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Ab initio study of the ellipticity of molecular high harmonic generation driven by linearly polarized laser fields¹ SANG-KIL SON, University of Kansas, DMITRY A. TELNOV, St. Petersburg State University, SHIH-I CHU, University of Kansas — A recent experiment has demonstrated that high-order harmonic generation (HHG) from aligned linear molecules can be elliptically polarized even driven by linearly polarized laser fields Phys. Rev. Lett. 102, 073902 (2009)]. We perform fully *ab initio* calculations of HHG from the ground and excited electronic states of H_2^+ with arbitrary orientation and detailed analyses for the polarization and phases of harmonic emissions to reveal theoretical origins of the ellipticity of molecular HHG. Our results predict that even for the one-electron system all harmonic emissions are elliptically polarized unless molecular alignment is parallel or perpendicular to the polarization of the driving laser field. The ellipticity of harmonic emissions is closely related to the symmetry of the molecular orbital and affected by two-center interference effects in HHG. For H_2^+ , the ellipticity becomes large for the ground state that is approximated by a symmetric combination of the atomic orbitals, whereas it becomes small for the first excited state approximated by an antisymmetric combination. This observation can be generalized for the ellipticity of HHG from linear molecules.

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