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## Ab-initio validation of a simple heuristic expression for the sequential-double-ionization contribution to the double ionization of helium by ultrashort XUV pulses

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**Synopsis** We study two-photon double ionization of helium by short XUV pulses by numerically solving the time-dependent Schrödinger equation in full dimensionality within a finite-element discrete-variable-representation scheme. Based on the emission asymmetries in joint photoelectron angular distributions, we identify sequential and non-sequential contributions to two-photon double ionization for ultrashort pulses whose spectrum overlaps the sequential ( $\hbar\omega > 54.4 \text{ eV}$ ) and non-sequential ( $39.5 \text{ eV} < \hbar\omega < 54.4 \text{ eV}$ ) double-ionization regimes.

For sufficiently long extreme ultraviolet (XUV) pulses, sequential and non-sequential two-photon double ionization of helium atoms are distinguishable by the central XUV photon energy. If this energy is above the second ionization potential (54.4 eV), emission proceeds sequentially, otherwise non-sequential correlated double ionization occurs [1]. For ultrashort XUV pulses, this distinction breaks down if their spectrum overlaps the sequential and non-sequential regimes [2].

Solving the time-dependent Schrödinger equation (TDSE) *ab initio*, we investigate joint photoelectron angular distributions (JADs) for two-photon double ionization of helium atoms in the sequential and non-sequential regimes. We find that strong electronic correlation results in similar angular distributions over a large range of energy sharings between the two photoelectrons [3]. Based on the emission asymmetry in JADs (ranging from -1 for emission in opposite to 1 for emission into the same hemisphere), we distinguish "sequential" and "non-sequential" contributions to double ionization by linearly polarized ultrashort XUV pulses.

Figure 1 displays the "effective sequential double-ionization (SDI) yield" and doubleionization emission-asymmetry dependence on the XUV-pulse duration. We define the effective sequential yield as the product of the effective interaction time (pulse duration) and the integrated XUV-pulse-spectral intensity for  $\hbar \omega >$ 54.4 eV (blue area in the inset of Fig. 1). For XUV pulses with a central photon energy of 50 eV, the SDI contribution reaches a maximum value at a pulse duration of 650 as, which we explain as being due to the competition between increasing temporal and decreasing spectral pulse widths.



Figure 1. Effective SDI yield for helium in a  $10^{14}$  W/cm<sup>2</sup> peak intensity XUV pulse with a central energy of 50 eV as a function of the XUV pulse duration. The stars are results of our *ab initio* TDSE calculations [1,3] for the double-emission asymmetry at extremely unequal energy sharing of the emitted electrons. The inset displays a typical XUV-spectral intensity, the blue area indicating the spectrum above the second ionization potential of helium.

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## References

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