Spectral signature of correlation back-reaction

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Abstract:

High-order harmonic generation from neutral He presents a distinctive trace of correlation back-reaction: a secondary plateau extending the emission towards higher frequencies. We identify a novel mechanism prior to ionization in which the field interacts with one of the electrons, while the other is excited to a Rydberg level through the Coulomb interaction.

Related publication:

A. De las Heras et al, Phys. Rev. Research (accepted 2020)

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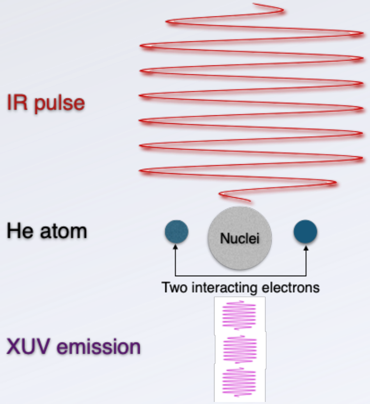
Challenge and theoretical models



The phenomenology of high harmonic generation is typically described with a single-active electron (SAE) occupying the outermost valence orbital of the atom or molecule under study. However, multi-electron effects can influence the ionization or recombination steps.

We look for traces of e-e correlation in the simplest multi-electron system.

Theoretical models 1D

Time-dependent Schrödinger equation

$$i\hbar\frac{\partial\Psi(t)}{\partial t}=H(t)\Psi(t)$$

EXACT 1D MODELS

He TAE: Two Active electrons (exact 1D description of the He atom) — V_{int} V_{ee} V_{int}

$$H^{TAE}=\frac{\left(\hat{p}_1+\frac{e}{c}A(t)\right)^2}{2m_e}+\frac{\left(\hat{p}_2+\frac{e}{c}A(t)\right)^2}{2m_e}-\frac{2e^2}{4\pi\varepsilon_0\sqrt{x_1^{\,2}+0.50a_0}}-\frac{2e^2}{4\pi\varepsilon_0\sqrt{x_2^{\,2}+0.50a_0}}+\frac{e^2}{4\pi\varepsilon_0\sqrt{(x_1-x_2)^2+0.32a_0}}$$

He⁺: Exact He⁺ description — V_{int}

$$H^{He+}=\frac{\left(\hat{p}+\frac{e}{c}A(t)\right)^2}{2m_e}-\frac{2e^2}{4\pi\varepsilon_0\sqrt{x^2+0.50a_0}}$$

APPROXIMATIONS

He IDL: Idle electron (Two correlated electrons but only one interacts with the laser) — V_{ee} V_{int}

$$H^{IDL}=\frac{\left(\hat{p}_1+\frac{e}{c}A(t)\right)^2}{2m_e}+\frac{\hat{p}_2^{\,2}}{2m_e}-\frac{2e^2}{4\pi\varepsilon_0\sqrt{x_1^{\,2}+0.50a_0}}-\frac{2e^2}{4\pi\varepsilon_0\sqrt{x_2^{\,2}+0.50a_0}}+\frac{e^2}{4\pi\varepsilon_0\sqrt{(x_1-x_2)^2+0.32a_0}}$$

He SAE: Single-active electron — V_{int}

$$H^{SAE}=\frac{\left(\hat{p}+\frac{e}{c}A(t)\right)^2}{2m_e}-\frac{e^2}{4\pi\varepsilon_0\sqrt{x^2+0.487a_0}}$$

Numerical results

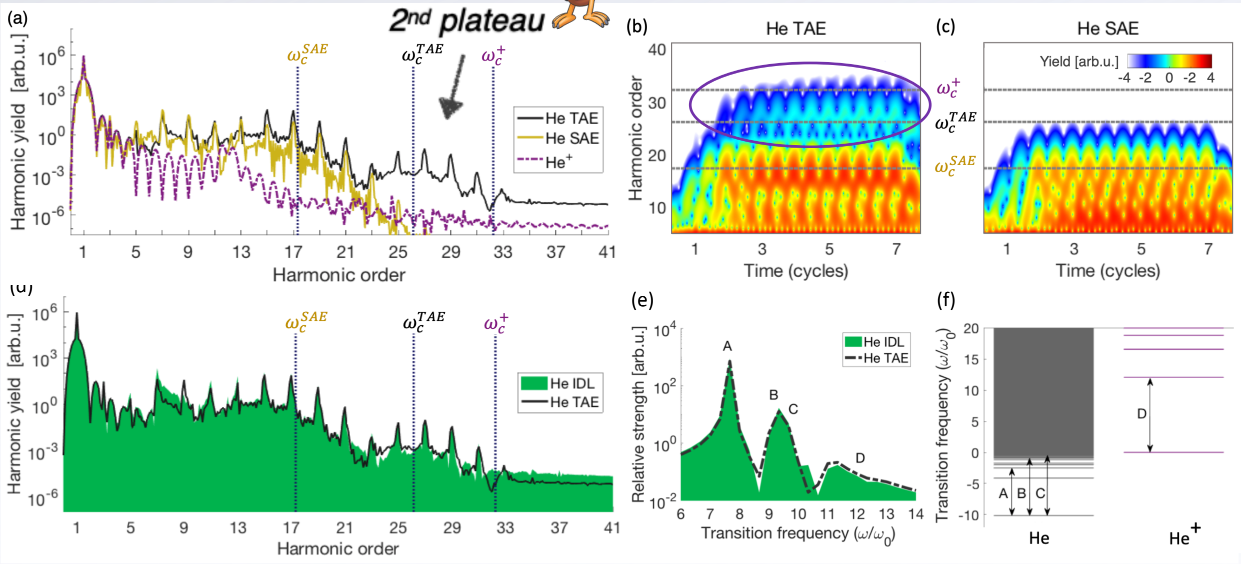
High-harmonic spectroscopy encodes information of e-e correlation!!

- A high-energy secondary plateau emerges in the TAE model with a significantly higher yield than the signal from He^+.

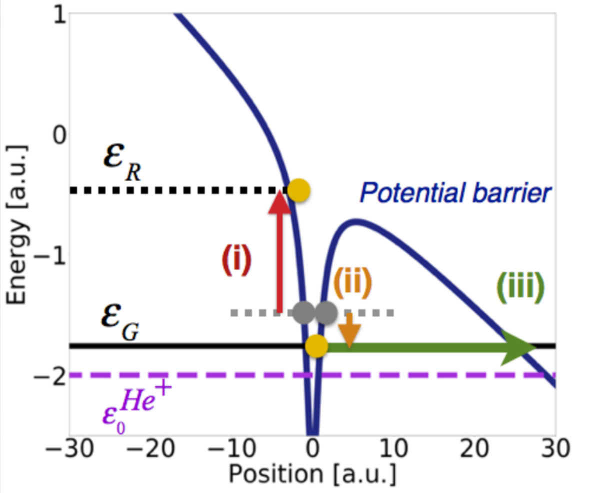
$\lambda = 515\, nm;\ I = 1.6{\times}10^{14}\ W/cm^2$

- The 2^{nd} plateau is preserved even if one electron is artificially disconected from the external field (IDL model in Fig. (d)).

- Single-excited states of neutral He are populated both in the TAE and IDL descriptions (Figs. (e) and (f))



Conclusions



The back-reaction mechanism leading to the secondary plateau involves the following steps:

(i) Single-excitation of the secondary electron mediated by e-e correlation.

(ii) The secondary electron back-acts on the primary electron, lowering its energy.

(iii) The primary electron tunnels the potential barrier from this modified level, then it is accelerated by the driving field and finally it recombines to the same single-excited state, emitting high-order harmonics.

Unlike other multi-electron phenomena[1,2] where the electron-field excitation is modified by cross-interactions with the rest of the electrons, back-reaction is a mechanism of auto-correlation.

Take-home message:

Complex multi-electron interactions can be monitored using high-harmonic spectroscopy!

Further insights: A. De las Heras et al, Phys. Rev. Research (accepted 2020)

[1] T. Tikhomirov et al, Phys. Rev. Lett. 118, 203202 (2017)
[2] P. Koval et al, Phys. Rev. Lett. 98, 043904 (2007)