Attosecond electron–spin dynamics in Xe 4d photoionization

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The photoionization of xenon atoms in the 70–100 eV range reveals several fascinating physical phenomena such as a giant resonance induced by the dynamic rearrangement of the electron cloud after photon absorption, an anomalous branching ratio between intermediate Xe+ states separated by the spin-orbit interaction and multiple Auger decay processes. These phenomena have been studied in the past, using in particular synchrotron radiation, but without access to real-time dynamics. Here, we study the dynamics of Xe 4d photoionization on its natural time scale combining attosecond interferometry and coincidence spectroscopy. A time-frequency analysis of the involved transitions allows us to identify two interfering ionization mechanisms: the broad giant dipole resonance with a fast decay time less than 50 as, and a narrow resonance at threshold induced by spin-flip transitions, with much longer decay times of several hundred as. Our results provide insight into the complex electron-spin dynamics of photo-induced phenomena.

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The absorption of X-rays by matter has been used since more than a century ago to understand the structure of its fundamental constituents. An X-ray photon absorbed by an atom triggers multiple electron dynamics. The emission of an electron from an inner shell is accompanied by ultrafast rearrangement of the electronic cloud, which simultaneously modifies the potential seen by the electron, sometimes leading to resonances in the emission spectrum. An outer-shell electron may fill an inner hole, while another electron is emitted, a process called Auger decay. Finally, the electron spin may be affected by the field induced by the ultrafast orbital motion, giving rise to spin flip, which is forbidden for purely electric dipole transitions. All of this complex hole or electron motion occurs on a rapid time scale, in the attosecond (1 as = \(10^{-18}\) s) range.

The interaction of xenon atoms with photons in the 70–100 eV range illustrates many aspects of the electron dynamics sketched above. Collective many-electron effects in the 4d shell\(^{13,14}\) lead to a broad “giant dipole” resonance in the photoionization cross-section, which is maximum at 100 eV\(^{7,8}\). Photoionization is accompanied by Auger decay, involving the 5s and 5p shells, leading to the formation of Xe\(^{2+}\) ions\(^9\) (see Fig. 1). Relativistic (spin–orbit) effects can be observed at threshold (in the 70–75 eV region), with, in particular, an anomalous branching ratio between the \(2D_{3/2}\) and \(2D_{5/2}\) final states of the ion, separated by a spin-orbit splitting of 2 eV\(^{10–12}\).

Attosecond pulses produced by high-order harmonic generation in gases\(^{13,14}\) enable measuring ultrafast electron dynamics, as shown in a series of seminal experiments\(^{15–22}\). Temporal information is obtained by pump/probe techniques combining attosecond pulses and a synchronized laser field. The reconstruction of attosecond beating by interference of two-photon transition (RABBIT) technique\(^{23}\), based on interferometry, allows the determination of the photoionization spectral amplitude in the complex plane. The temporal dynamics is then obtained by Fourier transform or more generally by time-frequency analysis\(^{16,24}\). This technique has been successfully used to measure photoionization time delays, due to electron propagation in the potential following the absorption of an extreme ultraviolet (XUV) photon. Most of these studies\(^{25–28}\), however, have concentrated on relatively simple systems, ionized from the valence shells.

In this work, we present measurements of photoionization time delays in the Xe 4d shell for different ionic states \(4d^{-1}(2D_{3/2})\) and \(4d^{-1}(2D_{5/2})\), denoted \(4d_{3/2}\) and \(4d_{5/2}\) in the following (see Fig. 1b). Auger-photoelectron coincidence spectroscopy is used to disentangle electrons from different photoionization and decay channels\(^{29}\). The RABBIT interferometric technique allows the extraction of a phase, or a time (or group) delay, from the photoelectron spectra. At high photon energy (between 80 and 100 eV), both \(4d_{3/2}\) and \(4d_{5/2}\) photoelectrons are emitted with the same positive time delay. Close to the 4d-ionization threshold (75–80 eV), the measured time delays differ by more than 100 as. Supported by relativistic random phase approximation (RRPA) theoretical calculations\(^{30}\), we show that this difference is due to the interference of the broad giant dipole resonance with a narrow threshold resonance due to relativistic spin-orbit effects.

### Results

The experiments were performed with attosecond pulse trains generated in neon by a femtosecond Ti:sapphire laser system, covering a spectral range from the 4d ionization threshold to the maximum of the giant dipole resonance (see Methods for details). A small fraction of the infrared (IR) laser beam was used as a probe with a variable time delay. The XUV and IR pulses were focused into Xe gas and the created electrons were detected by an electron spectrometer.

Photoionization to different ionic states followed by Auger decay produces a complex electron spectrum, with two sets of photoelectrons separated by 2 eV (see Fig. 1). Single Auger decay from Xe\(^+\) (e.g., \(4d_{5/2}\) to \(Xe^{2+}\) (e.g., \(5s^{-5p^{-1}}\)) leads to electrons at kinetic energies equal to the difference between intermediate and final state energies, spanning from 8.3 eV to 36.4 eV\(^{31}\) and thus overlapping with the photoelectrons ionized by 75–100 eV photons\(^{32}\). Figure 2 shows XUV-only (a) and XUV+IR (b) two-dimensional coincidence maps. For a given final state of Xe\(^{2+}\), Auger electrons detected in coincidence with photoelectrons contribute to a stripe with discrete spots related to absorption of different harmonics (with odd orders 53 to 63 in the figure), or absorption of harmonics and absorption or emission of an IR photon (sidebands 54 to 62). In addition, weak signals due to absorption or emission of an IR photon by the Auger electron, are observed (see, e.g., the difference between the blue and red curves in Fig. 2e at 9.8 eV). This coincidence technique requires long acquisition times, but allows us to disentangle unambiguously the \(4d_{3/2}\) and \(4d_{5/2}\) photoelectrons by the energy of the Auger electron.

Each sidband arises from the interference between two quantum paths as illustrated at the top of Fig. 2(d). The sidband signal oscillates as a function of the delay \(\tau\) between the attosecond pulse train and the probe IR field, according to,

\[
I_{SB} = A + B \cos(2\omega \tau - \phi),
\]

where \(A\) and \(B\) are constants, \(\omega\) is the IR frequency and \(\phi\) is a phase offset, which can be extracted by fitting with a cosine function. The phase offset \(\phi\) divided by the oscillation frequency (\(2\omega\)) can be written as the sum of two delays, \(\tau_{XUV} + \tau_{A}\). The first one is the group delay of the attosecond pulses, while the second, called atomic time delay, arises from the two-photon ionization process. As shown in previous work\(^{33,34}\) and as discussed in more details in the Supplementary Fig. 1, the variation of the atomic time delay \(\tau_{A}\), as a function of XUV photon energy or between two spin-orbit split final states, reflects, to a large extent, one-photon ionization dynamics. To remove the influence of \(\tau_{XUV}\) in our time delay measurements, we alternate experiments in Xe and Ne, and measure the time delay difference. Atomic time delays in Ne 2p can be measured and calculated with good accuracy. They are very small in the energy range considered\(^{33}\), so that the time delay differences between Xe and Ne are, to a very good approximation, absolute time delays in Xe (see Supplementary Fig. 2). The sidbands corresponding to the same photoelectron but different Auger final state are found to oscillate in phase within our error bar, which allows us to average the time delays over the different Auger decay channels.

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![Fig. 1 Excitation scheme. a Schematic illustration of Xe 4d photoionization (violet) and Auger decay processes (green) after absorption of XUV radiation. b Xe energy diagram showing the Xe\(^+\) intermediate and Xe\(^{2+}\) final states involved.](https://example.com/fig1.png)
coincidence with 4
Photoelectrons corresponding to absorption or emission of an additional IR photon (sidebands, labeled as S58 and S60 as an example) appear in the calculated branching ratio of the 4p→4p transitions. The standard error of the weighted mean, detailed in Methods section. In the estimation of the error bars is the sum of the signal with slow electron energy from 10 eV to 10.4 eV, i.e., the photoelectrons in coincidence with 4d→5s→2(3S0) Auger electrons. The projection on the slow electron energy axis c and d is the sum of the signal with slow electron energy from 10 eV to 10.4 eV, i.e., the photoelectrons in coincidence with 4d→5s→2(3S0) Auger electrons. The projection on the slow electron energy axis e shows the sum of the signal for the different Auger processes indicated on the right, with (red) and without (blue) IR field. A RABBIT energy scheme is indicated at the top of d.

Discussion
To understand the underlying physics behind the variation of the time delays, we examine the behavior of the RRPA transition matrix elements involved in Xe 4d single photon ionization. In the energy range considered in this work, photoionization is dominated by the transitions from the 4d shell to continuum f states, which we denote 4d→3f in the following. The contribution from 4d→3f transitions is one order of magnitude smaller in this energy region, as shown in the Supplementary Material. The asymptotic phase for a given channel is the sum of the Coulomb phase and a phase due to the short-range potential. The Coulomb phase is removed in the phases displayed in Fig. 3a, as well as in the calculation of the time delays, in order to focus on the short range effects (see Supplementary Fig. 1). Figure 4a shows that photoionization is dominated by 4dσ2→3fσ2 and 4dσ2→3fπ2, especially at high photon energy, in the region of the giant dipole resonance. In the threshold region, the 4dσ2→3fπ2 channel contributes significantly. This transition is accompanied by a spin flip, which points out the role of the spin-orbit interaction. The phases and time delays for the three channels (Fig. 4b,c) coincide above 80 eV photon energy, showing the first half of a π phase variation across the giant dipole resonance with a time delay of ~40 as. Below 80 eV, the three quantities plotted in Fig. 4a–c show a strong, oscillating, channel dependence, indicating a quantum interference phenomenon.

The dynamics behind this effect can be unraveled by calculating the Wigner representation of the auto-correlation function of the transition matrix elements, D(E), i denoting the channel, and E the electron
While the broad feature, maximum at 100 eV, obviously represents the giant dipole resonance (\(1^3S \rightarrow 1^1P\)), extracted from ref. 12. We exclude the Coulomb phase-shift in \(\delta_{ab}\), as the giant dipole resonance; (ii) A sharp resonance at low energy, around 75 eV, with a long decay of a few hundreds of attoseconds; (iii) Interferences between these resonances, leading to rapid oscillations of the Wigner distribution.

In conclusion, we have measured photoionization time delays in Xe using attosecond interferometry, giving us high temporal resolution, and coincidence spectroscopy, which allows us to avoid spectral congestion and to obtain a high spectral resolution. These time delays are positive and similar for the two spin-orbit split \(^{13}S \rightarrow 3^P\) states over a large energy range (up to 100 eV photon energy), except at threshold (75 eV) where they differ by 100 as. With the help of RRPA calculations for one- and two-photon ionization, we attribute this difference to the interference of several channels coupled by the spin-orbit interaction. A time-frequency analysis of the dominant transition matrix elements, allows us to unravel two main ionization processes, with very different time and energy scales: the broad giant dipole resonance, dominated by the \(1^3S \rightarrow 1^1P\) transition and the narrow resonances due to the \(1^3S \rightarrow 3^P\) and \(1^3S \rightarrow 3^D\) transitions, which have similar amplitudes in this region. This interference explains the difference in time delays for \(4d_{5/2} \rightarrow 4f_{5/2}\), as well as the anomalous branching ratio (Fig. 3c, d).

In Fig. 4d, we show the modulus, phase and time delay of the transition matrix element \(D_i(E)\) as a function of photon energy for the coupled channels (a–c) \(4d_{5/2} \rightarrow 4f_{5/2}\) (blue), \(4d_{5/2} \rightarrow 4f_{7/2}\) (red), \(4d_{5/2} \rightarrow 4f_{1/2}\) (brown) and eigenchannels (d–f) \(1^3P\) (black), \(3^P\) (magenta), \(3^D\) (orange), extracted from ref. 12. We exclude the Coulomb phase-shift in \(\delta_{ab}\) and \(\delta_{bc}\). A zero phase shift has been added to the phase of \(4d_{5/2} \rightarrow 4f_{5/2}\) for better comparison. The time delay is the energy derivative of the phase.

\[
W(E, t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} dE' \frac{(E + \frac{\epsilon}{2})^2}{(E - \frac{\epsilon}{2})^2} e^{-i W(E, t)},
\]

where \(\hbar\) is the reduced Planck constant. The results are shown in Fig. 5a for the \(4d_{5/2} \rightarrow 4f_{5/2}\) channel (see Supplementary Fig. 4a, for the other two channels). All three channels show similar features. (i) A broad resonance with a maximum around 100 eV and a short decay of a few tens of attoseconds, which are enabled by the spin-orbit interaction. A time-frequency analysis of the Wigner distribution.

To interpret the sharp spectral feature at 75 eV, we utilize the theoretical analysis performed in the seminal work of Cheng and Johnson12. Using a multichannel quantum defect theory approach40, results obtained within RRPA, similar to those of the present work, were analyzed and eigenchannel solutions were extracted. These eigenchannel solutions are completely decoupled from each other and can be used as a basis to describe coupled-channel transitions. They are labeled using the closest corresponding LS-coupled channel \((|dP^f_i\rangle + |dP^f_i\rangle)^{1^3P, 3^P, 3^D}\), where in all these cases it is only the \(j = \frac{3}{2}\) states that can be populated from the xenon ground state by one-photon absorption. Neglecting the weak contribution of the \(4d \rightarrow 4f\) transitions, the \(4d \rightarrow ef\) transitions are superpositions of \(dP^f_i\) \(1^3P, 3^P, 3^D\) eigenchannels (In the following, we drop the \(dP^f_i\) label).

Figure 4d–f present the modulus, phase and time delay of these three eigenchannels. The behavior of the three curves is much simpler than those in Fig. 4a–c. For each eigenchannel, a single resonance feature can be identified, with a peak for the modulus and time delay and a \(\pi\) phase variation across the resonance. While the broad feature, maximum at 100 eV, obviously represents the giant dipole resonance \((1^3S \rightarrow 1^1P)\), the narrow peaks at 75 and 76 eV exist because of the spin-orbit interaction that enables singlet to triplet mixing. The maximum of the time delay varies from a few tens \((1^3P)\) to a few hundreds \((3^D, 3^P)\) of attoseconds, in agreement with the results in Fig. 5a.

The difference in time delays can be further interpreted by examining the effective potential experienced by the escaping \(f\) photoelectron. We represent in Fig. 5b a mean-field average potential (red), as well as the potential modified by \(1^3S \rightarrow 1^P\) dipole polarization (screening) effects (black), which are included in the random phase approximation with exchange (RPAE) approach. These effects lead to an effective high and narrow potential barrier and therefore to a broad resonance, with a maximum at high energy, and a short decay time (see black dashed line). In contrast, an electron emitted in the triplet channels does not feel these dipole polarization effects and sees essentially the potential indicated in red, with a relatively low barrier only due to angular momentum and a long decay time (red dashed line). The time delay is directly related to the resonance lifetime, being equal to it at the maximum of the resonance44. Figure 5b even suggests that the increase of the temporal width of the broad resonance in Fig. 5a towards low energy might be due to the influence of the long tail of the screened potential (black).

The rapid variation of the amplitude, phase and delays of the three \(4d \rightarrow ef\) channels (Fig. 4a–c) at threshold can therefore be interpreted as a quantum interference effect between the “direct” dipole-allowed \(1^3S \rightarrow 1^1P\) transition and the spin-orbit-induced \(1^3S \rightarrow 3^P, 3^D\) transitions, which have similar amplitudes in this region. This interference explains the difference in time delays for \(4d_{3/2} \rightarrow 4f_{3/2}\) and \(4d_{5/2}\), as well as the anomalous branching ratio (Fig. 3c, d).

**Figure 4** Transition matrix elements. a–d Modulus, b–e phase and c, f time delay of the transition matrix element \(D(E)\) as a function of photon energy for the coupled channels (a–c) \(4d_{5/2} \rightarrow 4f_{5/2}\) (blue), \(4d_{5/2} \rightarrow 4f_{7/2}\) (red), \(4d_{5/2} \rightarrow 4f_{1/2}\) (brown) and eigenchannels (d–f) \(1^3P\) (black), \(3^P\) (magenta), \(3^D\) (orange), extracted from ref. 12. *We exclude the Coulomb phase-shift in \(\delta_{ab}\) and \(\delta_{bc}\). A zero phase shift has been added to the phase of \(4d_{5/2} \rightarrow 4f_{5/2}\) for better comparison. The time delay is the energy derivative of the phase.*

**Figure 5** Wigner representation and effective potentials. a Wigner representation \(W(E, t)\) for the \(4d_{5/2} \rightarrow 4f_{5/2}\) channel. The amplitude is indicated by the color code on the right. b Illustration of one-electron potentials for the \(1^3S \rightarrow 3^P, 3^D\) transitions (red) and \(1^3S \rightarrow 1^P\) (black). Dashed lines suggest possible electron trajectories in the two cases.
traditional spectral studies, provides increased understanding of the complex electron dynamics taking place in Xe 4d photoionization. Finally, shape resonances are ubiquitous in nature and the methods developed in this work should be useful to investigate the electronic properties of a variety of molecular systems (see recent work in N$_2$ [42] and CH$_3$I[43]).

**Methods**

**Experimental method.** The experiments were performed with 40-fs long pulses, centered at 800 nm with 1-kHz repetition rate from a Ti:sapphire femtosecond laser system. The laser was focused into a 6-mm long gas cell filled with Ne to generate high-order harmonics. A 200-nm-thick Zn filter was used to filter out the infrared (IR) and most of the harmonics below the Xe 4d threshold (67.5 eV for 4d$_x^2$). The filtered harmonics thus span the 4d ionization region from the threshold to the maximum of the giant dipole resonance (100 eV). A small fraction (30%) of the IR beam, split-off before generation, is used as a probe with a variable time delay. The XUV and IR pulses were focused in an effusive Xe gas jet. The electrons were detected by a magnetic bottle electron spectrometer, which combines high collection efficiency and high spectral resolution up to E/E$\Delta$F – 80.

**Data analysis.** Each data point in Fig. 3 is the arithmetic mean weighted with the uncertainty estimated from the cosine fitting to Eq. (1). In each measurement, we average the time delays of electrons pairs corresponding to the same photoelectron but different Auger decay. For N measurements yielding N data points: $\tau_1, \tau_2, \ldots, \tau_N$ with corresponding uncertainties: $\sigma_1, \sigma_2, \ldots, \sigma_N$, the weighted average can be calculated as:

$$\bar{\tau} = \sum_{i=1}^{N} \frac{w_i \tau_i}{\sum_{i=1}^{N} w_i}, \quad \text{where} \quad w_i = \frac{1}{\sigma_i^2}$$

(3)

where $w_i$ is the weight. The uncertainty for each measurement is estimated from the fit of the RABBIT oscillation to a cosine function. The uncertainty on the time delay difference, $\tau_A - \tau_B$, can be expressed as:

$$\sigma = \sqrt{\sigma_A^2 + \sigma_B^2}$$

(4)

The error bars of the experimental results indicate the standard error of the weighted mean and can be calculated as:

$$\sigma_{\bar{\tau}} = \frac{1}{\sqrt{N(N-1)}} \sum_{i=1}^{N} \omega_i (\tau_i - \bar{\tau})^2$$

(5)

where $\omega_i = \sum w_i$.

**Theoretical method.** Theoretical calculations consisted in calculating one-photon and two-photon matrix elements within lowest-order perturbation theory for the radiation fields, using wavefunctions obtained by solving the Dirac equation, and including electron correlation effects within the RPAE for one-photon XUV photoionization. The complex-valued two-photon matrix elements$^{44,45}$ are calculated following the procedure described in$^{46,47}$ for the non-relativistic case. Briefly, the absorption of one ionizing photon is treated within the RPAE approximation and a perturbed wave function is calculated. Exterior complex scaling is used in order to be able to finite numerical box. The two-photon matrix element is dominated by a one-photon field of light.

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Author contributions

S.Z., D.B., R.J.S., M.I., L.N., H.L., R.W. and C.L.A. performed the experiment. R.J.S. and R.F. provided part of the experimental setup. J.V., J.M.D. and E.L. performed RRPA calculations. S.Z., D.B., J.M.D., G.W., M.G., E.L. and A.L. worked on the analysis and the theoretical interpretation. S.Z. and A.L. wrote the main part of the manuscript. All authors gave feedback on the manuscript.

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Competing Interests

The authors declare no competing interests.

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