Photoelectron spectroscopy of laser-dressed atomic helium

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Photoelectron emission from excited states of laser-dressed atomic helium is analyzed with respect to laser intensity-dependent excitation energy shifts and angular distributions. In the two-color XUV (extreme ultraviolet) – IR (infrared) measurement, the XUV photon energy is scanned between 20.4 eV and the ionization threshold at 24.6 eV, revealing electric dipole-forbidden transitions for a temporally overlapping IR pulse (≈10^{12} W/cm^2). The interpretation of the experimental results is supported by numerically solving the time-dependent Schrödinger equation in a single-active-electron approximation.

Photoelectron spectroscopy is a powerful technique to obtain compositional and structural information about matter and to investigate light–matter interactions in general. It has been successfully employed and continuously developed over many decades in atomic and molecular physics [1]. Photoelectrons carry information about the electronic bound and continuum states of the corresponding atom, as well as information about the absorbed and emitted photons.

With the advent of intense optical lasers, multiphoton absorption in atoms and molecules became feasible, enabling the observation of a variety of new phenomena, e.g., multiphoton excitation microscopy [2], resonance-enhanced multiphoton ionization (REMPI) [3], Doppler-free two-photon spectroscopy [4, 5], and high-harmonic generation (HHG) [6, 7], to name just a few. One step further in the investigation of light–matter interactions is the implementation of two-color ionization and excitation schemes, which reveal laser-induced continuum structures [8] and light-induced structures (LIS) [9]. In the former case, the dressing laser field couples bound states to the continuum, giving rise to a resonant structure [10–13]. In the latter case, the ground state is coupled to excited states beyond the one-photon allowed dipole transition via two-color photo-excitation [14–17].

In this Letter, we report the use of XUV (extreme ultraviolet) radiation with tunable wavelength provided by the free-electron laser in Hamburg (FLASH) in combination with a synchronized IR laser to obtain a detailed picture of excited states in laser-dressed atomic helium. The XUV photon energy is scanned over the 1snp^{1P} Rydberg excitation-series to a value just below the ionization threshold. The superimposed IR pulses (800 nm wavelength) arrive with a freely adjustable time delay with respect to the excitation pulses. Their intensity is too low to ionize He in its ground state, but strong enough to ionize it from excited states that are temporarily reached via a combination of XUV and IR photons. The ionization yield and angular distributions are analyzed as a function of the XUV photon energy, the IR time-delay, and the IR intensity. In the case of temporally overlapping pulses, by absorption of one XUV and one or more IR photons, one electron is lifted from the ground into a continuum state through laser-dressed excited states, including those that are not accessible by pure single-photon excitation. The interpretation of the experimental results is supported by numerical calculations based on the time-dependent Schrödinger equation (TDSE) within the single-active electron (SAE) approximation.

This two-color scheme has the clear advantage over, e.g., single-color REMPI setups, that the dominant contribution to the excitation energy is delivered by only one XUV photon, and hence the laser intensity can be kept low. Therefore, field-induced changes to energy levels and fragmentation are minimized. This opens up precision-spectroscopic studies of atoms and molecules under less-perturbing conditions.

The experiment was carried out with the reaction microscope (REMI) endstation [18, 19] at the free-electron laser (FEL) FLASH2 [20, 21]. FLASH2 features variable-gap undulators that allow to quickly change the photon wavelength [22] over a broad spectral range. During the measurements, the XUV photon energy was scanned in steps of 0.2 eV from 20.4 eV to just below the ionization threshold of atomic helium at 24.6 eV [23]. The FEL pulse-length was about 40 fs full-width at half maximum (FWHM) in intensity, and the pulse energy (< 10 nJ) was reduced to a level such that two-XUV-photon absorption in He can be neglected. Synchronized, but with a timing jitter of several ten femtoseconds, the IR probe-laser (800 nm) was superimposed collinearly with the XUV beam. In order to ensure optimal temporal over-

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lap with the FEL pulses, an IR pulse-duration of about 90 fs FWHM was chosen. The IR pulse energy and the focusing conditions were adjusted such that intensities in the order of up to $10^{13}$ W/cm$^2$ were reached in the target. With a diameter of about 30 µm the focus of the IR beam was significantly wider than the FEL focus ($\approx$ 10 µm). XUV and IR radiation were linearly polarized and aligned parallel to each other. Both beams were focused onto a dilute supersonic gas jet of atomic helium in the center of the REMI, which is equipped with two time- and position-sensitive detectors [24] to collect all charged fragments (electrons and ions) within the full 4$\pi$ solid angle. Time-of-flight and position information is used to retrieve the particles’ momentum vectors at the time of ionization.

During the XUV photon-energy scans, the FEL pulse energy and the FEL beam diameter change slightly. Together with the energy-dependent absorber-foil transmissions and mirror reflectivities, this leads to small variations in the photon flux. In our analysis, these effects are corrected by normalizing the data for each XUV energy with the simultaneously recorded yield of H$_2^+$ ions, which stem from a constant and weak background of H$_2$ gas in the REMI chamber. The ionization cross section of H$_2^+$ was taken from [25].

In the experiment two sets of data were taken, one with an IR intensity of approximately $I_{\text{high}} \approx 8 \times 10^{12}$ W/cm$^2$ and one with $I_{\text{low}} \approx 1 \times 10^{12}$ W/cm$^2$. We note that the temporal jitter between FEL and IR, which is comparable to the IR pulse duration, leads effectively to a lowering of the average IR intensity for the case of overlapping pulses. The influence of this imperfect overlap of both pulses increases with the IR intensity in the same way as the contribution of nonlinear multi-photon transitions increases. Therefore, in the comparison to theory, a smaller difference in intensity between the low and high IR intensity case was chosen in our calculations in order to mimic the corresponding experimental conditions.

The theoretical part of this study is based on numerically solving the TDSE in the SAE approximation. Electrons are assumed to be non-interacting, while the ground state is effectively described as a 1s1s$'$ 1S state, where the 1s is close to the He$^+$ orbital and the 1s$'$ is treated like a valence orbital. As always in theory, the binding energies of the $n\ell$ valence electrons are not exact. While 1sn$\ell$ Rydberg states with angular momenta $\ell \geq 2$ have very accurate binding energies, this is not quite the case for p-electrons and particularly for s-electrons, due to the small or missing centrifugal barrier. Since excitation energies are measured from the ground state, much of the remaining discrepancies are due to the binding energy of the 1s$'$ orbital.

Specifically, we used the same one-electron potential as Birk et al. [26],

$$V(r) = -\frac{1}{r} - \left(\frac{1}{r} + 1.3313\right) \exp(-3.0634r),$$

where $r$ is the distance from the nucleus, to calculate the valence orbitals. The difference of excitation energies compared to the recommended excitation from the NIST database [23], is less than 0.2 eV even in the worst-case scenario and does not alter the essential conclusions presented below. We will sometimes omit the inner 1s electron to simplify the notation, keeping in mind that only two-electron singlet spin states are accessible, since spin-forbidden transitions are negligible.

The laser parameters were chosen according to the available knowledge regarding the actual experimental conditions. The XUV pulse duration was taken as 40 fs (FWHM value of a peak intensity of $1 \times 10^{10}$ W/cm$^2$ with a Gaussian envelope) and the IR pulse duration as 80 fs. While the XUV photon energy was varied over a range in steps of 0.05 eV, the central IR photon energy was held fixed at 1.55 eV (800 nm). Since both beams are linearly polarized along the same direction, the initial state can be propagated very efficiently and accurately. Specifically, we used an updated version of the code described by Douguet et al. [27].

![FIG. 1: Electronic energy level scheme of helium with different ionization pathways through intermediate excited states. The XUV energy scanning range is indicated by the blue box.](image1)

![FIG. 2: Photoelectron yield measured for two different delays between the XUV and IR pulses. Both curves are normalized to their maximum yield, and statistical error bars are much smaller than the line thickness.](image2)
Over the XUV scanning range, the helium atom can be excited from the 1s² 1S ground state to a 1snp 1P excited state for specific XUV photon energies, according to the electric-dipole selection rules. The excited atom can be ionized by absorbing one or more IR photons (ℏω = 1.55 eV) of a subsequent laser pulse, promoting the weakly bound electron into the continuum. This mechanism is depicted in an energy-level scheme in the upper part of Fig. 1.

Experimental data for the corresponding process are shown in Fig. 2, where the yield of photoelectrons is plotted against the XUV photon energy for a non-overlapping temporally delayed IR pulse with intensity $I_{\text{high}}$. Clearly visible are the yield enhancements for XUV energies that match the 1snp 1P excitation energies in helium (2p at 21.2 eV, 3p at 23.1 eV, 4p at 23.7 eV etc. [23]). Also shown in Fig. 2 is the photoelectron yield for XUV and IR pulses in temporal overlap (yellow distribution). Compared to delayed ionization, the 1P-excitation peaks remain while an additional maximum appears around 22.4 eV. This feature was observed and described in transient-absorption measurements as a LIS [9, 15, 28, 29]. As XUV and IR radiation are simultaneously present, the helium atom can undergo dipole-forbidden (for single photons) transitions, provided the XUV photon absorption is accompanied by the absorption or emission of IR photons of the dressing laser field. In the simplest and dominant case, one XUV photon and one IR photon combined drive $1s^2 \rightarrow 1sn$ or $1s^2 \rightarrow 1sd$ transitions. By absorbing additional IR photons, the excited atom is ionized.

This mechanism is depicted in the lower part of Fig. 1. Direct two-photon absorption couples the ground state to 1sns and 1snd states. In contrast to P-state ionization, peaks of LIS emerge at XUV energies matching the energy of the excited state plus/minus one IR photon.

In Fig. 3 the measured yield distributions for overlapping pulses are shown for the two cases of low and high IR intensity. The purple distribution in the background is recorded with $I_{\text{low}}$, while the yellow distribution is again for the significantly larger intensity $I_{\text{high}}$. The latter exhibits a gradual decrease in the yield from the peak corresponding to the 2p state at 21.2 eV over the 3p state at 23.2 eV up to the higher np states (not visible due to the resolution). This overall decrease with rising XUV energy can be explained by the energy dependence of the cross section for the excitation step [30]. The excitation probabilities are directly mapped to the ionization yield in the case of $I_{\text{high}}$, where the IR intensity is large enough to ionize all excited states independent of the number of photons (N) needed. For low IR intensity (purple distribution in Fig. 3), on the other hand, the scaling law $R \sim I^N$ of the transition rate $R$ with the laser intensity $I$ in multiphoton processes becomes relevant [31]. Therefore, the ionization yield of the 2p state, which requires $N = 3$ IR photons, is reduced compared to the yield from the 3p and higher np states ($N = 1$).

Two approaches are employed to assign the LIS to specific field-free atomic excited states. First, we analyze the calculated distributions for excitation in combination with ionization and compare them with the experimental ionization yield. Second, the inspection of the measured photoelectron angular distribution allows us to deduce the intermediate bound state that the electron was emitted from.

Fig. 4 shows theoretical predictions for XUV and IR pulses in temporal overlap. The calculated ionization probability is plotted against the XUV photon energy for two different IR intensities, color-coded in yellow and purple for high and low IR intensity, respectively (similar to the measurement shown in Fig. 3). In addition to the ionization yield, our calculations predict the population

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**FIG. 3:** Photoelectron yield measured for temporally overlapping XUV and IR pulses. The distribution for high IR intensities ($I_{\text{high}}$) is shown in yellow (light gray), the distribution for low IR intensities ($I_{\text{low}}$) in purple (dark).

**FIG. 4:** Calculated ionization probability for two different IR intensities. Arrows in the top panel indicate the calculated positions of excited (black) and light-induced (red) states. + (−) IR denotes the emission (absorption) of an IR photon.
distribution of excited atomic states at the end of the pulses as a function of the XUV energy and the IR intensity. The most prominently populated states in the theoretical excitation-probability distributions are marked by arrows in Fig. 4. They serve as an indicator of the role of the respective excited states and their population (either direct or light-induced) at a given XUV energy en route to ionization. This allows assignment of the peaks in the ionization probability distribution (bottom panel of Fig. 4) to the excited states from which the atom is ionized.

Electronic energy levels experience an AC Stark shift due to the IR laser field [32, 33]. This shift is clearly seen for the 3p state in theory (see Fig. 4), but is much smaller in the experiment (see Fig. 3). However, experimental Stark/ponderomotive shifts can be seen in the photoelectron kinetic energy spectrum (not shown here). Overall, we see good agreement between experiment and theory when comparing the purple distributions in Figs. 3 and 4. Similar to the experiment, the calculated ionization yield shows a reduced contribution of the 2p state relative to the 3p state when the IR intensity is lowered. Moreover, the strengths and the positions of the LIS peaks, which only appear in temporal overlap, are well reproduced by the calculations. Small shifts in energy are attributed to the already mentioned inherent inaccuracies of electronic binding energies in the SAE model. Most importantly, for low IR intensity the dominating LIS peak is found around 21.8 eV as in the experimental counterpart. As the enhancements by other excited states appear at distinctively different positions, and are much less pronounced, this dominating LIS peak at 21.8 eV (purple distribution in Fig. 3) can be assigned to the 3d state.

The situation changes for high IR intensity where the dominant LIS peak is shifted to a larger XUV energy of about 22.4 eV in experiment (yellow in Fig. 3). Comparison with theory indicates that in this case ionization proceeds through the Stark-shifted 3d and 2s excited states. Both contribute to the dominating LIS peak at about 22.2 eV according to our state assignment in Fig. 4. Relative to the 3d state, the 2s contribution becomes more relevant at high intensity because, in order to populate the 2s state, the atom absorbs one XUV photon while emitting one IR photon. Ionization takes place by absorbing another three IR photons. In contrast to this effective four IR-photon transition, ionization via the 3d state involves only two IR photons. We note that the large shift of the 3d LIS with IR intensity seen in the experiment is also consistent with the calculation by Chen et al. [9].

The electronic structure of the dominant LIS involving both the 2s and 3d excited states at 22.4 eV for high IR intensity in Fig. 3 can also be deduced from the photoelectron angular distribution (PAD). This is shown in Fig. 5 where the yield of electrons is plotted as a function of their emission angle Θ with respect to the laser polarization axis.

The distribution exhibits a typical “F-like” shape, indicating the angular quantum number of the continuum state to be L = 3. Starting from a 1S state (L = 0), an angular quantum number of L = 3 can only be reached by absorbing at least three photons. For an atom in a 1D state (L = 2), absorbing one IR photon is sufficient to obtain an F-like PAD. One can deduce the populated bound states by taking the photoelectron kinetic energy (0.4 eV) minus the IR photon energies (each 1.55 eV) while also accounting for the Stark-/ponderomotively shifted continuum level. One finds that the matching states are the 3d and 2s excited states. The yellow solid line in Fig. 5 shows the calculated PAD for the considered LIS. We find very good agreement between experiment and theory, thereby supporting our interpretation of the mechanism.

The three-dimensional PAD of the dominant LIS is also contained in the measured three-dimensional photoelectron momentum distribution, shown in the inset of Fig. 5. Dots in the plot represent a bin in momentum space, with the yield within each bin being color-coded. The energy range was chosen from 0.3 eV to 0.5 eV, resulting in a spherical shell of dots in the plot. The projections on the walls show the integrated yield along one specific direction. The two small maxima in Fig. 5 are found as two rings around the polarization axis, while the large maxima are found in the three-dimensional distribution for maximal absolute p_x momentum, i.e. along the polarization axis of IR and XUV.

To summarize: We have measured and analyzed photoelectrons stemming from laser-dressed atomic helium. The helium atoms were photo-excited over a large energy range by XUV FEL radiation in the presence of a moderately strong IR laser (~10^{12} W/cm^2). The scheme allowed us to reveal light-induced structures alongside
the $^1P$ Rydberg series. The observed excitation energies of the LIS for varying IR intensities were supported by TDSE calculations. The photoelectron angular distribution and the kinetic energy were used to assign the LIS. The dominant LIS in the photoelectron yield of laser-dressed helium was identified as stemming from the $1s3d^1D$ excited state for an IR intensity of about $1 \times 10^{12} \text{W/cm}^2$, while for a higher IR intensity of about $4 \times 10^{12} \text{W/cm}^2$ the $1s2s^1S$ state also contributes significantly.

Our investigation complements previous transient-absorption measurements on light-induced structures, but brings up additional aspects. While transient-absorption measurements reveal LIS in the absorption spectrum without the need for ionization, our measurement is sensitive to the excited states from which electrons are emitted and allows to measure and assign angular distributions of the corresponding continuum final states.

The presented analysis reveals the preference to populate the $1s3d^1D$ state rather than the $1s3s^1S$ state. This is clearly shown in the high IR intensity case, where all excited states get ionized, so the yield is independent of the number of ionizing photons. We find the propensity of helium in its ground state to increase angular momentum by two-photon absorption, thus driving a bound-bound transition. This can be understood in the context of Fano’s propensity rule [34], originally stated between bound and continuum states, and the propensity analysis of continuum-continuum transitions by Busto et al. [35], both stating the propensity to increase the angular momentum in photo-absorption.

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