

Like massive particles may carry two types of angular momenta, namely spin and orbital angular momenta (SAM and OAM, respectively), the massless photon can be assigned two such characteristics¹. It was recognized earlier that SAM is associated with the circular polarization of light beams, but only 20 years ago was the OAM of light associated with beams with a tilted wavefront². The unique properties of twisted light beams and their large availability in the infrared and visible spectral regions lead to the emergence of countless applications, from quantum information³ to microscopy⁴, nanoparticle manipulation⁵ or fine structuring of materials using pulsed lasers⁶. In the extreme ultraviolet (XUV) spectral range, technological applications using OAM beams were anticipated^{7,8}, building on predictions of specific light matter interactions^{9,10}. However, because of the lack of sources, most of these predictions could not be confronted to the experiment. Here we first demonstrate a route based on high harmonic generation to synthesize and characterize helically shaped XUV trains of attosecond pulses. Using this photon source, we also report the experimental synthesis of electronic vortices with attosecond time structure. These breakthroughs pave the route for the study of a series of fundamental phenomena and the development of new ultrafast diagnosis tools using either photonic or electronic vortices or springs^{11,12}, for instance, the observation of new kinds of dichroisms^{7,8} or the visualization of dislocation strain fields in bulk material¹³.

The most common implementation of macroscopic beams carrying OAM, typically produced using spiral staircase phase plates, displays a helical phase front, with a phase singularity associated with zero intensity along the axis⁵. At the photon level, the twisted wavefront of the beam translates into a quantized momentum, with its component along the beam axis taking discrete $\ell\hbar$ values, where ℓ is a positive or negative integer. Laguerre-Gaussian modes (LG) are eigensolutions of the paraxial wave equation and form a natural set for these helically phased beams. Lately, motivated by theoretical predictions, the development of sources of XUV beams carrying OAM was undertaken on both quasi-continuous synchrotron installations¹⁴, and on femtosecond free electron lasers^{15,16}. High-harmonic generation (HHG)-based XUV sources constitute a table top, ultrashort and largely tunable alternative to those large-scale instruments. They show unrivalled stability specifications, especially useful for pump-probe experiments targeting attosecond temporal resolution¹⁷. They are based on the upconversion of a high-intensity femtosecond visible-infrared laser beam into the XUV range through highly nonlinear interaction with a gas. When driving HHG with helically phased light beams, momentum conservation predicts a ‘multiplicative’ rule for OAM transfer¹¹, similar to what is known in low-order nonlinear processes⁵. The OAM of the harmonic order q is then given by the quantum number $\ell_q = q\ell_1$, where ℓ_1 is the OAM of the driving field. Two groups measured the helicity of such a harmonic beam: first, Zürich *et al.*¹⁸ unexpectedly observed, on a single harmonic, that $\ell_q = \ell_1$. It was argued that this disagreement with theory, which was suggesting violation of momentum conservation, was due to parametric instabilities preventing higher-order vortices from propagating towards the detector¹⁹. Later, Gariépy *et al.*²⁰ succeeded, through fine interferometric measurements performed on three harmonics, in demonstrating the expected multiplicative rule. Therefore, OAM–HHG holds the potential for producing XUV pulses of attosecond duration carrying OAM. The shape of such a broad comb of phase-locked helically phased harmonics has been theoretically described¹¹. Such attosecond light springs, as they were dubbed¹², could in turn be a unique source to tailor attosecond electron springs through photoionization. Currently,

twisted electron beams, primarily generated through tailoring Gaussian electron beams in the quasi-static regime^{21,22}, are actively studied for future applications in fields covering spectroscopy of diluted and condensed matter²³, microscopy and particle physics²⁴.

In the present work, we report on the synthesis of attosecond XUV ‘light springs’. First, we show that the multiplicative OAM transfer rule for HHG is valid in our experimental conditions—without relying on any diffractive or interferometric technique. Importantly, this approach is directly scalable to an arbitrary spectral range. We then present the results of a two-colour ionization experiment in which we are able to characterize the attosecond structure of the generated light spring, which is in turn used to generate attosecond helically shaped electron beams.

Results

Link between OAM and divergence in HHG. Twisted attosecond XUV pulses hold great promises for a variety of applications. However, as raised up in ref. 20, a critical point is the characterization of OAM on a broad wavelength range. For instance, the study of helical dichroism in solids is envisioned close to transition-metal K-edges⁷, which lie at very short wavelengths. In much the same way, the extremely short time resolutions attainable using HHG-based sources are only reached when using a broad spectrum covering a large number of harmonics. In the visible-infrared range, the characterization is usually done directly using diffractive optical elements or interference schemes⁵. These approaches were transferred to the XUV domain in the aforementioned lines of work^{18,20}, requiring the resolution of fringes with extremely low spacing because of the short wavelength. These procedures clearly get ever more difficult to implement as the wavelength gets shorter. Here, for our broad harmonic comb, we use a different approach to measure the OAM of the harmonic field, which does not rely on interferometric measurements. It is based on another signature of the OAM value: the ring-shaped intensity radial profiles associated with the vortices’ phase singularity. In particular, it can be shown that the multiplicative law, $\ell_q = q \times \ell_1$, is the only one that yields a single ring with a constant diameter proportional to $\sqrt{|\ell_1|}$ over the whole spectrum. Indeed, unlike what is stated in the Supplementary Information of ref. 18, the enlargement of the ring’s diameter because of the increase in the OAM is exactly compensated for by the smaller diffraction observed at shorter wavelengths. This statement is supported by both a simple analytical model (see Supplementary Notes 1 and 2) and the results of numerical simulations of HHG²⁵ illustrated in Fig. 1. The simulations were carried out in a similar way as in ref. 11 but taking into account all aspects of a real experiment, including the propagation of the infrared beam towards the gas target (see Methods for more details). As shown in Fig. 1a, the harmonics at focus have a ring-shaped intensity as well as a spiral-shaped phase running q times 2π along the ring. When propagated to the far field, the harmonic profiles show one intense central ring (insets of Fig. 1b), suggesting that the generation is dominated by the emission of a single LG mode. Note that extra faint rings appear around it. They result from both the nonlinearity of the process and an interplay between two different contributions to HHG, associated with distinct electron quantum trajectories at the single atom level (so-called ‘short’ and ‘long’), as explained in ref. 26.

The diameter of the harmonic rings for two different values of ℓ_1 is plotted in Fig. 1b. As expected, the diameter is mostly constant apart from a jump around H25, which is precisely where the ‘short’ and ‘long’ trajectory contributions are merging. This leads to interferences and a modulation of the spatial profile of the harmonics, similarly to what is observed with Gaussian