

timescale. As explained in the Methods section, this is again consistent with momentum conservation. Supplementary Fig. 5 shows that, if we dress the harmonics with a Gaussian infrared beam, the oscillations wash out as expected. The RABBIT trace also demonstrates the spectral phase-locking of the high harmonic orders, thus confirming the attosecond ‘light spring’ structure of the XUV bursts generated with a helically phased infrared beam.

## Discussion

The two sets of measurements reported above, that is, the spatial intensity profile of the harmonics and their GD finally provides us with all data needed to reconstruct the full spatiotemporal shape of the emitted attosecond electron beam. We hereafter focus in the case of photoelectrons with linear momentum along the propagation axis, which directly reproduce the spring structure of the XUV beam. As lately predicted<sup>11,12</sup>, owing to the low GD over our spectral range, we get two intertwined spring-like structures (Fig. 3c). The helical shape is a direct consequence of the ionizing XUV light structure, while the presence of two spirals is reminiscent of the generation of odd harmonics only. A cut in the spatial domain displays a double lobe structure, while the temporal profile shows a train of attosecond pulses lasting  $\sim 200$  as (Fig. 3d). An interesting characteristic of this structure is that it leads to an identical attosecond pulse train when observed at different azimuthal positions, only delayed by  $7.4 \text{ as} \cdot \text{degree}^{-1}$  (half a period of the infrared driving laser (1,330 as) in 180 degrees). Turning back to light springs, as shown in ref. 11, the corkscrew structure still holds in the case of single attosecond pulses. We believe that this property makes such pulses powerful tools for transient absorption spectroscopy experiments, which require tunable pump–probe delays on the attosecond timescale. Here one may angularly map the attosecond time delay in a single shot, getting rid of any stability requirement. The dynamical range for the delay scan is here 1.33 fs, that is, the infrared laser half optical cycle, which could easily be increased up to several femtoseconds by increasing the driving field wavelength.

In conclusion, we reported on the synthesis and full characterization of attosecond XUV pulses and electron beams, both carrying OAM. The experimental evidence for the transfer of OAM is obtained over an extremely broad spectral range down into the XUV region. This divergence-based analysis method can easily be generalized to arbitrarily large spectral bandwidths. The attosecond structure of the generated XUV pulse train was determined through a photoionization-based cross-correlation technique. First, it confirms the conservation of the standard photoionization selection rules when using helically phased XUV light beams. Second, it opens the route to the manipulation of attosecond electron beams carrying OAM, which will use the large panel of attosecond tools developed in the past 15 years, may it be through high harmonics spectroscopy or XUV–infrared pump–probe experiments. In particular, helical dichroisms were predicted in the XUV spectral range and still remain to be observed. Multicolour HHG using such tailored beams could also provide all-in-one pump–probe schemes, taking advantage of the spatial encoding of the delay in the azimuthal coordinate of these beams. Finally, the accurate characterization of these new light beams will pave the way to the study of the controversial coupling between SAM and OAM in matter during photoionization.

## Methods

**Theoretical details.** In this paragraph we provide details about the simulation of HHG using a driving laser carrying an OAM. For the infrared beam, we accounted for the full experimental set-up sketched in Fig. 2a. To this end, the beam was propagated through the different optical elements, by means of the Huygens–Fresnel integral, up to the gas jet entrance. We consider a spatially and

temporally incident Gaussian beam of 6.25 mm waist and 50 fs duration (full width at half maximum, FWHM). It is propagated through a SPP and focused by a 1 m focal length lens, in the middle of a 500  $\mu\text{m}$  wide (FWHM) Lorentzian argon jet. The wavelength  $\lambda$  is 800 nm and the phase plate here imposes a  $\ell_1 \times 2\pi$  phase shift along a circle centred on the beam axis ( $\ell_1 = 1$  here). The equivalent Gaussian beam waist at focus  $w_0$  is 40  $\mu\text{m}$ , leading to an equivalent Rayleigh range  $z_R$  of 6.5 mm. The maximum gas pressure is 10 mbar and the laser peak intensity at focus is  $1.5 \times 10^{14} \text{ W cm}^{-2}$ . This is the starting point for HHG calculations. The structure of the LG mode, which does not possess the cylindrical symmetry of a Gaussian beam, required performing four-dimensional (three-dimensional (3D) in space + time) simulations. In brief, the coupled propagation equations for the driving laser and harmonic fields are numerically solved on a 3D spatial grid, in the paraxial and slowly varying envelope approximations, using a standard finite-difference method. Calculations are performed in a frame moving at the group velocity of the laser pulse, that is, the speed of light in vacuum here. Both electron and atomic dispersions are taken into account. We first computed the driving field at a given time  $t$  in the pulse envelope and position  $z$  along the propagation axis. This field is then used for calculating the space- and time-dependent ionization yields and dipole strengths. Ionization rates are modelled using Ammosov–Delone–Kraïnov tunneling formula<sup>34</sup>, while dipoles are computed in the strong field approximation, following the model described in ref. 35. Once these two terms are obtained, the equation of propagation for the harmonic field is solved. The calculation is repeated for each  $t$  and  $z$ . The SPP inducing the OAM is modelled by  $\varphi(x, y) = \ell_1 \tan^{-1} \frac{y}{x}$ , where  $x$  and  $y$  are the coordinates in the plane perpendicular to the propagation axis. The results of the calculations are reported in Fig. 1. As expected, HHG occurs along the peak intensity ring of the generating infrared beam. The FWHM of the profile obtained by a cut along a radius of this ring (hereafter called the thickness of the ring) is a fraction of the thickness of the ring of the driving field, a consequence of the high nonlinearity of HHG. As for the spatial phase, a typical example is given in Fig. 1 by the 15<sup>th</sup> harmonic order (H15), showing a spiral running 15 times  $2\pi$  along the ring. This implies that the H15 photons carry 15 times the OAM of the fundamental frequency photons. This behaviour is consistent with the multiplicative law of perturbative nonlinear optics, and was observed here for all computed harmonic orders, from H11 to H33. Finally, to mimic the experiment, we simulate the propagation of the XUV beam towards an observation plane located 80 cm downstream the gas cell. When reaching the detector the harmonic beam still displays a ring shape, suggesting that generation at focus is dominated by the emission of a single LG mode. The ring diameter increases by a factor of  $1.4 \approx \sqrt{2}$  when doubling the helicity of the phase. This may be guessed from analytical considerations presented in Supplementary Note 1, which show, under reasonable hypotheses, that the divergence of harmonics goes like  $\sqrt{|\ell_1|}$ .

**Experimental details.** The experimental set-up is sketched in Fig. 2a. The experiments were performed using the LUCA laser server at CEA Saclay, which delivers 30 mJ, 50 fs, 800 nm pulses at 20 Hz. The laser Transverse Electromagnetic Mode TEM<sub>00</sub> was converted using a 16-level SPP manufactured by SILIOS Technologies. For RABBIT measurements, the laser is split into two uneven parts using a mirror with a 8 mm hole. The main (outer) part of the beam is focused by a 1 m focal length lens (L) into a gas jet provided by a piezoelectric driven valve (Attotech). We could verify that even after reflection off a drilled mirror, the infrared focus kept a donut shape and so did the harmonics, with again a constant divergence. A diaphragm (D) removes the remaining infrared beam, while the harmonics are focused by a 0.5 m focal length toroidal mirror into the sensitive region of a 1 m long magnetic bottle electron spectrometer time of flight (MBES). A SiO<sub>2</sub> plate serves as further attenuation of the infrared beam. An extra plain weak infrared beam (4 mm diameter, energy of 70  $\mu\text{J}$ ) may be superimposed and synchronized with the XUV beam in the MBES with a delay controlled by a piezoelectric transducer. Focus imaging in the sensitive region of the electron spectrometer reveals a thick donut profile for the dressing beam, ensuring a homogeneous dressing of the XUV. For intensity measurements, the drilled mirrors are replaced by plain mirrors and the lens has a 2 m focal length. The harmonics are collected downstream the MBES on a photon spectrometer made of a variable line spacing Hitachi grating (001-0437) and a micro channel plate coupled to a phosphor screen. In order to reveal the 2D spatial profile of the harmonics while spectrally resolving them, the MCP are placed 8 cm before the flat field of the grating. The phosphor screen is imaged with a Basler A102f CCD camera. The observation distance from the source to the MCP is 115 cm. We could generate harmonics in argon gas with  $\ell_1 = 1, 2$  and 3 (Fig. 1) and neon gas with  $\ell_q = 1$  up to H41 (Supplementary Fig. 3). In all cases, we observed a constant ring diameter over the whole XUV spectral range, confirming the general validity of our first measurements in argon. The maximum value of OAM obtained was  $\ell_{19} = 57$  using  $\ell_1 = 3$  in argon.

**RABBIT with a beam carrying an OAM.** The theory of quantum interferometry used to characterize attosecond XUV pulses usually consider flat wavefronts for both the XUV and dressing infrared beam. Here we anticipate the wavefront of the XUV to be tilted. To be more specific, we assume that the spatial phase of the  $q$ -th harmonic within the beam is  $\Phi_q(R, \theta) = \ell_q \theta$ , where the  $(R, \theta)$  polar coordinates refer to the beam axis ( $R = 0$ ). The measured SB intensity is an average of the