Dynamical multielectron correlation in high harmonic generation

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Strong-field light-matter interaction has various applications. One prominent example is high harmonic generation (HHG), which serves not only as the standard tool for the generation of ultrashort laser pulses, but also as a promising way for the direct measurement and control of electron dynamics in atoms and molecules. Although the HHG process can be adequately treated as a one-electron problem in many cases, it fails when electron correlation plays a role [1], which is far from fully explored yet and is now attracting increasing interests.

In this contribution, we report a fascinating mechanism leading to a drastic cutoff extension of the HHG spectra, induced by dynamical electron correlation between recolliding and inner-shell core electrons. This mechanism was first predicted for a one-dimensional multielectron model atom [2] and later extended to a one-dimensional two-electron model molecule [3]. Here we numerically simulate HHG from real alkali atoms using the recently developed first-principle three-dimensional time-dependent complete-active-space self-consistent-field (TD-CASSCF) method [4, 5], which enables accurate description of many-electron atoms and molecules for a broad range of laser parameters with reasonable computational cost. Taking lithium atom for example, we have successfully computed HHG spectra in an intense 1200-nm laser pulse, shown in Fig. 1. Convergence of the spectra with respected to the number of orbitals is achieved (red and black curves). In comparison with single-active-electron case (blue curve), the multi-electron spectrum exhibits two remarkable features: (1) a resonance-like peak at around 60th harmonic order with intensity 2 orders of magnitude higher than the plateau and (2) a second plateau with cutoff position at 158th harmonic order, far beyond the first cutoff at 87th harmonic order. These two features, which also exist in the HHG spectrum from other alkali atoms, are confirmed to be clear manifestations of multielectron effects. Therefore, we demonstrate an important aspect of strong field phenomena, i.e, electrons interact not only with lights but also between electrons themselves.

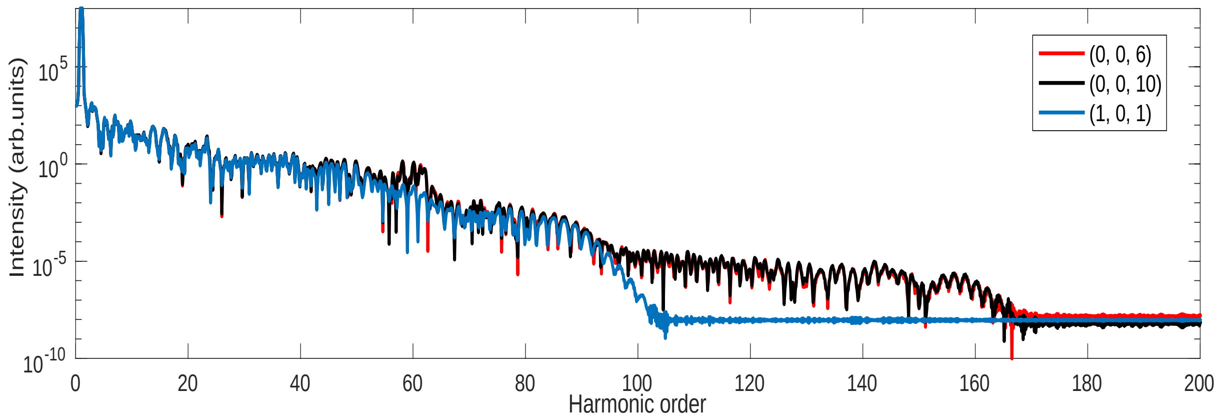

Figure 1: HHG spectra of Li irradiated by a laser pulse with a wavelength of $\boldsymbol{\lambda = 1200\ nm}$, a peak intensity of $\boldsymbol{I = 2 \times 10^{14}}$ W/cm^2 and the foot-to-foot pulse duration of $\boldsymbol{\tau = 8T}$ with $\boldsymbol{T}$ being the optical period of the laser field, obtained with the TD-CASSCF method with various decomposition of orbital spaces. The integers (n_{FC}, n_{DC}, n_A) in the legend specify the numbers of frozen-core, dynamical-core and active orbitals, respectively.

References

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