Contribution ID: **7** Type: **not specified**

Observation of resonant two-photon ionization of He via a doubly excited state using superradiant FEL pulses

Friday 15 January 2021 16:40 (20 minutes)

High-brilliance Free Electron Lasers (FELs) greatly increase the range of accessible experimental conditions and physical phenomena for non-linear studies of atoms, molecules and clusters with VUV and soft X-ray radiation. The seeded Free Electron Laser FERMI in Trieste (FEL-1: 100–20 nm. FEL-2: 20–4 nm) [1] provides typical pulse durations from 90 to 20 fs [2]. However in order to track atomic and molecular dynamics occurring on a faster timescale (i.e. proton motion after ionization [3], isomerization in molecules [4], Auger decay processes in atoms [5]) or to exploit the effect of simultaneous absorption of photons by matter (i.e. collective ionization in clusters [6], double core-hole excitation [7]) the scientific community is pushing for even shorter pulses ideally below 10 fs.
A recent innovative FEL operational scheme, superradiance [8], has demonstrated the generation of few-femtosecond pulses at FERMI [9] further increasing the range of multiphoton, non-linear studies.
We will show how the compression of the pulses is achieved in such an operational scheme and how the temporal duration has been experimentally characterized by means of auto-correlation measurements with two-photon, above-threshold ionization of Ar. In addition, we present results of a first experiment on He performed at the Low Density Matter beamline [10] with superradiant pulses.
Two-photon ionization of He, via the resonant 2s2p doubly excited state [11], was investigated by photoelectron spectroscopy. The superradiant pulse duration was less than the lifetime of this intermediate excited state, allowing us to explore the physics of the resonant excitation. The spectra showed that in this system, two-photon ionization led mostly to excited final states, that is, He ions with an electron in a shell higher than 1s. This result is in excellent agreement with detailed calculations.
The experiments also showed that a common problem with two-photon ionization by FEL light, contamination by second harmonic radiation which creates a background for the two-photon signal, was not a significant problem for superradiant operation.
The success of this experimental campaign opens the possibility to implement the superradiant configuration as one of the operational modes of FERMI available for users'proposals.

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Track Classification: WavemiX 2021