Probing Fixed-in-Space Molecular Structures With High Harmonic Generation[1]

Anh-Thu Le

Department of Physics, Kansas State University, Manhattan, KS 66506, USA

The recently developed quantitative rescattering theory (QRS) gives a simple and accurate description for high-order harmonic generation (HHG) from atoms or aligned molecules. According to the QRS, HHG spectra can be expressed as a product of a returning electron wave packet and the photo-recombination differential cross section of the *laser-free* continuum electron back to the initial bound state. This factorization opens possibilities for a relatively simple method for retrieval of the molecular frame photoelectron angular distributions (MFPAD) and the phase of the transition dipole with HHG from aligned molecules. Using the QRS combined with accurate photoionization transition dipole moments for fixed-in-space molecules obtained from state-of-the-art molecular photoionization calculations, we show that both the magnitude and phase of the high-order harmonics observed in recent experiments on aligned CO_2, O_2 and N_2 can be quantitatively reproduced. Furthermore, we show that the contribution from multiple molecular orbitals, the polarization state and ellipticity of the harmonics can be all studied quantitatively for aligned molecules.

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