Atomic resonances in intense XUV electric fields

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Abstract: Driven by intense XUV free-electron lasers, atomic resonances behave different than in the weak-field limit. Employing nonlinear wave-mixing spectroscopy, signatures of strong coupling are revealed and discussed.

Resonant electronic transitions are at the heart of light-matter interaction. Oscillating at extreme ultraviolet (XUV) and x-ray frequencies, one can address element-specific transitions which involve atomic inner-shell and core electrons. Going beyond the weak-field limit of conventional absorption spectroscopy, with intense XUV and x-ray sources these transitions can be strongly driven, which leads to a characteristic change in the absorption spectra. A key quantity hereby is the dipole response, with its coherent decay time typically ranging in the femtosecond and attosecond regime. These short times are directly connected, e.g., to the autoionization and Auger lifetimes of the excitations, being embedded in the ionization continuum. With the availability of intense and ultrashort XFEL sources, new possibilities now arise to coherently control this dipole response prior to its decay, leading to new opportunities for the coherent control of site-specific light-matter interaction.

In this talk I will give a brief overview of our recent experimental activities that are centred on nonlinear wave-mixing and light-matter interaction in moderately dense atomic gas targets in the vicinity of short-lived resonant transitions. We have developed a versatile beamline for XUV-pump XUV-probe transient absorption spectroscopy that was operated at the open-port beamline BL2 at the free-electron laser in Hamburg (FLASH) [1]. Being driven in intense XUV light at 50 eV photon energy, resonant ac Stark shifts of transiently produced ionic neon have been observed [2]. Given the nonlinear interaction of time-delayed XUV electric fields in neon, the experiment further carries signatures of spectral interference and a coherent enhancement of light diffraction at the few-femtosecond timescale in temporal pulse overlap. Furthermore, one can make use of the ionizing nature of the 50 eV radiation and thereby effectively change the XUV transmission properties of the neon target in a time-resolved manner. Probing the spectrally resolved transmission with a time-delayed XUV pulse thus directly encodes its spectro-temporal content with sensitivity to the frequency chirp of FEL pulses [3]. Last but not least, addressing a resonant two-electron transition in helium at 60 eV photon energy, signatures of strong coupling of an autoionizing doubly excited state with the helium ground state have been observed as characteristic changes in the asymmetric Fano resonant line shape [4]. These findings imply that a short-lived population inversion of both electrons in helium can be achieved.

References

[1] Ding T *et al.* 2021 *Faraday Discuss.* DOI: 10.1039/d0fd00107d
[2] Ding T *et al.* 2019 *Phys. Rev. Lett.* 123 103001
[3] Ding T *et al.* 2021 *Nature Communications* 12 643
[4] Ott C *et al.* 2019 *Phys. Rev. Lett.* 123 163201