Multi-electron effects in Harmonic Generation

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In the last several years, harmonic generation has become the centre-piece of attosecond physics [1] facilitating the generation of ultrashort and high energy laser pulses [2,3]. The process has also unlocked an array of techniques for probing complex electronic structure and dynamics [1,4], and has allowed real time observation of multi-electron effects in ultrafast processes [5,6].

Harmonic generation (HG) is commonly thought of as a single-electron process, and is modelled as such by the widely accepted 'three-step' model- an electron is first ionized, then driven in a laser field, before recolliding with its parent ion. Upon recollision the electron gives up the energy it has absorbed from the laser field in the form of a photon of a harmonic of the driving laser frequency, and is recaptured by the ion. While a great many computational models have been based upon this single-active electron picture, a body of evidence has emerged demonstrating the importance of multi-electron effects in the HG mechanism [5-10]. In order to understand these phenomena it is thus important that high quality theoretical approaches are developed which can describe multi-electron and multichannel (multiple orbital) effects from first principles.

To this end we have developed time-dependent R-matrix theory, and extended the method to account for HG. A study of the harmonic yield from argon in 2×10^{12} Wcm^{-2} laser pulses in the wavelength range of 200-248 nm was used to demonstrate the competition of 3s and 3p electrons in the HG process. This interference leads to a resonance in the fifth harmonic at energies corresponding to the $3s3p^6np$ bound states [9]. A similar effect is observed in HG from singly ionized argon, wherein multi-electron interference leads to an order of magnitude decrease in the harmonic yield at particular energies. The Ar^+ system also demonstrates multichannel interference- the close spacing of the ionization thresholds leads to an interference between residual ion states, and a consequent reduction in the harmonic yields. Interestingly, ionization events leaving the ion in the first excited state, rather than the ground state, are the dominant pathway to HG [10].

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