that the inversion is caused by contributions from multiple orbitals with a strong influence from the ionization process. Our results have implications for HHG-based orbital tomography, because in principle this technique could be used to examine the structure or dynamics of molecules of arbitrary complexity.

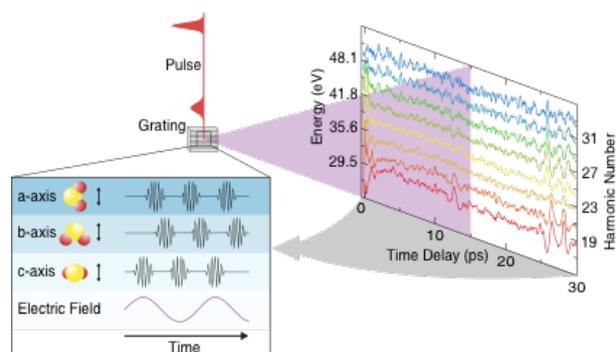


Figure 1: *The alignment decomposition method. The data show an inversion around 12 and 27 ps.*

References

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