

High-Harmonic Generation in Polyatomic Molecules

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Synopsis: We investigate signatures of suppressed ionization by observing high-harmonic generation in polyatomic chlorocarbon molecules CH_2Cl_2 , CHCl_3 and CCl_4 using 800nm infrared light. An extension of the high-harmonic cut-off in all molecules was observed compared to atomic Xe which has a similar ionization potential. Ellipticity measurements show broader electron wave-packet spreading for highly polarizable molecules, demonstrating the influence of multi-electron dynamics in the high-harmonic generation process.

Strong field laser interactions with atoms and molecules lead to incredible nonlinear processes such as above threshold ionization, laser induced electron diffraction, and high harmonic generation (HHG). HHG has quickly become the quintessential method to push the field of ultrafast science into the attosecond domain[1]. The semi-classical model for HHG accurately describes the underlying physics behind this process for a simple atom but the influence of multi-electron dynamics in a polyatomic molecule can lead to results unexplainable by the atomic model.

In this paper we report on experimental results demonstrating suppressed ionization in polyatomic molecules and the influence of polarizability on the HHG process.

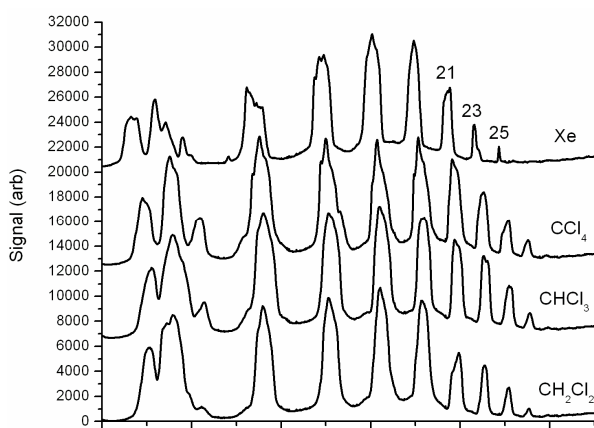


Fig. 1 Harmonic spectra of Xe and chlorocarbons

Signatures of suppressed ionization in HHG in linear di- and tri-atomic molecules have been demonstrated by comparison of extended cut-offs with their partner atoms having the same ionization potentials (IP)[2]. We choose three chlorocarbon molecules, CH_2Cl_2 , CHCl_3 , and CCl_4 because (a) they have similar IP's (11.33, 11.37, and 11.47 eV respectively), (b) they all possess three-dimensional tetrahedral geometry, (c) their highest occupied molecular orbitals (HOMO) exhibit similar symmetries but also

contain varying degrees of complexity, and (d) they have different electron polarizabilities (6.5, 8.5 and 10.5 \AA^3 respectively). **Fig. 1** shows the harmonic spectra for Xe and the chlorocarbons at an intensity of $5 \times 10^{14} \text{ W/cm}^2$. The harmonic cut-off is extended in the chlorocarbons compared to Xe (IP = 12.13 eV) indicating the role of suppressed ionization. Saturation intensity measurements for the chlorocarbons along with their observed HHG spectra and HOMO symmetries indicate that destructive interference is greater in CCl_4 than the others.

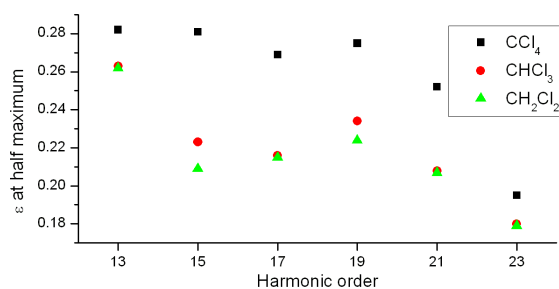


Fig. 2 Ellip. at half-maximum signal for each harmonics

Ellipticity dependence of HHG provides information on the electron wave-packet spreading. Our results show that the half-widths at half-maximum for all harmonics in CCl_4 are consistently larger (see **Fig. 2**). The electron wave-packet spreading depends only on the ionization potential and, in large molecules, on the polarizability [3,4]. Since the IP of the molecules under consideration were similar, this is the clear evidence of the role of multi-electron dynamics in HHG in larger molecules.

We expect that experiments using longer wavelength light will provide new insight into the multi-electron influence on HHG in polyatomic molecules.

References

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