Quantum Control of Atomic Transitions with a Strong-Field Extreme-Ultraviolet Free-Electron Laser

A Trabattoni[1,2]

[1]*DESY, 22607 Notkestr. 85, Hamburg, Germany. Contact Phone: +49 4089986048*
[2]*Institut für Quantenoptik, Leibniz Universität Hannover, 30167 Welfengarten 1, Hannover, Germany. Contact Phone: +49 4089986048*
Contact Email: andrea.trabattoni@desy.de

Modern free-electron lasers (FELs) are establishing a new paradigm for the study of light–matter interactions, spanning phenomena from metastable nuclear transitions to ultrafast core-electron dynamics. In this framework, recent advances in the spectro-temporal shaping of ultraintense XUV to X-ray pulses are enabling the exploration of quantum-control strategies and non-perturbative photoexcitation regimes. These capabilities open new avenues for creating and manipulating correlations in atomic ensembles using XUV/X-ray radiation, as well as for efficiently populating narrow-linewidth transitions. Developing robust quantum-control protocols in this newly accessible XUV to X-ray regime is therefore a key objective(9; 10). Rapid adiabatic passage (RAP), originally introduced in nuclear magnetic resonance spectroscopy, represents a particularly powerful control technique.

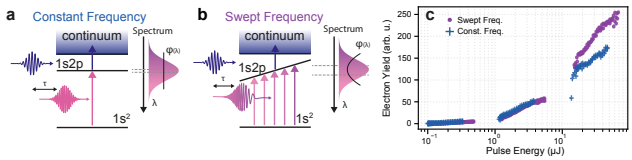

Figure 1: Experimental Results: a, Level diagram of the constant frequency excitation of the helium atom through the XUV FEL pulse, followed by ionization through a 266 nm UV pulse. b, Level-diagram of the RAP driven helium excitation and ionization with an FEL pulse of swept frequency (chirped). c, Experimental photoelectron yield of the 1.3 eV emission line as a function of FEL energy, and chirped versus constant frequency FEL pulse

It enables efficient and robust photoexcitation while remaining insensitive to intensity fluctuations and spatial inhomogeneities of the driving field. So far, however, RAP has been largely confined to spectral regions ranging from the radio-frequency domain to the ultraviolet. In this work, we demonstrate quantum control of an atomic transition by implementing rapid adiabatic passage in the XUV spectral range using an ultraintense, chirped FEL pulse. Specifically, we show that the narrow-linewidth, long-lived (around 1 ns) helium 2p state can be efficiently populated by exploiting the high peak intensity and pulse-shaping capabilities of a broadband seeded FEL. The achieved excitation yield significantly exceeds that obtained with conventional weak-field resonant excitation. These findings, combined with ongoing advances in ultraintense FEL technology, open new perspectives for coherent control at free-electron lasers, with potential applications ranging from nuclear laser spectroscopy to nonlinear X-ray science. The experiment was carried out at the seeded FERMI FEL at ELETTRA, Trieste. The FEL photon energy was tuned to the 1s-to-1s2p transition in helium at 21.18 eV and focused into an atomic helium gas jet (d = 1.5 mm). The excited-state population was probed via linear ionization using a UV laser pulse with a duration of 40 fs, a wavelength of 266 nm, and a delay of 1 ps. The angle-resolved photoelectron momentum distribution was recorded with a velocity-map imaging (VMI) spectrometer, while the corresponding photoions were detected using a time-of-flight spectrometer. The photoline at 1.3 eV energy is a direct probe of the FEL-driven relative population of the He 2p state. The FEL was operated at two conditions, (i) delivering XUV pulses of around 80 fs duration and constant carrier frequency (Fig. 1a) and (ii) a chirped pulse (group-delay dispersion $\sim$ -1000 fs^2) of around 110 fs duration with a continuous frequency sweep (Fig. 1b). In condition (i) the FEL pulse is resonantly exciting the transition with a constant frequency. In condition (ii), the photoexcitation is driven through rapid adiabatic passage. The transient dressing of the transition causes a broader absorption window and an excitation probability of approximately equal to 1 can be reached. To investigate the transition from perturbative to non-perturbative excitation and the onset of the adiabatic population enhancement, the photoelectron spectra for conditions (i) and (ii)

were collected as the FEL intensity was varied over 3 orders of magnitude. The result is reported in Fig. 1c as a function of FEL pulse energy, corresponding to a peak intensity range approximately between 10^{11} to 10^{14} W/cm^2. The intensity-dependent electron yield is showing an approximately exponential trend. At low energies, the yield is slightly larger for the unchirped XUV pulse, while for pulse energies larger than 10 µJ (peak intensity $\sim 10^{13}$ W/cm^2), RAP becomes the major effect and the yield is enhanced in the presence of the chirped FEL pulse. These results are consistent with theoretical models describing the multi-level dynamics and incorporate the full three-dimensional intensity distribution in the interaction volume (not shown), verifying that we are observing the onset of non-perturbative photoexcitation and the effect of RAP in the population enhancement.

With this work, we establish rapid adiabatic passage as an effective and robust quantum-control protocol for atomic transitions with an intense pulse-shaped XUV free-electron laser. Compared to conventional resonant excitation, which is strongly affected by intensity-dependent Rabi dynamics and spatial inhomogeneity across the focused interaction volume, the chirped FEL pulse yields a higher net excitation at high pulse energies, and is robust against intensity fluctuations. Looking ahead, spectro-temporal shaping of intense XUV pulses will allow for high-fidelity manipulation of excited atomic ensembles in the XUV opening a route to exploring correlation-driven phenomena such as superradiance, or to efficiently populate narrow-linewidth nuclear transitions.