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Theory of Dynamic Chemical Imaging with infrared laser Pulses¹

CHII-DONG LIN, Kansas State University

When an infrared laser pulse interacts with atoms or molecules, electrons which are released from the target earlier may be driven back by the laser field to recollide with the target ions. These recollisions incur the well-known strong field phenomena of high-order harmonic generation (HHG) as well as high-energy above-threshold ionization (HATI) photoelectrons. Since infrared laser pulses with duration of a few femtoseconds are now widely available, it is clear that HHG and HATI spectra can be used to extract the structural change of a transient molecule with temporal resolution of a few femtoseconds in a typical pump-probe setup. Based on the rescattering concept, recently we have established a quantitative rescattering theory (QRS) where the HHG yield can be expressed as the product of a returning electron wave packet with the photo-recombination cross section, and the HATI spectra can be expressed as the product of a similar wave packet with the elastic differential cross sections (DCS) between the target ion with free electrons. The QRS has now been applied to compare with experimental HHG spectra from molecules, from which molecular frame photoionization cross sections and the phases can be extracted. The QRS has also been applied to study the HATI spectra extensively, to retrieve the DCS of electron-ion collisions. Using the extracted DCS for electron energies in the typical range of the returning electrons, we have shown that the target structure can be retrieved. From the returning electron wave packet, we can also obtain the laser parameters, including pulse duration, peak intensity and carrier-envelope-phase, from the measured HATI spectra.

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