Two-electron autoionization dynamics in helium driven by intense XUV fields of a free-electron-laser radiation source

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Synopsis We investigate the 2s2p doubly excited state in helium via extreme-ultraviolet (XUV) absorption spectroscopy and resolve spectroscopic line-shape changes where the autoionizing transition is driven by intense XUV fields of a free-electron-laser radiation source.

The helium atom represents a prototype system where fundamental two-electron correlation dynamics can be investigated with a high degree of theoretical understanding [1,2]. In particular for the case of double excitation of both electrons with a single XUV photon, correlation effects become prominent. For the lowest possible excitation scenario, originating from the $1s^2$ helium ground state, both electrons can be promoted into the $N = 2$ shell, i.e., into the dipole-allowed 2s2p doubly excited state. Correlation effects manifest via the electron–electron interaction (Coulomb repulsion) and lead to the mixing of different two-electron configurations, in this case including the singly ionized 1s*E*p continuum, giving rise to the well-known Fano interference and auto-ionization pathways that are spectroscopically observed as asymmetric line shapes [3].

In previous studies we have experimentally investigated such resonant two-electron transitions in response to moderately strong visible to near-infrared ultrashort laser pulses at tunable intensity. With pulse durations significantly shorter than the autoionization lifetime, a continuous tunability from asymmetric Fano to symmetric Lorentzian line shapes was observed [4], with direct access to quantum dynamical phases that allowed the laser-control of two-electron wave packets [5].

Here we consider the more fundamental case of strongly driven double excitation in helium using XUV-only fields from the free-electron-laser source FLASH in Hamburg with intensities in the range of 10^{12} to 10^{14} W/cm^2. As can be seen in Fig. 1, significant changes in the absorption spectrum across the 2s2p two-electron transition at 60.15 eV are revealed.

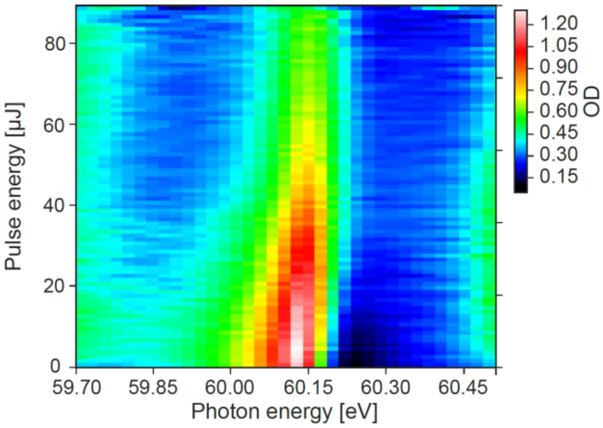

Figure 1. Asymmetric Fano absorption profile (optical density, OD) of the 2s2p single-photon double-excitation transition in helium, and its changes upon interaction with increasing FEL pulse energy.

The spectroscopic access to the absorption line shape enables a direct study of electron correlation effects driven by intense high-frequency fields. A detailed investigation of its theoretical description, including the statistical nature of the employed FEL radiation source is underway. In the future, such studies may guide the way for a deeper understanding and control of strongly driven core-level transitions in the x-ray domain at novel XFEL facilities.

References

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