Electron spin polarization in strong-field ionization of xenon atoms

Alexander Hartung1*, Felipe Morales2, Maksim Kunitskii3, Kevin Henrichs1, Alina Laucke1, Martin Richter1, Till Jahnke1, Anton Kalinin1, Markus Schöffler1, Lothar Ph. H. Schmidt1, Misha Ivanov2, Olga Smirnova2 and Reinhard Dörner1

As a fundamental property of the electron, the spin plays a decisive role in the electronic structure of matter, from solids to molecules and atoms, for example, by causing magnetism. Yet, despite its importance, the spin dynamics of the electrons released during the interaction of atoms with strong ultrashort laser pulses has remained experimentally unexplored1,2. Here, we report the experimental detection of electron spin polarization by the strong-field ionization of xenon atoms and support our results with theoretical analysis. We found up to 30% spin polarization changing its sign with electron energy. This work opens the new dimension of spin to strong-field physics. It paves the way to the production of sub-femtosecond spin-polarized electron pulses with applications ranging from probing the magnetic properties of matter at ultrafast timescales3 to testing chiral molecular systems with sub-femtosecond temporal and sub-Ångström spatial resolutions.

Short laser pulses provide an electric field that can be strong enough to suppress the binding potential of an atom or molecule and lead to field ionization. Electrons that pass over the barrier, or tunnel just under it, and emerge from the atom are subsequently driven by the laser field. So far, nearly all of the works exploring electronic behaviour after ionization have solely used the binding energy of the electron and the shape of the barrier as the defining properties, omitting the other fundamental property of the electron—its spin. This is even more surprising as a few pioneering theoretical works have indicated the importance of the spin of the outgoing electron in strong-field ionization2-5. From a fundamental quantum standpoint, the spin of the liberated electron should not be ignored as there is correlation between this electron and the ion left behind and ionization is known to trigger spin–orbit dynamics in the ion4. Noble gas atoms, with their closed shells and overall vanishing spin, provide an ideal starting point for the study of such strong-field spin effects.

For single-photon ionization, the spin polarization of the photoelectrons that are ejected from the outermost orbital has been thoroughly studied both experimentally5 and theoretically6-9. However, the physics behind spin polarization in this case is completely different from the strong-field regime discussed here. In the single-photon case, photoelectrons of the same energy populate a small set of continuum angular momentum states, as dictated by the dipole selection rules. Different phase shifts for each such set of continuum states then lead to spin polarization of the energetically degenerate electrons8. The single-photon case was generalized to the weak-field multiphoton regime2-5, uncovering the importance of intermediate resonances. In contrast, in the case of over-barrier strong-field ionization discussed here, electrons of different spin (but same binding energies) are substantially shifted in their kinetic energy in the continuum. The shift is due to the different direction of the bound state momentum relative to the sense of rotation of the laser field and the correlation between the bound state momentum and the spin of the released electron, see Fig. 2 and the discussion below.

For the present study we exposed xenon atoms to circularly polarized 780 nm, 40 fs laser pulses. The peak intensity was estimated to be $I_0 \sim 3.3 \times 10^{14}$ W cm$^{-2}$ at the centre of the focal spot. The intensity is well above the saturation intensity of Xe (refs 10, 11), leading to ionization on the rising edge of the pulse, as expected in the saturation regime. The effective intensity $I_{\text{eff}}$ at which most of the electrons were released can be extracted from the peak position of our experimental electron energy distribution, which for circularly polarized pulses is around the electron ponderomotive energy $U_e = e^2 F^2/2m_\text{e} \omega^2$ where $F$ is the electric field amplitude, $\omega$ is the laser frequency and $e$ and $m_\text{e}$ are the electron charge and mass (see ref. 12). This estimate yields $I_{\text{eff}} \sim 1.4 \times 10^{14}$ W cm$^{-2}$, consistent with field ionization with little or no tunnelling. We measured the kinetic energy and spin polarization of the electrons with a time-of-flight spectrometer equipped with a commercial Mott polarimeter10. Figure 1 shows the measured spin polarization as a function of the electron kinetic energy (see the Supplementary Information). Positive values correspond to a surplus of electrons with spins parallel to the propagation axis of the laser. The error bars show statistical errors only. The blue curve was generated by solving the time-dependent Schrödinger equation (TDSE).

---

1Institut für Kernphysik, Goethe Universität Frankfurt, Max-von-Laue-Straße 1, D-60438 Frankfurt am Main, Germany. 2Max-Born-Institut, Max-Born-Straße 2A, D-12489 Berlin, Germany. *e-mail: hartung@atom.uni-frankfurt.de

---

**Figure 1** | Spin polarization of the electrons ejected by strong-field ionization of Xe parallel to the direction of light propagation by circularly polarized laser pulses. The spin polarization is defined as the weighted difference between spin-up and spin-down electrons (see the Supplementary Information). Positive values correspond to a surplus of electrons with spins parallel to the propagation axis of the laser. The error bars show statistical errors only. The blue curve was generated by solving the time-dependent Schrödinger equation (TDSE).
function of the electron kinetic energy. The experimental results are in good agreement with our numerical simulations.

The basic physics behind our observation is explained in Fig. 2a, which shows a schematic of the ionization process from the highest 5p \( j = 3/2 \) orbital of Xe. We chose the quantization axis to be the light propagation direction (orange), with the positive projection of the total angular momentum \( m_\ell \) being in the light direction. The two 5p \( j = 3/2 \) orbitals with \( m_l = 3/2 \) (red) and \( m_l = -3/2 \) (blue) are degenerate in energy. For \( m_l = 3/2 \) the electron spin is oriented upwards (downwards) and the electrons with the initial tangential to the direction of the laser field. Different spin states (red, spin-up electrons; blue, spin-down electrons) therefore correlate with the different shifted energy distributions, leading to energy-dependent spin polarization. b, Theoretical energy distribution of \( s \) and \( p \) states. The intuitive picture in a is confirmed by results obtained from the numerical solution of the time-dependent Schrödinger equation, for a single active electron in the initial \( p \) state. The effective potential for the electron motion is chosen to fit the 5p \( j = 3/2 \) ionization potential of Xe. For the initial \( s \) state, this potential was modified to maintain the same binding energy as for the \( p \) state. The heights of the three distributions were normalized to unity.

Figure 2 | Ionization process and theoretical energy distribution of \( s \) and \( p \) states. a, Schematic of the ionization process of the Xe 5p state. The 5p \( j = 3/2 \) states of Xe are predominantly ionized. In \( j = 3/2 \) \( |m_j = 3/2 \) states the angular momentum and spin of the electron are parallel. The initial rotational state of the electron results in an offset momentum in the direction of the streaking momentum imparted on the electron by the laser field. Different spin states (red, spin-up electrons; blue, spin-down electrons) therefore correlate with the different shifted energy distributions, leading to energy-dependent spin polarization. b, Theoretical energy distribution of \( s \) and \( p \) states. The intuitive picture in a is confirmed by results obtained from the numerical solution of the three-dimensional time-dependent Schrödinger equation, for a single active electron in the initial \( p \) state. The effective potential for the electron motion is chosen to fit the 5p \( j = 3/2 \) ionization potential of Xe. For the initial \( s \) state, this potential was modified to maintain the same binding energy as for the \( p \) state. The heights of the three distributions were normalized to unity.

The basic physics behind our observation is explained in Fig. 2a, which shows a schematic of the ionization process from the highest 5p \( j = 3/2 \) orbital of Xe. We chose the quantization axis to be the light propagation direction (orange), with the positive projection of the total angular momentum \( m_\ell \) being in the light direction. The two 5p \( j = 3/2 \) orbitals with \( m_l = 3/2 \) (red) and \( m_l = -3/2 \) (blue) are degenerate in energy. For \( m_l = 3/2 \) the electron spin is oriented upwards (downwards) and the electrons with the initial tangential to the direction of the laser field. Different spin states (red, spin-up electrons; blue, spin-down electrons) therefore correlate with the different shifted energy distributions, leading to energy-dependent spin polarization. b, Theoretical energy distribution of \( s \) and \( p \) states. The intuitive picture in a is confirmed by results obtained from the numerical solution of the three-dimensional time-dependent Schrödinger equation, for a single active electron in the initial \( p \) state. The effective potential for the electron motion is chosen to fit the 5p \( j = 3/2 \) ionization potential of Xe. For the initial \( s \) state, this potential was modified to maintain the same binding energy as for the \( p \) state. The heights of the three distributions were normalized to unity.

The basic physics behind our observation is explained in Fig. 2a, which shows a schematic of the ionization process from the highest 5p \( j = 3/2 \) orbital of Xe. We chose the quantization axis to be the light propagation direction (orange), with the positive projection of the total angular momentum \( m_\ell \) being in the light direction. The two 5p \( j = 3/2 \) orbitals with \( m_l = 3/2 \) (red) and \( m_l = -3/2 \) (blue) are degenerate in energy. For \( m_l = 3/2 \) the electron spin is oriented upwards (downwards) and the electrons with the initial tangential to the direction of the laser field. Different spin states (red, spin-up electrons; blue, spin-down electrons) therefore correlate with the different shifted energy distributions, leading to energy-dependent spin polarization. b, Theoretical energy distribution of \( s \) and \( p \) states. The intuitive picture in a is confirmed by results obtained from the numerical solution of the three-dimensional time-dependent Schrödinger equation, for a single active electron in the initial \( p \) state. The effective potential for the electron motion is chosen to fit the 5p \( j = 3/2 \) ionization potential of Xe. For the initial \( s \) state, this potential was modified to maintain the same binding energy as for the \( p \) state. The heights of the three distributions were normalized to unity.

The basic physics behind our observation is explained in Fig. 2a, which shows a schematic of the ionization process from the highest 5p \( j = 3/2 \) orbital of Xe. We chose the quantization axis to be the light propagation direction (orange), with the positive projection of the total angular momentum \( m_\ell \) being in the light direction. The two 5p \( j = 3/2 \) orbitals with \( m_l = 3/2 \) (red) and \( m_l = -3/2 \) (blue) are degenerate in energy. For \( m_l = 3/2 \) the electron spin is oriented upwards (downwards) and the electrons with the initial tangential to the direction of the laser field. Different spin states (red, spin-up electrons; blue, spin-down electrons) therefore correlate with the different shifted energy distributions, leading to energy-dependent spin polarization. b, Theoretical energy distribution of \( s \) and \( p \) states. The intuitive picture in a is confirmed by results obtained from the numerical solution of the three-dimensional time-dependent Schrödinger equation, for a single active electron in the initial \( p \) state. The effective potential for the electron motion is chosen to fit the 5p \( j = 3/2 \) ionization potential of Xe. For the initial \( s \) state, this potential was modified to maintain the same binding energy as for the \( p \) state. The heights of the three distributions were normalized to unity.
this Letter is different from predictions made in ref. 1 that are based on a spin polarization mechanism that operates purely in the tunneling regime and illustrated for a short range of potentials where the barrier cannot be suppressed. Tunnelling through the barrier in a circularly polarized field will lead to an initial momentum distribution after tunnelling shifted away from zero, even for an s state. As a result, all of the energy distributions and the zero crossing of the spin polarization are shifted to higher energies ($U_0 + U_1$ in this regime). Thus, the zero crossing of the spin polarization is a sensitive measure of the kinematics of the ionization process.

The significant degree of spin polarization in electrons ejected from Xe atoms by a strong ultrashort laser pulse opens new directions for strong-field physics. As shown in the Supplementary Information, spin polarization is also important during laser-driven electron recollision with the parent ion in elliptically polarized laser fields. As the recollision signal decreases with ellipticity $\epsilon$ (whereas spin polarization increases with $\epsilon$), there is a trade-off between the two. Substantial spin polarization can be achieved while keeping the strength of the recollision signal well within experimental capabilities, see Supplementary Figs 1 and 2.

The coherence of the recollision process dictates that the degree of spin polarization should be considered in correlation with the state of the parent cation. Supplementary Figs 1 and 2 show that spin polarization can exceed 50% for recollision with the Xe$^+$ (P$_{1/2}$) core state at ellipticities where the recollision signal is still adequate for experiment. Thus, the spin and angular momentum polarization will be important in recollision-driven processes such as laser-induced electron diffraction and holography, correlated multiple ionization and high-harmonic generation using single- and especially multicolour chiral laser fields, which can ensure a strong recollision signal while still maintaining sensitivity to the orientation of the electron’s initial angular momentum. This would allow chiral molecules to be probed with sub-femtosecond temporal resolution and sub-Ångström spatial resolution. Our results show that orbital imaging can be extended to probe stationary and dynamical currents, in molecular orbitals for example. The application of modern few-cycle circularly polarized pulses would allow the production of sub-femtosecond spin-polarized electron pulses, which can then be used to probe the magnetic properties of matter on ultrafast timescales. Finally, spin polarization of the ejected electron is firmly linked to the creation of the parent ion in an initially spin-polarized state. Spin–orbit coupling then leads to internal circular electron and spin currents, as has been predicted.

Methods
Methods and any associated references are available in the online version of the paper.

Received 16 October 2015; accepted 2 May 2016; published online 13 June 2016

References
1. Barth, I. & Smirnova, O. Spin-polarized electrons produced by strong-field ionization. Phys. Rev. A 88, 013401 (2013).
2. Barth, I. & Smirnova, O. Hole dynamics and spin currents after ionization in strong circularly polarized laser fields. J. Phys. B 47, 204020 (2014).
3. Krieger, K., Dewhurst, J. K., Elliott, P., Sharma, S. & Gross, E. K. U. Laser induced ultrafast demagnetization: an ab initio perspective. Preprint at http://arxiv.org/abs/1406.6607 (2014).
4. Goulielmakis, E. et al. Real-time observation of valence electron motion. Nature 466, 739–743 (2010).
5. Heckenkamp, C., Schäfers, F., Schönhense, G. & Heinze, M. Experimental characterization of the Xe 5p photoionization by angle- and spin-resolved photoelectron spectroscopy. Z. Phys. D 2, 257–274 (1986).
6. Fano, U. Spin orientation of photoelectrons ejected by circularly polarized light. Phys. Rev. 178, 131 (1969).
7. Lambropoulos, P. Spin-orbit coupling and photoelectron polarization in multiphoton ionization of atoms. Phys. Rev. Lett. 30, 413 (1973).
8. Lambropoulos, P. On producing totally polarized electrons through multiphoton ionization. J. Phys. B 7, L33 (1974).
9. Dixit, S. N., Lambropoulos, P. & Zoller, P. Spin polarization of electrons in two-photon resonant three-photon ionization. Phys. Rev. A 24, 318 (1981).
10. Larocheille, S., Talebpour, A. & Chin, S. L. Non-sequential multiple ionization of rare gas atoms in a Ti:Sapphire laser field. J. Phys. B 31, 1201–1214 (1998).
11. Hankin, S. M., Villeneuve, D. M., Corkum, P. B. & Rayner, D. M. Intense-field laser ionization rates in atoms and molecules. Phys. Rev. A 64, 013403 (2001).
12. Corkum, P. B., Burnett, N. H. & Brunel, F. Above-threshold ionization in the long-wavelength limit. Phys. Rev. Lett. 62, 1239 (1989).
13. Burnett, G. C., Monroe, T. J. & Dunning, F. R. High-efficiency retarding-potential Mott polarization analyzer. Rev. Sci. Instrum. 65, 1893–1896 (1994).
14. Salieres, P. et al. Feynman’s path-integral approach for intense-laser-atom interactions. Science 292, 902–905 (2001).
15. Meckel, M. et al. Laser-induced electron tunneling and diffraction. Science 320, 1478–1482 (2008).
16. Spanner, M., Smirnova, O. Corkum, B. P. & Ivanov, M. Y. Reading diffraction images in strong field ionization of diatomic molecules. J. Phys. B 37, L243 (2004).
17. Husmanns, Y. et al. Time-resolved holography with photoelectrons. Science 331, 61–64 (2011).
18. Blaga, C. et al. Imaging ultrafast molecular dynamics with laser-induced electron diffraction. Nature 483, 194–197 (2012).
19. Pullen, M. G. et al. Imaging an aligned polyatomic molecule with laser-induced electron diffraction. Nature Commun. 6, 7262 (2015).
20. Milosevic, D. B., Becker, W. & Koppold, R. Generation of circularly polarized high-order harmonics by two-color coplanar field mixing. Phys. Rev. A 61, 063403 (2000).
21. Fleischer, A., Økr, O., Diskin, T., Sidorenko, P. & Cohen, O. Spin angular momentum and tunable polarization in high-harmonic generation. Nature Photon. 8, 543–549 (2014).
22. Kfir, O. et al. Generation of bright phase-matched circularly-polarized extreme ultraviolet high harmonics. Nature Photon. 9, 99–105 (2015).
23. Medissaouka, L. et al. Generating isolated elliptically polarized attosecond pulses using bichromatic counterrotating circularly polarized laser fields. Phys. Rev. Lett. 115, 153001 (2015).
24. Mancuso, C. A. et al. Strong-field ionization with two-color circularly polarized laser fields. Phys. Rev. A 91, 031402 (R), (2015).
25. Baltuska, A., Paulus, G. G., Lindner, F., Kienberger, R. & Krausz, F. in Femtosecond Optical Frequency Comb: Principle, Operation, and Applications (eds Ye, J. & Cundiff, S.) 263–313 (Springer, 2005).

Acknowledgements
The experimental work was supported by the Deutsche Forschungsgemeinschaft. A.H. and K.H. acknowledge support from the German National Merit Foundation. M.I. acknowledges support from the EPSRC Programme Grant EP/I032517/1 and the United States Air Force Office of Scientific Research program no. FA9550-12-1-0482. F.M. and O.S. acknowledge support from the DFG grant SM 292/3.

Author contributions
All authors edited and commented on the manuscript. A.H. and M.K. carried out the measurements. A.H., M.K., K.H., M.R., T.J., A.K., M.S. and L.S. built up the experimental set-up. A.H. and A.L. analysed the experimental data. F.M. and O.S. carried out the numerical simulations. R.D. supervised all work.

Additional information
Supplementary information is available in the online version of the paper. Reprints and permissions information is available online at www.nature.com/reprints. Correspondence and requests for materials should be addressed to A.H.

Competing financial interests
The authors declare no competing financial interests.
Experimental measurements. A commercial KMLabs Dragon Ti:Sa laser system (40 fs, 780 nm, 0.5 mJ per pulse) was employed for the measurements. We used a quarter-wave plate to produce circular polarization from the initially linearly polarized light. The ellipticity of the electric field for left circular polarization $e = 0.96$ (for right circularly polarized light $e = 0.93$) was measured with a Glan polarizer and a rotational stage. The laser pulses were focused into the Xe gas target by a lens of $f = 10$ cm. The emitted electrons travelled through a 50 cm field-free drift-tube to a commercially available Mott detector\(^1\), which is capable of measuring the spin polarization of an electron beam. Owing to spin–orbit interactions the differential cross-section of the electrons scattered at high-Z atoms (in our detector thorium target is used) is spin-dependent. A Mott detector utilizes this effect and measures the scattering asymmetry $A$ of electrons. $A$ is given by $A = \frac{N_{1} - N_{2}}{N_{1} + N_{2}}$, where $N_{1,2}$ are the number of electrons scattered upwards and downwards, respectively. $A$ is related to the spin polarization $P$ by $A = S_{\text{eff}}P$, where $S_{\text{eff}}$ is a constant of proportionality defined by the detector geometry. For the instrumental scaling factor $S_{\text{eff}}$ of our Mott polarimeter we used $S_{\text{eff}} = -0.15$.

The literature value is between $-0.15$ and $-0.25$ for an acceleration voltage of 25 kV. In the measurements 18 kV was used and we therefore reduced $S_{\text{eff}}$ accordingly\(^2\).

In addition to statistically measuring the polarization we gained information on the kinetic energy of each electron by recording its time of flight. Instrumental imperfections in our set-up (for example different detection efficiencies) would lead to asymmetry, which is indistinguishable from the measured asymmetry caused by polarization. To cancel those, we made two measurements with left and right circularly polarized light. Between those measurements the polarization effect should just switch sign, whereas the instrumental asymmetry stays the same. To compare the two measurements in analysis a third microchannel plate detector was employed, lying in the plane of laser propagation and hence unaffected by a possible spin polarization in that axis. Small intensity differences between the measurements with left and right circularly polarized light were levelled out by comparing the energy distributions of this third detector. The distributions were superimposed by stretching one of them and assigning this stretch factor to the other two spin-measuring detectors.

Numerical simulations. For the theoretical calculations in Fig. 2, we have numerically solved the time-dependent Schrödinger equation using the single active electron approximation, with the electron in the initial $p^+$ or $p^-$ state (that is, co-rotating or counter-rotating with respect to the laser field). We used the effective model potential\(^3\) $V(r) = -(1/r) - ((Z - 1)\epsilon^* / r)$ with $r$ being the distance between electron and nucleus, $Z = 54$ and the parameter $\kappa = 1.2285$ adjusted to fit the lowest ionization potential of the Xe $5s$ shell, $I_p = 12.13$ eV. The electron spectrum was obtained by propagating the wave function for a further two cycles after the end of the laser pulse, during which electrons with 9 eV energy (as an example) move by an extra 170 a.u. away from the origin. Next, we extracted the continuum part of the wave function using a spatial square mask with a radius of 100 a.u. that eliminates the central part of the wave function near the core. This masking procedure is adequate, because for an intense circularly polarized pulse the electronic wave function is well-separated into the bound part near the core and the continuum part far away from the core. The remaining continuum part was then projected on the plane wave basis. The accuracy of this procedure has been monitored by varying the extra propagation time up to five cycles, by varying the radius and the width of the spatial mask and by comparing the spectra obtained with the same laser conditions and using the same procedure but for a hydrogen atom against the exact spectra obtained by projecting on the well-known exact continuum eigenstates of hydrogen. For the initial $s$ state (in Fig. 2), the effective potential was modified to maintain the same ionization potential binding energy as for the two $p$ states. We have used different pulse shapes to test the validity of the numerical analysis: two cycles sin$^2$ ramp up and two cycles sin$^2$ ramp down, four cycles sin$^2$ ramp up and four cycles sin$^2$ ramp down and a ‘long pulse’ with 2 cycles sin$^2$ ramp up and down and four cycles flat top. Aside from the angular asymmetry introduced by the two shorter pulses, the angle-integrated electron energy distributions and spin polarization remained essentially the same. All of the numerical data shown in the figures are for the long pulse. For the calculations in Fig. 2 the amplitude of the circularly polarized electric field was set to $F = 0.05$ a.u.

For the spin–orbit channel-resolved calculations in Fig. 3 we modified the effective potential to fit the ionization potential of the $5p^+$ state ($I_p = 13.44$ eV) following the recipe described in ref. 27. Specifically, an extra short-range potential step was added to the effective potential. The step is non-zero only at the first grid point, fixed at 0.5 a.u. The effective field intensity of $1.4 \times 10^{14}$ W cm$^{-2}$ was chosen such that the spin-integrated electron energy spectra for experiment and theory overlap each other. We then followed the prescription in ref. 1 to compute the spin polarization depicted in Fig. 1, performing further averaging over intensities $I = 1.1–1.4 \times 10^{14}$ W cm$^{-2}$.

References
26. Popruzhenko, S. V., Paulus, G. G. & Bauer, D. Coulomb-corrected quantum trajectories in strong-field ionization. Phys. Rev. A 77, 053409 (2008).
27. Müller, H. G. Tunneling excitation to resonant states in helium as main source of superponderomotive photoelectrons in the tunneling regime. Phys. Rev. Lett. 83, 3158–3161 (1999).