High-harmonic generation enhanced by dynamical electron correlation

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Synopsis

We theoretically investigate correlated electron dynamics in high-harmonic generation (HHG), using all-electron ab initio simulations for three-dimensional real alkali-metal atoms. The resulting harmonic spectra exhibit a plateau extended beyond the usual cutoff and a prominent resonance peak, which are clear manifestations of dynamical electron correlation.

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.

Here, 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 muticonfiguration time-dependent Hartree-Fock (MCTDHF) method [4-6], which enables accurate description of many-electron atoms and molecules for a broad range of laser parameters. Taking sodium atom for example, we have successfully computed HHG spectra in an intense 1200-nm laser pulse, shown in Fig. 1. In comparison with single-active-electron (SAE) case, the multi-electron spectrum exhibits a second plateau with cutoff position at 133 eV, far beyond the first cutoff at 90 eV. Moreover, a prominent resonance peak shows up at 33 eV. These two remarkable features, which also exist in the HHG spectrum from other alkali atoms, is confirmed to be clear manifestations of multielectron effects. This demonstrates that high-harmonic spectroscopy provides new possibilities to explore dynamical electron correlation in strong laser pulses.

Figure 1. HHG spectra of Na computed by MCTDHF method (blue solid curve) and SAE approximation (red dashed curve). The laser pulse has a wavelength of \( \lambda = 1200\,\text{nm} \), a peak intensity of \( I = 2 \times 10^{14}\,\text{W/cm}^2 \). The vertical dashed line guides the position of resonance peak.

References

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