Multielectron trace of back reaction in high-harmonic generation

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Abstract: We describe back-reaction as a novel correlation mechanism in the two-electron dynamics of helium atoms exposed to intense laser fields. The electron-electron correlation information is encoded as a high-energy secondary plateau in high-harmonic spectroscopy.

Correlation is the main source of complexity in the dynamics of microscopic systems. The many body problem remains as a computational challenge for numerical simulations in laser-mater interactions and hence, most of the theoretical approaches rely on the single-active electron approximation. To disentangle the most fundamental aspects of complex interactions, two-electron systems, such as the helium atom, offer an extraordinary scenario. We report a correlation mechanism based in a back-reaction interaction between both electrons [1]. By solving the one-dimensional time-dependent Schrödinger equation of He, we identify a distinct trace of this mechanism in high-order harmonic generation (HHG).

Among the different correlation mechanisms, back-action refers to a two-way correlation, in which the dynamics of a given physical system affects itself through the intermediation of a coupled second system. In the shake-up photoionization of helium, back-action leads to a measurable time delay using attosecond streaking [2]. However, back-action can give rise to other observable effects, which can be advantageous to track the correlated dynamics behind it. We demonstrate that the back-action signature in high-harmonic spectroscopy is a secondary plateau of high-order harmonics extending the emission towards higher frequencies, beyond the standard cutoff frequency.

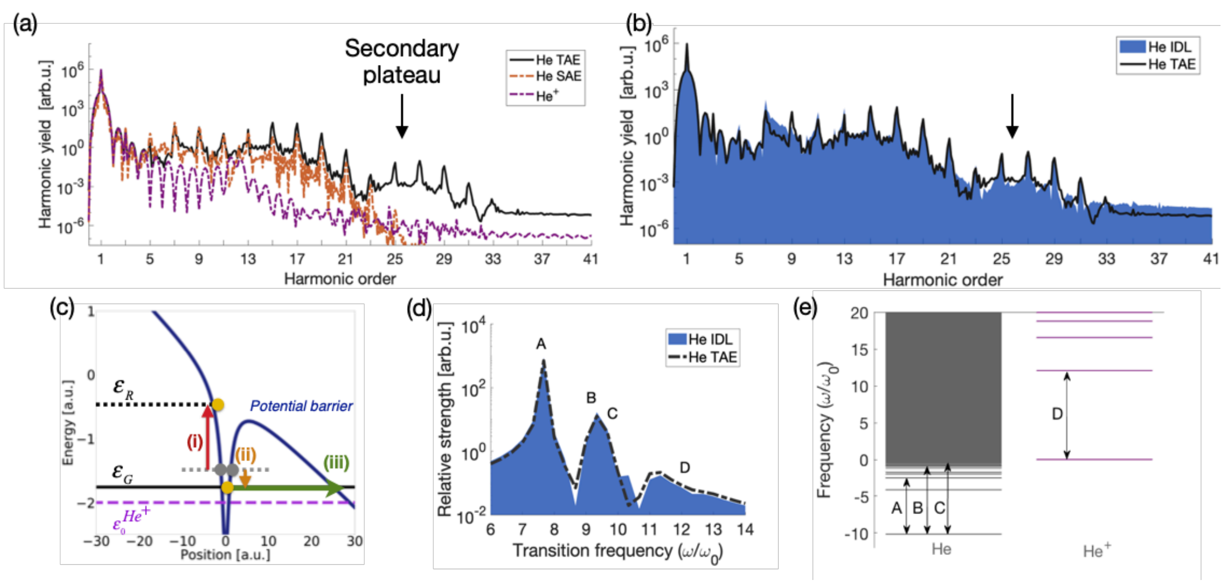

Fig. 1. (a) HHG spectra with two-active electrons (He TAE, solid black line), a single-active electron (He SAE, dashed orange line) and the exact SAE model of He+ (He+, dashed purple line) at a wavelength of 515 nm and 1.6×10^{14} W/cm^2 peak intensity. (b) The HHG spectra when disconnecting one electron from the laser field (He IDL, blue area) agrees with the TAE calculation. (c) Scheme of back reaction before tunnel ionization: one electron is excited to a Rydberg state by e-e correlation (i), whereas the other is knocked down to a deeper level (ii), from which it tunnels (iii). (d) Relative strength of the atomic dipole excitations and (e) energy levels of He and He+.

Figure 1(a) shows the comparison of the HHG spectra from neutral He, within the two-active electron (TAE, solid black line) and the single-active electron (SAE, dashed orange line) descriptions, as well as from He+ (He+, dashed purple line). Both models of neutral He show an excellent agreement up to the 23rd harmonic. In this region, we observe a typical non-perturbative plateau of harmonics characterized by a monoelectronic behavior. Interestingly, the TAE spectrum also includes a secondary plateau at higher frequencies, which is substantially more intense than the emission from He+. The absence of the secondary plateau in the SAE approximation emphasizes its multielectron character.

Noticeably, the secondary plateau is well reproduced even if one electron is artificially turned off from the laser field —idle electron model (IDL), blue area in Fig 1(b) —, while maintaining the electron-electron interaction. This is a distinctive feature compared to other multielectron dynamics in HHG, which involve two electrons effectively interacting with the field [3,4]. Thus, Fig 1(b) confirms that the secondary plateau arises due to electron-electron correlation and points out the passive role of one electron in this HHG mechanism. Indeed, the IDL model is a useful numerical tool to know whether double ionization or double recombination are relevant, since its agreement with the TAE calculation immediately discards these two processes.

Finally, we observe resonance peaks corresponding to electronic transitions in neutral He (Figs. 1d-e) and indicating that single-excited states have been populated both in TAE and IDL. Consequently, the HHG process leading to the formation of the secondary plateau can be summarized in the following steps (depicted in Fig 1c): (i) single-excitation mediated only by electron-electron correlation; (ii) as a back-action, the inner electron is knocked down to a lower energy level; (iii) the inner electron tunnels the potential barrier from this modified level, then it is accelerated by the driving field and eventually, it recombines to the same single-excited state, emitting high-order harmonics. Note that as the inner electron sinks in the potential well, its ionization potential takes an intermediate value between the SAE and the He+. This implies, on one hand, a cut-off frequency higher than the SAE (lower than He+) and, on the other, an intermediate ionization rate between the SAE and the He+, which influences the HHG efficiency. Congruently, these expectations are in accordance with the results in Fig. 1(a).

In conclusion, our numerical simulations reveal back-reaction as a relevant aspect in HHG from correlated two-electron systems, and pave the way to monitor complex mutielectron correlations using high-harmonic spectroscopy.

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