

Attosecond Streaking of Correlated Two-Electron Transitions in Helium

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We present fully *ab initio* simulations of attosecond streaking for ionization of helium accompanied by shakeup of the second electron. This process represents a prototypical case for strongly correlated electron dynamics on the attosecond time scale. We show that streaking spectroscopy can provide detailed information on the Eisenbud-Wigner-Smith time delay as well as on the infrared-field dressing of both bound and continuum states. We find a novel contribution to the streaking delay that stems from the interplay of electron-electron and infrared-field interactions in the exit channel. We quantify all the contributions with attosecond precision and provide a benchmark for future experiments.

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With the development of light-wave synthesis with attosecond precision time-resolved investigations of ultrafast electronic dynamics in atoms, molecules, and solids on the atomic time scale came into reach [1–3]. Attosecond streaking is one of the most fundamental processes in *attosecond science* allowing for a mapping of time information onto the energy axis yielding a time resolution in the order of a few (tens) of attoseconds [4–7]. It is a variant of a pump-probe setting with an ultrashort extreme ultraviolet (XUV) pulse serving as pump and a phase-controlled few-cycle infrared (IR) field as probe [3]. First proof-of-principle studies addressed the direct time-domain measurement of the life time of the Kr($3d^{-1}$) hole by Auger decay of ≈ 8 fs [5] and the time-resolved photoemission from a tungsten surface by an energetic XUV pulse of ~ 300 as duration [6] providing insight into the relative time delay of ~ 110 as [6] (later ~ 85 as [8]) between photoemission from core levels relative to the conduction band. Even shorter delays were more recently determined in the elementary photoelectric effect where a time shift of the $2p$ relative to the $2s$ electron in neon as small as 21 as has been obtained in an attosecond streaking experiment by Schultze *et al.* [7]. These observations have triggered a flurry of theoretical investigations [7,9–16]. While time delays of atomic photoionization on the one-electron (or mean-field) level as well as additional effective time shifts due to the dressing of the outgoing photoelectron [11,12,14] (often referred to as “Coulomb-laser coupling” (CLC), [17]) could account for a delay of the order of 6 to 8 as, even the approximate inclusion of correlation effects failed to reproduce the experimentally observed delay [7,9,15,16]. Multielectron effects were studied in some detail in stationary scattering approaches, i.e., by analyzing the phase of the dipole transition matrix elements for photoionization in the absence of the streaking field. This *group delay* or Eisenbud-Wigner-Smith (EWS) delay (see, e.g., [18]) is not directly what is measured by a streaking experiment [11]. It was suggested that the probing IR field might be responsible for the larger delay

observed in the experiment [11,9]. There have been extensive time-dependent studies that simulated actual streaking experiments [11,12,15,16]. The role of electron correlations and its interplay with the laser field has remained an open problem. Recent work included some multielectron effects in the simulation of the full streaking setup, either with the *R*-matrix incorporating time method using a few-state representation of neon [15], or through a two-electron model system approximating the level structure of neon [16]. However, treating both the dressing of the atomic dynamics by the IR streaking field and the dynamical electron correlation exactly is still out of reach for neon but is possible for helium.

In this Letter we present streaking simulations for single ionization of helium including shakeup by solving the full two-electron time-dependent Schrödinger equation (TDSE) in the presence of the laser field without further approximation. In contrast to neon [7], where ionization from different atomic subshells was probed, shakeup ionization in helium relies strongly on electron-electron interactions. Here, the absorption of a single XUV photon leads to both emission of one electron and excitation of the other electron, which can only happen if the electrons share the photon energy and thus interact strongly. Studying ionization with and without shakeup may serve as benchmark for the interplay between mean-field, correlation, and field dressing effects.

We show that for ionization without shakeup the streaking time shifts can be very well accounted for within the framework of Coulomb-laser coupling and the Eisenbud-Wigner-Smith delay. For ionization accompanied with excitation of the second electron correlation becomes important as field dressing effects modify the electron-electron interaction in the exit channel and give rise to an additional apparent time shift. We quantify all the contributions with attosecond precision and provide a benchmark for future experiments.

In our computational approach (see, e.g., [19,20] for a detailed description) we solve the Schrödinger equation by

the time-dependent close-coupling method, [21–23]. For the radial discretization we use a finite-element discrete-variable representation (FEDVR) [24–26], and propagate in time using the short-iterative Lanczos (SIL) algorithm [27,28] with automatic time-stepping and error control. The laser fields are linearly polarized and treated in dipole approximation. The XUV pulse has a Gaussian envelope with a FWHM duration of 200 as and an intensity of $I_{\text{XUV}} = 10^{12}$ W/cm² (for which multiphoton processes can be neglected), while the 800 nm IR field has a sine-squared envelope with a FWHM duration of 3 fs and an intensity $I_{\text{IR}} = 4 \times 10^{11}$ W/cm². For these laser parameters, numerically converged streaking simulations are obtained with a partial wave expansion with total angular momenta up to $L = 8$ and one-electron angular momenta up to $l_{<} = 5$ and $l_{>} = 8$. We use an asymmetric radial box with an extension up to 960 a.u. in one direction and 96 a.u. for the other radial coordinate (which is large enough for ionic bound states up to $n = 8$ to be well represented). Each FEDVR element spans a length of 4.0 a.u. and contains a DVR of order 11. Atomic units are used throughout the paper.

Figure 1 shows the energy levels of helium and a corresponding single-ionization spectrum including shakeup peaks (“correlation satellites”). Because of the large difference in the binding energy between the $\text{He}^+(n = 1)$ and $\text{He}^+(n = 2)$ states of 1.5 a.u. (40.8 eV) the two peaks are well separated and resolvable [Fig. 1]. For a typical XUV pulse ($\hbar\omega = 100$ eV) with 200 as duration (≈ 9 eV spectral width), different shakeup channels (e.g., $n = 2$ and $n = 3$) are not resolved. However, numerically we have access to all channels separately. The electronic spectra for different delay times τ of the ionizing XUV pulse relative to the probing IR-field build up a *streaking spectrogram* [see Fig. 1(c)], where the spectra are shifted in momentum relative to the unperturbed asymptotic momentum \vec{p}_0 . The streaking spectrograms are obtained by projecting the propagated wave function $\psi(\vec{r}_1, \vec{r}_2, t)$ onto the single continuum constructed as a symmetrized product state of a *bound state* $\Phi_{n,l,m}(\vec{r})$ of the He^+ ion and a *Coulomb wave* $\psi_{\vec{k}}(\vec{r})$ with charge $Z = 1$ when the outgoing electron has reached sufficiently large distances.

The absolute time shifts t_S are extracted by a nonlinear least-squares fit of the modified final momentum $\vec{p}_f(\tau)$ of the different channels (taken as the first moment of the electron spectrum) [11],

$$\vec{p}_f(\tau) \approx \vec{p}_0 - \alpha \vec{A}_{\text{IR}}(\tau + t_S), \quad (1)$$

where α is a correction factor for the amplitude of the momentum shift induced by the streaking field. The resulting t_S contains information on the Eisenbud-Wigner-Smith (EWS) time delay t_{EWS} [29–31] of the atomic photoionization process in the absence of the streaking field as well as on the dressing of atomic and ionic states in the IR field. On the one-electron level the streaking time shifts t_S can be decomposed as [11,16]

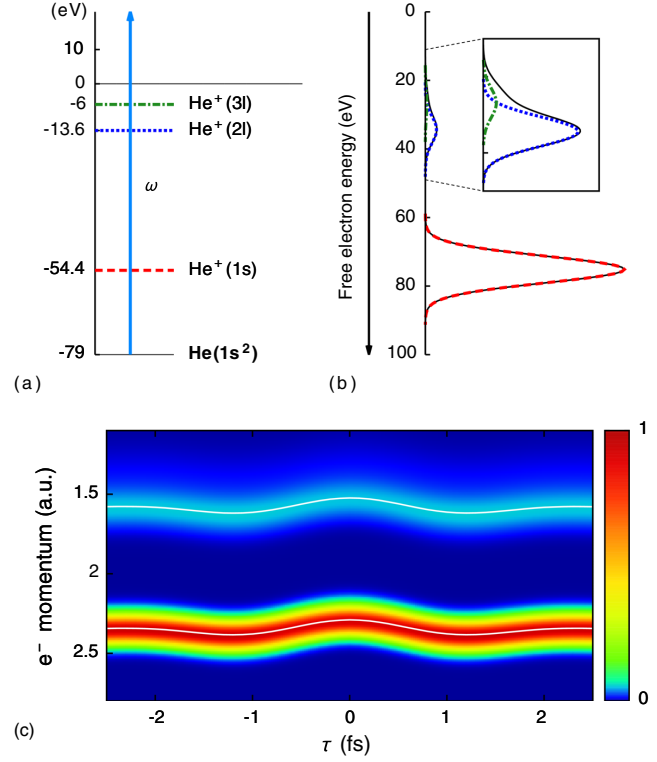


FIG. 1 (color online). Single-ionization of He: (a) energy levels of $(nl, \varepsilon l')$ final states; (b) electron energy distribution $P_{nl}^{\text{SI}}(\varepsilon, \theta = 0 \pm 10^\circ)$ created by a 200 as (FWHM) XUV pulse with mean energy $\hbar\omega = 100$ eV along the polarization direction with an opening angle of 10° . (c) Streaking spectrogram of (b) with a 3 fs streaking pulse with $\lambda = 800$ nm and intensity $I_{\text{IR}} = 4 \times 10^{11}$ W/cm², lower (high momentum) feature: shake-down, upper (low momentum): shakeup. Note the reversed direction of the energy (and momentum) axis in (b) and (c) to match the energy levels (a).

$$t_S = t_{\text{EWS}} + t_{\text{CLC}} + t_{\text{ISLC}}. \quad (2)$$

The EWS time delay t_{EWS} is given by the energy derivative of the phase of the dipole transition element $\langle \psi_f | \hat{z} | \psi_i \rangle$,

$$t_{\text{EWS}}(E) = \frac{\partial}{\partial E} \arg(\langle \psi_f(E, \theta = 0) | \hat{z} | \psi_i \rangle), \quad (3)$$

where E is the final continuum energy of the free electron, and the ejection angle θ is chosen along the laser polarization axis, as in the streaking spectrogram. The second term t_{CLC} is an apparent time shift due to the interaction of the outgoing electron with both the long-ranged Coulomb potential and the IR streaking field [11,12,14] which approximately scales as $\sim -E^{-3/2}$ [11,32]. We take $t_{\text{CLC}}(E)$ from the *reference* streaking shift $t_S^{\text{H}(1s)}(E)$ of the pure $Z = 1$ Coulomb potential at the asymptotic electron energy E ,

$$t_{\text{CLC}}(E) = t_S^{\text{H}(1s)}(E) - \frac{\partial}{\partial E} \sigma_1, \quad (4)$$

where we have subtracted the EWS delay of the Coulomb phase σ_1 in the final p continuum state for one-photon

ionization of H(1s). Alternatively, t_{CLC} can be extracted from a classical calculation [11]. The CLC time shifts are only weakly dependent on the streaking laser field parameters, in particular, the wavelength of the IR field [12] and the duration of the XUV pulse.

For strongly polarizable initial bound states another apparent time shift t_{ISLC} was found due to initial-state laser coupling [11,33]. Energy shifts of the initial state due to the interaction with the laser field are at the moment of ionization transferred to the final energy, and thus appear as apparent time shifts.

A novel scenario appears for true multielectron processes beyond the single-active electron (SAE) or mean-field level. We consider the prototypical two-electron process, the photoionization of He with shakeup or shakedown, where electron correlation is expected to play a significant role. Figure 2 shows the streaking time shifts t_S for ionization with and without shakeup in comparison with the prediction $t_{\text{CLC}} + t_{\text{EWS}}$ [Eq. (2)]. Initial-state distortions t_{ISLC} are negligible for the helium ground state He(1s²). For shakedown to the ground state in the ionic system He⁺(1s), i.e., direct ionization without shakeup, the experimentally accessible streaking shift t_S agrees remarkably well with Eq. (2). To calculate exact dipole matrix elements for single ionization with the correct boundary conditions even above the double ionization threshold Eq. (3), we use the extraction method of Palacios *et al.* [34] based on exterior complex scaling and apply it to the wave packet $\hat{z}|\phi_0\rangle$. For comparison we have also calculated the EWS time shift within the Hartree (mean-field) approximation where we take the ionic electron distribution to create an effective one-electron potential. When the bound electron is left in the ionic ground

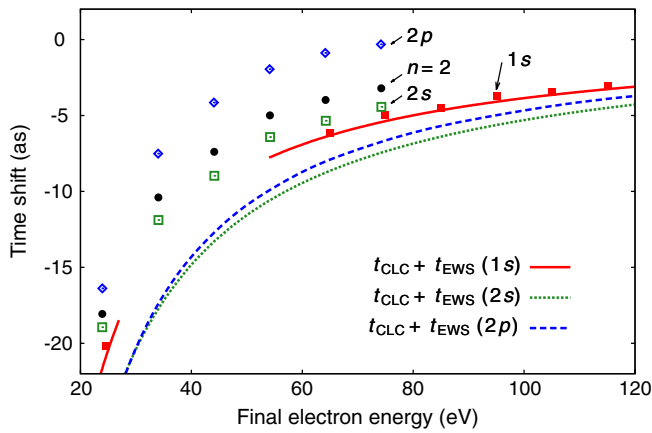


FIG. 2 (color online). Temporal shifts t_S (■: 1s, □: 2s, ◇: 2p, ●: $n = 2$) extracted from quantum mechanical streaking simulations and shifts predicted by Eq. (2), $t_{\text{CLC}} + t_{\text{EWS}}$ [lines, with $t_{\text{ISLC}} = 0$ for He(1s²)] for single ionization of helium into an opening angle of 10° with respect to the polarization axis with and without shakeup of the second electron as a function of the final electron energy. Time shifts that belong to the same XUV energy are thus shifted by $I_1^{(2)} - I_1^{(1)} = 40.8$ eV. Note that in the spectral region of resonances ($35 \text{ eV} \leq E \leq 54.4 \text{ eV}$ for an ionic 1s state), streaking time shifts are not well defined.

state, also the mean-field values $t_{\text{EWS}}^{\text{HF}}$ agree with the exact t_{EWS} to within less than one attosecond (not shown).

A surprisingly different picture emerges for shakeup to $n = 2$ (2s and 2p) where Eq. (2) fails. The streaking time shifts for shakeup predict a delay with respect to shakedown for all investigated photon energies which can not be accounted for by the corresponding EWS delays t_{EWS} . Obviously, the interplay between electron-electron interaction and the IR streaking field strongly influences the streaking delay of the outgoing wave packet. The additional time delay can be viewed as the result of the coupling of the dipole moment of the shaken-up ionic state to the streaking field. Because of the entanglement of the system, the outgoing electron still contains information on the ionic state. For the almost-degenerate n manifolds, this effect should be maximal for intershell eigenstates of the dipole operator, i.e., Stark states (nk) with $n = 2$ and $k = \pm 1$. Indeed, the resulting time delays (see Fig. 3) are strongly enhanced ($k = 1$) and diminished ($k = -1$) compared to ionic final states with well-defined angular momentum quantum number. There are two effects worth noting: First, the potential seen by the receding electron is modified by $V_D(\vec{r}) = \vec{d} \cdot \vec{r}/r^3$, where \vec{d} is the dipole

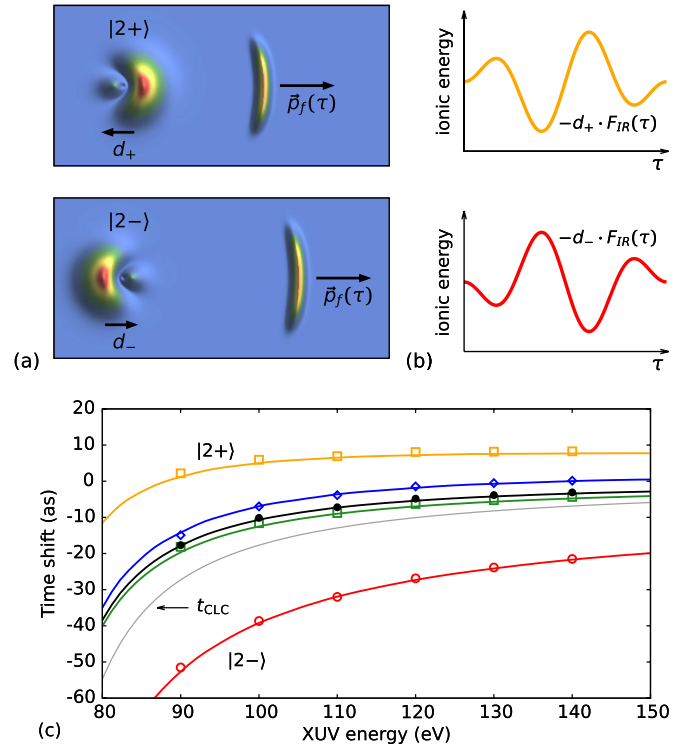


FIG. 3 (color online). (a) Illustration of the backaction of the shakeup Stark state on the emitted electron and (b) the ionic energy in the laser field which leads to an additional momentum shift of the outgoing electron. (c) Streaking time shifts t_S (points) along the forward laser polarization direction for the two ionic parabolic states ($n = 2, k = \pm 1$) in comparison with $n = 2, 2s$ and $2p$ states (see Fig. 2). In addition we show the complete prediction of t_S according to Eq. (5), $t_{\text{CLC}} + t_{\text{EWS}} + t_{\text{CLC}}^{e-e}$ (solid lines).

moment of the Stark state. However, this time shift is already included in the (field-free) EWS delay and thus gives no additional measurement-induced time shift. Second, creation of a shaken-up electron in a Stark state in the presence of the IR field entails an additional energy contribution $-\vec{d} \cdot \vec{F}_{\text{IR}}(t)$ of the ionic system (see Fig. 3). This time-dependent energy shift of the *bound* electronic final state of the residual ion becomes thus visible in the energy and momentum distribution of the streaked *ionized* electron. Since this energy shift is proportional to $\vec{F}_{\text{IR}}(t)$ it appears as an additional time shift $t_{\text{CLC}}^{e-e(2\pm)} = \text{atan}(-\omega_{\text{IR}} d_z^{\pm}/k_z)/\omega_{\text{IR}}$. This process can be viewed as a two-electron generalization of the streaking shift for one-electron states with permanent dipole moment first discussed in [10]. For final ionic Stark states the streaking delay $t_S^{(2\pm)}$ is indeed given to remarkable accuracy by the generalization of Eq. (2),

$$t_S = t_{\text{EWS}} + t_{\text{CLC}} + t_{\text{ISLC}} + t_{\text{CLC}}^{e-e} \quad (5)$$

(with $t_{\text{ISLC}} = 0$ for the $\text{He}(1s^2)$ initial state). This streaking-field induced two-electron effect also causes the additional time shifts for final ionic states with well-defined angular momentum, s and p , i.e., states with a negligible dipole moment due to inversion symmetry. If we expand the final ionic states into Stark states, $|2s/p\rangle = 1/\sqrt{2}(|2+\rangle \pm |2-\rangle)$ we can find an effective dipole moment for the final two-electron states,

$$d_{z,\text{eff}}^l = (d^+ |c_+|^2 + d^- |c_-|^2)/(2|c_l|^2), \quad (6)$$

where $c_\alpha = \langle 2\alpha, E\theta_0 | \hat{z} | \psi_i \rangle$. Note that the nonzero dipole moment in Eq. (6) is consistent with the inversion symmetry of the ionic *bound* state as this symmetry is broken by the selection of the emission direction of the streaked *continuum* electron. Even if the final state of the remaining ion is not detected, i.e., the time shift for the total $n = 2$ shell is measured, an averaged effect in the order of 5 as is visible (see Fig. 3). Thus probing the momentum spectrum (or energy shift, respectively) of the ionized electron contains information on the dynamics of the entangled ionic state.

We have thus identified an additional time shift t_{CLC}^{e-e} resulting from the back action of the excited bound state onto the continuum wave packet in the presence of the IR streaking field. Similar to the corresponding initial-state distortions, this additional time delay is not (or only weakly) dependent on the intensity of the IR field I_{IR} (within the range of useful intensities $10^{10} \text{ W/cm}^2 \leq I_{\text{IR}} \leq 10^{12} \text{ W/cm}^2$), cf. [11]. Thus, for photoionization of two-electron systems with shakeup, the total streaking time shift [Eq. (5)] contains an additional contribution due to simultaneous excitation of the second electron. Attosecond streaking phases can therefore reveal information on dynamical polarization in two-electron transitions. This information on the time-resolved bound-state excitation complements alternative techniques of transient x-ray

absorption spectroscopy [35] and transient bound-state excitation by shaped two-color laser fields [36].

We conclude by discussing possible experimental observations. While the streaking spectrogram for the final ground state, $\text{He}^+(1s)$, is well separated from that of two-electron excitation-ionization (Fig. 1), unambiguous observation of the latter requires the separation of the $n = 2$ from higher shells with $n \geq 3$. This would require an XUV pulse with a Fourier-limited width corresponding to a duration of $T_{\text{XUV}} \geq 500$ as (we have checked that our numerical results change within less than 1 as compared to the 200 as used above). Alternatively, measuring Ly_α photons resulting from the radiative decay of the excited $n = 2$ states in coincidence would allow to separate excitations of the $n = 2$ shell. Moreover, excitations of the $2s$ and $2p$ states could be separated by prompt vs delayed Ly_α coincidences in a weak DC electric field. A simple proof-of-principle experiment would be to monitor the large delay of shakeup into the $n = 2$ subshells, or an ensemble of excited manifolds ($n \geq 2$), with respect to streaking without shakeup ($n = 1$) as a function of the XUV energy (Fig. 4). This would give rise to time advances up to 12 as when the $n = 2$ signal can be separated from higher shakeup states and up to 27 as when the total electron spectrum for $n \geq 2$ is analyzed. The large difference in the delay for $n = 2$ and $n \geq 2$ shows how sensitive the extracted streaking time shifts are to small contributions of different shakeup states in the streaking spectrum [see inset in Fig. 1(b)]. The strong photon energy dependence of the relative delay between $n = 1$ and $n > 1$ is due to the variation of t_{CLC} with the final-state energy E . The ensemble of excited manifolds with $n \geq 2$ would correspond to the streaking delay between the two peaks of the total ionization spectrum [see Fig. 1(b), black line] which should be relatively straightforward and could serve as benchmark for the precision of the experimental streaking techniques.

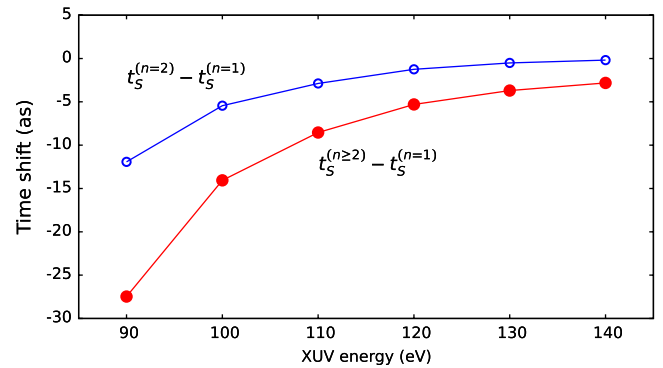


FIG. 4 (color online). Relative total temporal shifts $t_S^{(n>1)}(\omega) - t_S^{(n=1)}(\omega)$ between ionization with and without shakeup of the second electron extracted from quantum mechanical streaking simulations for helium for different XUV energies, for shakeup into $n = 2$ (blue line, open points) and sum of all shakeup states ($n \geq 2$, red line, solid points), lines guide the eye.

In summary, we have shown that for photoionization of helium streaking time shifts provide detailed information on the Eisenbud-Wigner-Smith time delay as long as Coulomb-laser coupling as well as laser-induced state distortion effects are accounted for. For two-electron excitation-ionization the interplay of electron-electron and IR-field interaction in the exit channel leads to additional and novel contributions to the time shift of the outgoing wave packet. We show that dynamical correlation effects can play a significant role for attosecond streaking experiments. Our theoretical *ab initio* results can serve as an accurate benchmark for experimental attosecond streaking setups.

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