Attosecond pulses can ionize atoms in a coherent process. Since the emerging fragments are entangled, however, each preserves only a fraction of the initial coherence, thus limiting the chance of guiding the ion subsequent evolution. In this work, we use \textit{ab initio} simulations of pump-probe ionization of helium above the $2s/2p$ threshold to demonstrate how this loss of coherence can be controlled. Thanks to the participation of $2\ell n\ell'$ states, coherence between the ionic $2s$ and $2p$ states, which are degenerate in the non-relativistic limit, results in a stationary, delay-dependent electric dipole. From the picosecond real-time beating of the dipole, caused by the fine-structure splitting of the $n = 2$ manifold, it is possible to reconstruct all original ion coherences, including between antiparallel-spin states, which are sensitive probe of relativistic effects in attosecond photoemission.

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Electronic motion in atomic systems occurs on a sub-femtosecond timescale \cite{1–3}. Attosecond XUV-pump IR-probe photoelectron spectroscopies have thus emerged as powerful tools to explore charge-transfer processes in complex systems \cite{4–8} and attosecond dynamics at the nanoscale \cite{9, 10}. The short duration of attosecond pulses generates coherent superposition of electronic states above the ionization threshold, bearing the promise of quantum control in the electronic continuum \cite{11, 12}. Since the photoelectron and ion form an entangled pair, however, as soon as the photoelectron leaves the interaction region, part of the coherence in the residual parent-ion is lost, and so is the chance of guiding any subsequent transformations of the target in a reproducible way. One way to limit the loss of coherence that accompanies photoionization is to polarize the target with a strong control field that forces the ion in a single polarized state \cite{13, 14}. In a theoretical study of the one-photon ionization of xenon, which can result in partial coherence between ions with the same parity, Pabst \textit{et al.} have shown this coherence increasing for pulses with shorter duration and higher central frequency, on account of the reduced role of inter-channel coupling at large photoelectron energies \cite{15, 16}. A more general control of the entanglement between photofragments can be achieved by leveraging the interference between different multi-photon ionization (MPI) paths \cite{17, 18}. In this latter approach, autoionizing resonances play a crucial role as intermediate states since they decay on a longer timescale than free photoelectron wavepackets \cite{19–24}. In fact, metastable states are essential intermediates in resonant multi-photon atomic ionization \cite{25, 26}, ultrafast electron decay \cite{27} and molecular dissociative photodissociation \cite{28, 29}.

In this work, we use \textit{ab initio} simulations to explore the control and reconstruction of the density matrix of the ensemble of $2s$ and $2p$ parent ions that emerge from the shake-up ionization of the helium atom with an XUV-pump IR-probe sequence of ultrashort pulses, linearly 
polarized along the $z$ axis [22, 24, 30]. Multi-photon excitations are key to entangle the 2$s$ and the 2$p$ states, which have opposite parity. In pump-probe ionization of helium, autoionizing states below the N=2 threshold are known to affect the branching ratio between shake-up channels [19], due to the interference between direct-ionization and resonant MPI paths. This same interference affects also the residual coherence between the 2$s$ and 2$p$ states of the He$^{+}$ ion. In the non-relativistic limit, these states are degenerate, and hence their coherence results in a permanent dipole moment. We demonstrate that the magnitude of the polarization can be controlled by changing the pump-probe delay. On a timescale of few picosecond, the dipole moment fluctuates even in absence of external fields, due to spin orbit interaction [31]. Our results show how the slow dynamics of such polarized-ion ensemble can be controlled with attosecond precision. Conversely, from these fluctuations, it is possible to reconstruct the relative phase between the 2$s_{m_s}$ and 2$p_{m_p,m_s}$ states in the ion wavepacket at the time of its inception. In particular, the method gives access to the coherence between the 2$s_{1/2}$ and the 2$p_{1,-1/2}$ states, which is a sensitive probe of relativistic effects in attosecond ionization, since it vanishes only in non-relativistic limit.

Figure 1a illustrates the pump-probe excitation process we simulate. A weak single attosecond XUV pulse, with 60.48 eV energy and a duration of 545 as, excites the neutral helium atom from the ground state to the N=2 shake-up ionization channels above the threshold, as well as to the DES below N=2 ionization threshold. The $sp_{2}^{+}$ and $sp_{3}^{+}$ states [32–36], which are 5.04 eV and 1.69 eV below the N= 2 threshold with a lifetime of 17.6 fs and 80.3 fs, respectively, are populated most efficiently [19]. The absorption of a single XUV photon cannot give rise to any coherence between ionic states with opposite parities, such as 2$s$ and 2$p$. An IR-probe pulse with 1.55 eV energy, 5.33 fs FWHM, and a controllable delay dresses the system at the time of the excitation and promotes the DES to shake-up ionization channels, above the N=2 threshold. Thanks to the presence of several interfering multi-photon processes, a coherence between degenerate 2$s$, 2$p$ ionic states now emerges. We simulate this process by solving the time-dependent Schrödinger equation for the atomic system in the presence of the external pulses is integrated with a unitary second-order propagator in a B-spline close-coupling basis [19, 22, 24]. Partial photoelectron amplitudes are computed at the end of the pulse by projecting the wavefunction in interaction representation on a complete set of multi-channel scattering states for the two-electron system [19, 33, 37], as a parametric function of the pump-probe delay $\tau$, $A_{\alpha\alpha\sigma}(\tau) = \langle \psi_{\alpha\alpha\sigma}^{\rightarrow} | \Psi_{f}(t; \tau) \rangle$, where $\alpha$ identifies the state of the parent ion, while $\vec{k}$ and $\sigma$ are the asymptotic photoelectron momentum and spin projection. The reduced density matrix for the parent ion, $\rho_{\alpha\beta}(\tau)$, is obtained tracing out the photoelectron quantum states $[38]$

$$\rho_{\alpha\beta}(\tau) = \sum_{\sigma} \int d^{3}k A_{\alpha\alpha\sigma}(\tau) A_{\beta\beta\sigma}^{*}(\tau).$$

The coherence between ionic states [15, 38] is defined here as $g_{\alpha\beta}(\tau) = \rho_{\alpha\beta}(\tau)/\sqrt{\rho_{\alpha\alpha}(\tau)\rho_{\beta\beta}(\tau)}$.

Figure 1c-e show the ion electron density immediately after the ionization event, for a pump-probe delay $\tau$ of 0, 1, and 2 fs, respectively, computed from the ab initio density matrix $\rho_{\alpha\beta}(\tau)$. The residual coherence results in a controllable polarization of the ion. Within...
the non-relativistic approximation, the 2s and 2p states are degenerate, and hence their dipole moment is stationary. On the femtosecond timescale of the present simulation for helium, the non-relativistic approximation is expected to be valid. On longer timescales, however, relativistic interactions can no longer be neglected. Spin-orbit coupling splits the 2p level into a 2P_{1/2} and 2P_{3/2} multiplet [39], and Lamb shift lowers the energy of the 2S_{1/2} level compared to 2P_{1/2} [40, 41], see Figure 1b. Due to these relativistic interactions, gathered in the fine-structure Hamiltonian \( H_f \), the density matrix undergoes periodic oscillations on a picosecond timescale,

\[
\rho(t; \tau) = e^{-iH_f t} \rho(\tau)e^{iH_f t},
\]

and so does the ion dipole moment, \( \langle \mu_z(\tau, t) \rangle = Tr[\rho(\tau)\mu_z(t)] \).

Figure 2.a shows the absolute value of the coherence between the 2s and 2p_{\pm} states as a function of the pump-probe delay. In the region where the two pulses overlap, ionization takes place in the presence of the IR probe pulse, which suppresses the channel in which the ion is polarized opposite to the IR field. As a consequence, the ion emerges strongly polarized, giving rise to the macroscopic polarization of the residual charge density shown in the inset and mentioned in reference to Figure 1. The density fluctuates with the same frequency as the IR period, whereas coherence is maximum every half IR period, near the peak of the IR. When the two pulses do not overlap, beyond 4 fs time delay, the coherence exhibits weaker modulations due to the beating between the MPI amplitudes from the multiple intermediate doubly-excited states below threshold. The change in the charge density can be better appreciated from the left-right density asymmetry. Figure 2.b shows the window Fourier transform of the dipole moment with respect to the time delay,

\[
\tilde{\mu}(\tau_w, \omega_\tau) = \frac{1}{\sqrt{8\pi^3\sigma_w}} \int d\tau e^{i\omega_\tau \tau - (\tau - \tau_w)^2/2\sigma_w^2} \mu(\tau),
\]

where \( \sigma_w = 2.4 \) fs, which features clear peaks, as a function of \( \omega_\tau \), each corresponding to the beating between a pair of doubly excited states. The spectrum is dominated by the beating between the pair of doubly excited states \( sp_{2}\pm -sp_{3}\pm, sp_{3}\pm -sp_{4}\pm, sp_{4}\pm -sp_{5}\pm, \) and \( sp_{5}\pm -sp_{6}\pm \), which in the Figure are labelled 2 - 3, 2 - 4, 3 - 4, and 4 - 5, respectively. The beating with the \( sp_{5}\pm \) state decay more rapidly than the others, due to the short lifetime of this state (~18 fs). All peaks exhibit strong modulations as a function of the window central delay, which shows that the pump-probe delay can be used as a femtosecond knob to control the degree of coherence of the ion.

The splitting of the \( n = 2 \) level causes the electric dipole to oscillate in real-time, on a picosecond timescale. Figure 3a shows the ionic dipole as a function of both pump-probe delay and real time. When the pump and probe overlap, the dipole fluctuates with a period of \( \sim 6 \) ps, with its phase flipping periodically between 0 and \( \pi \), giving rise to a checkerboard structure. Since the \( N = 2 \) manifold splits into three levels, the real-time beating contains two distinct frequencies, \( 24.5 \times 10^{-6} \) a.u. and \( 2.1 \times 10^{-6} \) a.u. [40]. From the picture, only the faster beating is clearly visible, since it is considerably stronger than the other. Furthermore, the longer period, \( \sim 72 \) ps, is close to a multiple of the faster period, of 6 ps, which reduces its visibility further. Nevertheless, from the Fourier Transform of the signal, both components can be accurately retrieved. The 6 fs beating dominates the real-time evolution of the dipole even when the pump and probe pulses do not overlap. In contrast to the overlapping case, the phase of the oscillation now changes gradually as a function of the pump-probe delay. Indeed, in this time-delay range, the ion coherence originates from resonant multiphoton interferences. As a result, the relative phase of the DES, which is encoded in the beating of the ion’s permanent dipole shortly after the end of the pulse sequence, as a function of the pump-probe delay, is reflected, stretched by three orders of magnitude, on the real-time oscillation of the ion dipole on the picosecond timescale. Figure 3b shows the real and imaginary part of \( \rho_{2s\sigma, 2p_{\pm}\sigma} \), computed at the end of pulse, which both dictates and can be retrieved from the long-time evolution of the dipole under the effect of fine-structure interactions.

The present excitation scheme has a duration of few tens of femtoseconds, i.e., two orders of magnitude smaller than the spin precession period caused by the fine-structure splitting. As long as the electron spin does not affect the excitation process, therefore, the dipole expectation value at the end of the pulses is dictated only by the coherence between the \( 2s_\sigma \) and \( 2p_{0,\sigma} \) states (the coherence is the same for \( \sigma = \pm 1/2 \)), whereas the coherence between the \( 2s_\sigma \) and the \( 2p_{0\sigma}, -\sigma \) states is zero. At larger times, the non-stationary character of the \( 2p_{0\sigma} \) configurations emerges, and the dipole moment is observed to oscillate. When the fine-structure is taken into account, the time dependence of the dipole moment is dictated by two independent non-vanishing coherences, between the \( 2S_{1/2,1/2} \) state and the two \( 2P_{1/2} \) states, for \( j = 1/2 \) and \( j = 3/2 \), which beat at different frequencies, and can hence be separately measured,

\[
\langle \mu_z(\tau, t) \rangle \propto \sum_{j=\frac{1}{2},\frac{3}{2}} P_j(\tau) \cos(\omega_j t - \phi_j(\tau)),
\]

where \( \omega_j = E_{2P_j} - E_{2S_{1/2}} \). Neglecting the small differences in their radial wave functions we can write the \( 2P_{1/2} \) and \( 2P_{3/2} \) fine-structure states in terms of the \( 2p_{0\sigma} \) spin orbitals just by coupling the orbital and spin angular momentum, \( |2P_{\mu}\rangle = \sum_{m\sigma} |2p_{m\sigma}\rangle C^{\mu}_{m\sigma} |1m,\frac{1}{2}\sigma\rangle \), where \( C^{\gamma}_{aa,bb} \) are Clebsch-Gordan coefficients [31, 39]. The spin-free character of the ultrashort excitation process, there-
Where we have used the fact that the zero elements of the density matrix between 2s and 2p states of the H^+ ensemble with a dominant period of ~ the fluctuation of the dipole moment of the N=2 level evolves in time, due to relativistic effects, resulting in the real (blue-solid) and imaginary (purple-dashed) part of the \( \rho_{2s,2p_{0}} \). This circumstance allows us to predict, from the simulation. The real and imaginary components reconstructed from the FT of the real-time oscillation of the dipole are in perfect agreement with the simulated results and hence they are not shown.

Therefore, causes the coherence between the \( |2S_{1/2,1/2}\rangle \) and \( |2P_{1/2,1/2}\rangle \) states to be in geometrical fixed proportion to the coherence between the \( |2S_{1/2,1/2}\rangle \) and \( |2P_{3/2,1/2}\rangle \) states. This circumstance allows us to predict, from the ab initio spin-free attosecond pump-probe simulations, the time evolution of the dipole at large times. Conversely, from the phases and the relative amplitude of the dipolar beatings on the picosecond time scale, regardless if measured or simulated, it is possible to reconstruct the relative amplitude and phases of the coherences in the \( \{2s_{\sigma},2p_{\sigma'\sigma'}\} \) basis, at the end of the ultrashort sequence,

\[
\rho_{2s,2p_{\sigma'\sigma'}} = \sum_{j=\pm 1/2} C_{1\sigma-\sigma',1/2}^{j\sigma} \rho_{2S_{1/2},2p_{j,\sigma'}} \tag{5}
\]

where we have used the fact that \( m = \sigma - \sigma' \), since the z component of the total angular momentum (orbital plus spin) is conserved. The off-diagonal terms \( \rho_{2S_{1/2},2p_{j,\sigma'}} \) are related to the observable beating parameters, \( \rho_{2S_{1/2},2p_{j,\sigma'}} = P_{j} e^{i\phi_{j}} / \mu_{2p_{j,\sigma'}} \). To check the self-consistency of this method, we have used it to reconstruct, from the long-time dipole fluctuation, the complex \( \rho_{2s,2p_{\sigma'\sigma'}} \) coherences. The non-vanishing quantity \( \rho_{2s,2p_{0}} \) so retrieved coincides with the one directly computed from the ionization wave function at the end of the pulse, plotted in Figure 3b. Our ab initio codes, which is based on the non-relativistic approximation, predicts the ratio \( R = \rho_{2s,2p_{0}} / \rho_{2s,2p_{0}} \) to be zero. Our numerical reconstruction of this ratio from the asymptotic dipole beating recovers also this ab initio prediction, thus confirming the accuracy of the reconstruction procedure. Conversely, \( R \) is expected to vanish only on the basis that spin-orbit coupling and other fine-structure interactions do not intervene in the ultrafast ionization process. An experimental measurement of \( R \), therefore, would open a new sensitive window on relativistic effects in attosecond ionization.

Is it possible to gain experimental access to the relative amplitude and phase of the picosecond dipole beatings? The ps dipole signal lies in the microwave range, and it can, in principle, be measured using microwave spectroscopy [42, 43]. Since the optical density of the ionic ensemble is expected to be extremely small, however, measuring dipole oscillations with purely optical methods would likely represent a major challenge. An alternative method to measure the coherence of the 2s and 2p states is to map it to the population of the N=3 level by means of a combination of the 2nd and 3rd harmonics of the IR probe pulse, together with a delayed 5th harmonics. These transitions require a temporal resolution of about one picosecond, and hence their synchronization is not as challenging as the attosecond synchronization between the initial pump and probe pulses. By changing the delay between \( 2nd+3rd \) and 5th harmonics, it is possible to change the total population transferred to the N=3 level in a well-known way. A final, intense IR pulse ionizes the N=3 states, whose population is finally measured by detecting the doubly-charged ion signal. The details of these possible experiments are beyond the scope of the present theoretical investigation.

In Conclusion, we have shown that the multiphoton ionization of Helium gives rise to 2s/2p coherence that can be controlled with the pump-probe delay, on a femtosecond time scale. When the two pulses overlap, a strong polarization of the ion is caused by the strongly polarized IR. A weaker modulation of the charge density exists due to beating between intermediate DES even when there is no overlap between the pulses. We also demonstrate that the spin-orbit splitting causes a slow evolution of the dipole that maps the phases of DES on a picosecond time scale. This slow evolution allows us to reconstruct the relative amplitude and phases of the off-diagonal terms in the density matrix at the time of ionization. This reconstruction protocol not only gives access to the coherent state of the ion. It also offers a way to measure the coherence between states with anti-parallel spin projection at the time of the ionization, which quantifies the effect of relativistic interactions on attosecond photoemission processes.
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