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Probing time-resolved large-amplitude molecular vibrations with high-order harmonics generated by ultrashort laser pulses[1]

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We present a theory that incorporates the vibrational degrees of freedom in high-order harmonic generation (HHG) of molecules with intense infrared lasers [1]. This theory extends the previously developed quantitative rescattering theory (QRS) for HHG from fixed-nuclei molecules, by accounting for the effect of lasers on the vibrational wavefunctions. The induced time-dependent transition dipoles for each fixed nuclear geometry are added up coherently, weighted by the laser-driven time-dependent nuclear wave packet distribution. We show that the nuclear wave packet can be strongly modified by the driving laser. The validity of this model is first checked against results from the solution of the time-dependent Schrödinger equation for a model system. The theory is then applied to explain time-resolved HHG spectra for molecules measured in pump-probe experiments.

Work done in collaboration with Anh-Thu Le, Kansas State University; Robert Lucchese, Texas A&M University; and Toru Morishita, University of Electro-communications, Japan.

[1] Anh-Thu Le, Toru Morishita, R. R. Lucchese and C. D. Lin, Phys. Rev. Lett. **109**, 203004 (2012).

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