Information from Optical Properties of High-order Harmonic Signals

Paul A. J. Sherratt and Tamar Seideman *

Department of Chemistry, Northwestern University,
2145 Sheridan Rd, Evanston, IL 602018-3113, USA
(Dated: December 27, 2011)

Abstract

High-order harmonic generation (HHG) is a highly non-linear process in which a medium in an intense laser field emits coherent radiation at integer multiples of the driving frequency. It provides a sensitive probe at attosecond and sub-Ångström resolution of the underlying electronic structure and molecular dynamics.

We address theoretically and numerically the possibility of birefringence, observing ellipticity in HHG from aligned molecules driven by linearly polarized fields, a subject of controversy in the recent literature with significant implications. We illustrate how the phase of the continuum electronic wavefunction, and hence the underlying molecular potential, is responsible, at least in part, for the ellipticity observed in harmonic spectra. We find that the emission of elliptically polarized harmonics is a general phenomenon, yet qualitatively determined by the molecular properties. The sensitivity of the ellipticity to the parameters in our model invites the use of ellipticity measurements as a probe of both the continuum wavefunction and the underlying molecular potential.

Studies of the possibility of molecular dichroism from HHG driven by elliptically polarized fields is in its infancy. We show early experimental and theoretical results highlighting further important information contained within the harmonic signal concerning both geometrical and interference effects and how they manifest themselves as dichroism in the HHG signal.

*p-sherratt@northwestern.edu