"High Harmonic Generation from Aligned Molecules as a Route to Molecular Continua"

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High harmonic generation (HHG) is a fascinating process, wherein ionization takes place close to the maximum of the electric field, generating a free-electron wave packet in the continuum that follows the electric field oscillations. If the field is linearly (or close to linearly) polarized, the electron will revisit the core, with a certain probability of recombining while emitting high harmonics of the incident radiation. Nonadiabatic alignment by moderately intense laser pulses is a general technique of establishing field-free (post pulse) orientational order and a controllable refractive index in molecular media with a growing variety of applications. The combination of HHG with nonadiabatic alignment was shown to provide information about the electronic structure of the bound molecule.

We derive a rigorous framework for calculation of high harmonic signals from aligned molecules that accounts for both the electronic and the rotational wave packet dynamics as well as for their correlation. The former was intensively discussed in the recent literature whereas the latter is new and fascinating. By providing a closed form expression for the time dependence of the signal, the formalism explains the observations of a large number of experiments. Interestingly, it points to the information content of harmonic intensities regarding the rotational coherences of the target molecule and illustrates the relation between the harmonics polarization and the phase of the underlying continuum electronic wavefucntion. Finally, we fit high accuracy experimental harmonics to our formal harmonics to extract new insights regarding the continuum electronic dynamics.