

Consistent theoretical check of the three-step model of high-harmonic generation for sulfur dioxide SO₂

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One of the goals in strong-field physics is to achieve a real-time imaging of the electronic structure of molecular systems and thereby gain the ability to “watch” dynamical phenomena such as chemical reactions in real time. The proposed time-resolved imaging schemes that are based on linear-polarized ultrashort laser pulses can be roughly divided into two categories, direct imaging and rescattering based. The first category relies on the strong angular dependence of the ionization on the polarization axis of the incident electromagnetic field. This dependence reflects the shape of the outer part of the electronic orbitals [1, 2].

The rescattering-based concepts study phenomena that take place when the ejected electron recollides with the parent ion. One such observable is the high-harmonic generation (HHG) spectrum that has been used for orbital imaging [3] or for monitoring the nuclear motion in molecules such as water [4]. The three-step model [5] is by far the most popular way for calculating HHG spectra. The essence of the model is to approximate the HHG amplitude by a product of three independent amplitudes (therefore three-step-model): the ionization amplitude, the amplitude describing the propagation of the unbound part of the electronic wavepacket in the laser field (ignoring the interaction with the remaining ion), and the recombination amplitude with the parent ion. Clearly, in view of the popularity of the three-step model it is important to investigate its validity. We note that such a check by comparing the outcome of a HHG experiment with the prediction of the three-step model is often limited by the number of approximations that necessarily have to be done in the calculation of the different amplitudes for systems more complicated than one- or two-electron atoms. Already for a simple molecule, nuclear motion (both rotations and vibrations) as well as many-electron effects would have to be considered. Furthermore, experimental aspects like uncertainties in the pulse shape and focal-volume effects cannot be ignored. The omission of these effects may severely bias the conclusions from such a comparison.

Motivated by a recent and on-going experiment on SO₂ [6], we have chosen this molecule as a test case for a consistent check of the three-step model for HHG. For this purpose, the HHG spectra for a number of orbitals and orientations are calculated with our recently developed solver of the time-dependent Schrödinger equation (TDSE) within a gauge-invariant single-active-electron approximation [1]. Furthermore, also the orientation-dependent ion yields are extracted from this TDSE solution. Using the identical electronic-structure model and basis set, the recombination amplitudes are also calculated. This allows for the alternative calculation of the HHG spectra based on the three-step model. Evidently, in this way a very consistent check of the three-step model is achieved.

References

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