Core-resonance line-shape analysis of atoms undergoing strong-field ionization

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Received 27 June 2022, revised 22 September 2022
Accepted for publication 7 October 2022
Published 18 November 2022

Abstract

Using attosecond transient absorption spectroscopy for time delays where the near-infrared pump and the extreme ultraviolet (XUV) probe pulses overlap, sub-cycle structures in the build-up of absorption lines in xenon ions are investigated as a function of the pump intensity during strong-field ionization. We observe a half-cycle-periodic change in the line-shape asymmetry of the ionic 4d–5p resonances. Analyzing the line shapes, we find that in particular the phase of the induced dipole emission is modified, and the magnitude of this phase modulation decreases with increasing laser intensity. We discuss the influence of ground state depletion on interfering pathways involved in XUV-assisted strong-field ionization.

Keywords: strong-field ionization, attosecond transient absorption spectroscopy, R-matrix with time-dependence (RMT)

(Some figures may appear in colour only in the online journal)

1. Introduction

The (r)evolution of femto- to attosecond physics was launched by harnessing the sub-cycle dynamics in strong-field ionization (SFI) for high-order harmonic generation (HHG) [1, 2]. By generating coherent attosecond pulses of light [3, 4] for use in experiments, SFI has enabled the study of dynamics on ever shorter time scales [5–7]. In addition, other techniques such as laser induced electron diffraction [8] and high-harmonic spectroscopy [9, 10] take direct advantage of SFI as a crucial first step. Thus, SFI is not only a fundamental interaction between light and matter but also a powerful tool for the study of atomic and molecular structure and dynamics used in laboratories around the world.

The time-resolved investigation of SFI has attracted attention from the early stages of attosecond physics [11–16]. Attosecond transient absorption spectroscopy (ATAS) is particularly well suited for this task, since the all-optical approach probes the system while it is being ionized. Studies using ATAS investigated coherences in strong-field generated ions [12, 17, 18] and uncovered the non-monotonic increase of ion population through reversible polarization dynamics [19]. With ATAS the investigation of ionization dynamics benefits due to the inherent charge-state specificity [18] and site-selectivity [20] of the extreme ultraviolet (XUV) transitions and features attosecond time resolution through the use of ultrashort probe pulses.

Through XUV spectrometers with high spectral resolution, absorption lines can not only be investigated with respect to their energy and line/oscillator strength but also via their specific shape. Within the framework of time-resolved line-shape analysis [21, 22], a continuous transition between a symmetric Lorentzian to an asymmetric Fano line is the result of a change in the phase of the dipole that is at the origin of an absorption...
line [23]. This dipole phase $\phi_0$ can typically be extracted by fitting a line-shape model to the resonance as well as through the direct reconstruction of the temporal dipole response [24]. Together with an absorption line’s amplitude, the dipole phase constitutes an additional experimental observable that is linked to the energy structure and ultrafast coupling of states in a quantum system.

For the time-resolved study of SFI of many-electron atoms, Pabst et al [21] investigated the deformation of spectral lines for the case of overlapping pump and probe pulses in ATAS. Focusing on different couplings in the Hamiltonian, it was found that the origin of a non-zero dipole phase $\phi_0$ — and hence an asymmetric line shape — lies in the non-perturbative dressing of the entire multi-electron system during ionization with both strong-field near-infrared (NIR) and weak attosecond XUV pulses.

In this kind of ATAS experiment, SFI is driven by focusing a few-femtosecond NIR laser pulse on the target atom. The intensities needed to ionize are in the regime of $10^{14}$ W cm$^{-2}$. In the heavy noble gases, like krypton or xenon, the liberation of a valence $p$-shell electron then opens up excitation channels for the XUV pulse to promote an electron from a more strongly bound $d$-shell to the valence-hole. These transitions are characteristic to the ion and observing their formation as the XUV-probe pulse scans over the NIR-pump pulse allows study of the build-up and coherence of the ion population on the attosecond timescale [17].

Here we perform a systematic analysis of the line shape of an ionic core-to-valence transition in xenon by using ATAS at different NIR pump intensities $I_{\text{NIR}}$ and the attosecond time delay $\tau$ when the pulses overlap in time. Extracting the dipole phase $\phi_0(I_{\text{NIR}}, \tau)$ from the measured line-shape, we identify an indirect ionization pathway — SFI of neutral xenon from an XUV core-excited virtual state — which interferes with the direct pathway of valence-shell SFI, leading to delay-dependent asymmetry changes of a xenon ion XUV absorption line shape. More specifically, we observe delay-dependent NIR-half-cycle oscillations of the line-shape asymmetry whose amplitude decreases with increasing NIR pump intensity. We attribute this effect to the depletion of the neutral ground state, and hence a weakening of the interfering virtual pathway, which is confirmed by estimating the remaining neutral Xe population and supported by ab initio simulations [25].

2. Experiment

The experiment is performed in a classical pump-probe scheme (see figure 1). Intense 760 nm NIR pulses ionize the xenon target and populate the $5p^{3/2}$ and $5p^{1/2}$ valence-hole states. The same NIR pulse has also been used upstream for the generation of the XUV attosecond pulse. Due to the intrinsic phase locking of the underlying mechanism of HHG, small drifts over a longer time-scale of the short-term stable carrier-envelope phase of the NIR pulses have a negligible effect on the results reported below. Time-delayed XUV probe pulses interrogate the ion population through the resonant 4d–5p transitions by means of excitation of a 4d core electron. The intensity of the NIR can be adjusted with a piezo-controlled closed-loop iris aperture. The NIR pulse duration is characterized by a $d$-scan setup [26] to be 4.5 fs full-width at half maximum (FWHM). For more information about the experimental beamline see reference [27]. The measurements reported here were performed at several different NIR intensities in the range of $1–2.5 \times 10^{14}$ W cm$^{-2}$, corresponding to Keldysh parameters of $\gamma \lesssim 1$, such that ionization is predominantly driven in the tunneling regime [28].

Recording XUV absorption spectra downstream of the target, the spectroscopic signature of the Xe$^+$ ion is imprinted in three absorption lines between the dipole-allowed fine-structure configurations of the $5p^{-1}$ valence-hole and the $4d^{-1}$ core-hole states that can be probed between 55 eV and 58 eV. In the results that follow we focus only on the first of these transitions, labelled $T_1$ in figure 1. The absorbance of the xenon target at 18 mbar backing pressure is quantified in terms of a differential optical density AOD [29, 30]. Here, the absorption spectra are referenced by spectra at a fixed time-delay position at which the probe pulse arrives 17 fs before the pump pulse, with negligible effect on the XUV absorption of the target. These reference spectra at negative time delay are recorded in an alternating manner for every time-delay step. A linear combination of these measured reference spectra is used to calculate the actual reference spectrum for each time delay. The coefficients of this linear combination are found by edge-pixel referencing, identifying regions in the XUV spectrum that do not carry resonant absorption signatures of the xenon ions. The method is further described in chapter 4.3 of reference [30], which is very similar to the one published in reference [29]. To obtain high-quality data we averaged 36 time-delay scans for each of the 10 pump intensities between $1–2.5 \times 10^{14}$ W cm$^{-2}$. In each of these 360 measurements we scan the region of temporal overlap between the XUV and NIR pulses with a delay resolution of 85 as.

3. Theory

Our theoretical simulations employ the R-matrix with time-dependence (RMT) code [25, 31, 32]. RMT is a fully non-perturbative, ab initio approach to solve the TDSE for multi-electron atoms and molecules driven by short, intense laser pulses. The RMT calculations employ a non-relativistic description of the Xe atom [33], comprising Hartree–Fock orbitals up to the $5p$, and all $4d^{-1}6\ell$, $5s^{-1}6\ell$ and $5p^{-1}6\ell$ continuum channels up to a maximum total angular momentum of 40. The NIR pulse has a central wavelength of 760 nm with a Gaussian envelope (FWHM 4.5 fs). The attosecond pulse has a Gaussian envelope (FWHM 250 as) and a central photon energy of 61 eV. This energy is chosen to match the energy gap between the 4d and 5p states in the calculated atomic structure, which is larger than the experimental value of 55 eV. The theoretical results shown are thus shifted down by 5.5 eV to aid comparison with the experiment. The RMT simulations provide the time-dependent expectation value of the dipole, from which the absorption spectrum can be calculated [34].
pulses at time delay $\tau$ pass an iris to adjust the NIR intensity before being focused into the target cell filled with xenon gas. Further downstream the NIR light is blocked by a 200 nm thick aluminum (Al) foil filter. The transmitted XUV light is dispersed by a grating and recorded with an XUV camera. The XUV spectrum shown together with the TAS data is the average of all reference spectra recorded at the second highest intensity. (b) Level scheme of xenon with the targeted ion transitions. Dashed arrows indicate the coupled-field mechanism.

**Figure 1.** (a) Sketch of the experimental setup. The concentric XUV (purple) and NIR (red) beams of attosecond XUV and few-cycle NIR pulses at time delay $\tau$ pass an iris to adjust the NIR intensity before being focused into the target cell filled with xenon gas. Further downstream the NIR light is blocked by a 200 nm thick aluminum (Al) foil filter. The transmitted XUV light is dispersed by a grating and recorded with an XUV camera. The XUV spectrum shown together with the TAS data is the average of all reference spectra recorded at the second highest intensity. (b) Level scheme of xenon with the targeted ion transitions. Dashed arrows indicate the coupled-field mechanism.

4. Observation

Figure 2 shows a pump-probe delay scan, obtained from experiment and from RMT calculations, centered around the transition labeled T1 in figure 1 ($5p_1^2 \rightarrow 4d_{5/2}^1$) at an NIR intensity of $(1.91 \pm 0.21) \times 10^{-14}$ W cm$^{-2}$ as an example of the emergence of an ionic absorption line as the XUV pulse passes over the NIR pump pulse. The T1 resonance lies high above the Xe$^{2+}$ ionization threshold and is therefore short-lived with a lifetime of $\tau = 6$ fs after which it decays through the Auger–Meitner effect. The RMT calculations do not account for the Auger–Meitner effect. However, this is simulated in post-processing by applying an exponential decay to the time-dependent expectation value of the dipole. At late positive time delays, far out of the temporal overlap region, one observes a symmetric Lorentzian profile with a line width of $\Gamma = 110$ meV [35]. Half-cycle structures (given the full-cycle duration $T_{\text{NIR}} = 2.5$ fs) manifest in the build-up of the ionic line as observed previously [19] and give proof of the excellent timing stability of our setup. Furthermore, it is apparent that the asymmetry of the absorption line is modulated with a half-cycle periodicity as well. The RMT calculations show the same half-cycle periodicity of the absorption line asymmetry, providing good agreement with experiment. In the following, we perform an in-depth line-shape analysis of this ionic resonance (T1) at different NIR pump intensities.

5. Analysis

A line-shape model is used to perform a fit to the observed absorption line. The model was presented by Pabst et al [21] and notably used and verified in the analysis of reference [19]. The model can be connected to the dipole-control model [22] for a decaying dipole subjected to an impulsive phase shift. It corresponds to a generalized absorption profile parametrized as a function of the dipole phase shift $\phi_0$. According to reference [21], the time-delay-dependent absorption $\text{OD}(\omega, \tau)$ is given by:

$$\text{OD}(\omega, \tau) = \frac{(\varrho L)_{\text{in}}}{\ln(10)} 4\pi\alpha_{\text{in}} \sum_i z_0(\tau) \times \frac{\Gamma(\tau)/2 \cos(\phi_0(\tau)) + (\omega - \omega_\tau) \sin(\phi_0(\tau))}{(\omega - \omega_\tau)^2 + \Gamma(\tau)^2/4} + C(\tau).$$

The model is used to fit the optical density, from both our experiment and simulation, as a function of the photon energy $h\omega$ for every time delay $\tau$. For the experimental data, the sum runs over all transitions $T_i$, which are added up incoherently. The simulation uses a non-relativistic description and thus contains only one transition, T1. The model can be used to describe asymmetric line shapes and the dipole phase shift $\phi_0$ is connected to the Fano asymmetry parameter $q$ via $\phi_0 = 2 \arg(q - i) [23]$. For $\phi_0 = 0$ it describes a symmetric Lorentzian profile. There are three possible, time-delay dependent fit parameters for each absorption line; the amplitude/line strength $z_0$, the dipole phase $\phi_0$ and the line width $\Gamma$. $\varrho L$ is the experimental pathlength-density product of the target gas (in atomic units) and $C(\tau)$ describes a global time-delay-dependent background to account for broadband, spectrally flat features stemming from the neutral resonances at higher energies [37].

The fit results for the experimental data for T1 are plotted in figure 3. The color coding of the lines represents the pump intensity which increases from blue to red. In panel (a) the delay-dependent line strengths, which are proportional to...
Figure 2. ATAS scan centered on the Xe\(^{+}\) transition \(T_1\) in the time-delay overlap region at an NIR intensity of \((1.91 \pm 0.21) \times 10^{14} \text{ W cm}^{-2}\).

(a) Measured data and (b) the corresponding reconstruction from the line-shape fits. (c) The same 4d–5p resonance as simulated with our \textit{ab initio} RMT approach and (d) the corresponding fit. Delay of probe with respect to pump, pump centered at zero. The data presented may be found at [36].

the effective valence-hole population, capture the build-up of the ion lines. The non-monotonic overshoots with half-cycle periodicity confirm the previous findings by Sabbar \textit{et al} [19]. The same half-cycle periodicity is observed in the simulation (not shown). The intensity-dependent evolution demonstrates how we scan from low intensities, just sufficient for enough spectroscopic ion signal for line-shape analysis, to a regime where single-ionization saturates in favor of double-ionization. The latter is also confirmed by the observation of Xe\(^{2+}\) ionic transitions (not shown) appearing above \(1.6 \times 10^{14} \text{ W cm}^{-2}\).

The RMT method does not have capability for double ionization, and so the simulated results become less reliable at higher NIR intensities.

Panels (b) and (c) for the dipole phase \(\phi_0(\tau)\) and the line width \(\Gamma(\tau)\) reveal the difficulties of the fitting procedure. When the ion population is low, the line strength \(z_0\) is correspondingly small, hence the remaining line-shape parameters are more difficult to quantify and increasingly susceptible to noise in this regime. To visualize this without the need of obstructive layers of error bars, the transparency of these plots is set to be inversely proportional to the line strength. The most remarkable observation from the line-shape fits is the course of the phase evolution, which similarly to the line strength reveals half-cycle oscillations on top of a systematic trend of an increasingly negative dipole phase when scanning from late to earlier time delays. In addition, the oscillation amplitude is intensity-dependent and decreases for higher intensities.

6. Discussion

To isolate the intensity dependence of the temporally oscillating dipole phase \(\phi_0\), we subtract the slowly varying cycle-averaged evolution from this phase. Figure 4 shows the resulting peak-to-peak differences \(\Delta \phi\) of the phase modulation for each pump intensity. The reported peak experimental intensities are inferred from average beam power with an uncertainty of up to 11\%, which may account for the systematic shift between simulation and experiment in figure 4. With additional RMT calculations (not shown) we have ascertained that focal-volume averaging does not change the reported results. In the following, we focus on this oscillatory feature of the line-shape asymmetry, because it is experimentally accessible and can be directly quantified for future benchmarks of multi-electron strong-field dynamics theory.

While the asymmetry of the absorption lines oscillates strongly with half-cycle periodicity at low pump intensities, the modulation is substantially reduced at the highest measured intensity of \(2.5 \times 10^{14} \text{ W cm}^{-2}\). We now estimate the relative abundance of the neutral and ionic species in the target by computing their path-length-density products \(\rho L = \text{OD} \ln(10)/\sigma\) from the optical density and the cross-section of a charge-state-specific transition following Beer–Lambert’s law. We identify the depletion of the neutral xenon ground state to dominate at the highest intensities, which is also in agreement with a saturation of the line strength \(z_0\) at and above \(2.33 \times 10^{14} \text{ W cm}^{-2}\) (figure 3). This estimated abundance of Xe\(^{+}\) in the experimental results agrees well with the values extracted directly from the RMT calculations, although there is a systematic overestimation in the simulations which may further account for differences in figure 4. However, by changing the independent variable from laser intensity to ground state abundance we may remove the systematic differences between the simulated and experimental results, and demonstrate unambiguously the physics behind the observed oscillations. We observe a near-perfect correlation (Pearson correlation coefficient \(r = 0.99, r_{\text{sim}} = 0.98\)) between the dipole phase oscillation amplitude and the ground-state abundance (see figure 5).
Figure 3. Fit results of the line-shape model (equation (1)) to the experimentally measured resonance T1 at different NIR intensities as denoted by the colors and the legend (intensity increasing from blue to red color). (a) Line strength $z_0$, (b) the dipole phase $\phi_0$ and (c) line width $\Gamma$ each as a function of XUV-NIR time delay $\tau$. The transparency shading in (b) and (c) encodes the extracted line strength. See text for further details and discussion.

Figure 4. (a) Measured, delay-dependent dipole phase $\phi_0(\tau)$ of transition T1 after subtracting the cycle-averaged trend. For better visibility the individual lines (ordered from higher intensity at the top to lower intensity at the bottom) are offset by 0.5 rad and plotting range of each line is limited to the region where the amplitude is greater than 5% of the maximum. (b) Amplitudes of the filtered phase modulation defined as $\Delta \phi = \max[\phi_0(\tau)] - \min[\phi_0(\tau)]$ from both measurement and simulation. The line and symbol colors represent NIR intensities as given in figure 3.

According to the theoretical work by Pabst et al [21] the non-perturbative dressing of the entire neutral N-electron system and in particular the mixing of the excited N-electron states with the neutral ground state in the combined XUV and NIR electric fields is the predominant cause of a non-zero dipole-phase shift. The excitation pathway corresponding to this dressing is sketched in figure 1(b).

By coupling to the same final state this non-perturbative pathway interferes with the direct, sequential SFI channel. A tractable mechanism to understand the observed line-shape changes is the consequence of transient couplings within the Fano picture [22, 23]. In the static helium atom, the interference of directly- and auto-ionizing pathways leads to an asymmetric Fano line shape in the doubly-excited states. Here, the time-dependent admixture of the neutral ground state through the two-color field leads to a transient coupling that distorts the ionic dipole only when the XUV pulse coincides with the maximum of an NIR field cycle. Hence, we observe a signature of the dipole phase of the Xe$^+$ ionic resonance with half-cycle periodicity, the strength of which is correlated with the neutral-state population. This non-sequential ionization pathway could be dubbed XUV-assisted SFI since it enables direct SFI of an otherwise inaccessible core-electron.

The interpretation that this mechanism is at the heart of the observed dipole-phase modulations is corroborated by its intensity dependence through the correlation with the neutral xenon abundance. Depletion of the neutral ground state impedes this pathway and thus, the interference amplitude decreases. This reasoning further confirms the theoretical findings by Pabst et al and extends the observations that were made on the overshoots in the line strength of the ion build-up during SFI [19] to now include the complementary phase information with attosecond resolution.
Figure 5. (a) Relative abundance of neutral xenon and the ionic species as a function of pump intensity. Estimations based on the line strength and cross sections \([35,38]\) of the following transitions: \(\text{Xe}: \, 4d^{10} - 4d_{5/2}; \, \text{Xe}^+: \, 5p_{1/2} - 4d_{1/2} (T_1), \, \text{Xe}^{2+}: \, 5p_{1/2} - 4d_{3/2}.\) \(\text{Xe}_{\text{sim}}\) is the neutral groundstate population extracted from the simulations. (b) Correlation between neutral xenon ground-state abundance and \(\text{Xe}^+\) resonant dipole-phase oscillation amplitude. The symbol colors represent NIR intensities as given in figure 3.

7. Conclusion

We present results of ATAS measurements on the pump-intensity- and delay-dependent ionization of xenon atoms. By fitting a line-shape model to the transitions in the ionic species \(\text{Xe}^+\) and \(\text{Xe}^{2+}\), insights into the ionization dynamics are extracted. This line-shape analysis reveals a modulation of the line-shape asymmetry with half-cycle periodicity with respect to the pump pulses and, specifically, a decrease in modulation strength of the dipole phase in near-perfect correlation with rising pump intensity. Along the lines of previous work \([19,21]\), we attribute this delay-dependent oscillation to the interference of non-sequential XUV-assisted SFI with the direct SFI and the subsequent excitation of core-hole excited states in xenon ions. In this context the intensity dependence of the observed feature is interpreted as the depletion of the neutral xenon ground-state which is the common starting point for both channels.

With complementary, delay-dependent phase information from the oscillations that are featured in both the line strength and dipole phase, a detailed investigation of their synchronicity can now be conducted on the attosecond timescale.

We expect attosecond delays of the interfering pathway of XUV-assisted SFI to be directly encoded in the sub-cycle timing of line-strength and dipole-phase evolution, hence opening a new direction for quantifying attosecond ionization delays \([39]\) with attosecond transient absorption spectroscopy.

Acknowledgments

The RMT code is part of the UK-AMOR suite, and can be obtained for free at reference \([40]\). This work benefited from computational support by CoSeC, the Computational Science Centre for Research Communities, through CCPQ. The authors acknowledge funding from the UK Engineering and Physical Sciences Research Council (EPSRC) under Grants EP/P022146/1, EP/P013953/1, EP/R029342/1, EP/T019530/1, and EP/V05208X/1. This work relied on the ARCHER2 UK National Supercomputing Service (www.archer2.ac.uk), for which access was obtained via the UK-AMOR consortium funded by EPSRC.

Data availability statement

The data that support the findings of this study are openly available at the following URL/DOI: https://doi.org/10.5281/zenodo.6683951.

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