

contributions of all emitted electrons in the interaction region. If we write the intensity coming from one point in this region, neglecting the contributions of the so-called atomic phase, we get for the 2ω oscillating component³³:

$$\begin{aligned} \text{SB}_{q+1}(\omega, R, \theta) &= \cos[2\omega\tau_0 + \varphi_{q+2} - \varphi_q + \Phi_{q+2}(R, \theta) - \Phi_q(R, \theta) - 2\Phi_{IR}(R, \theta)] \\ &= \cos[2\omega\tau_0 + \varphi_{q+2} - \varphi_q + (\ell_{q+2} - \ell_q - 2\ell_1)\theta], \end{aligned}$$

where ω is the angular frequency of the driving laser and φ_q the spectral phase of the q -th harmonic order. For the intensity to keep oscillating after integration over θ , which is the operation mode of our detector, the θ -dependent term must vanish, giving $\ell_{q+2} - \ell_q = 2\ell_1$. In particular, if dressing with $\ell_1 = 1$, this condition is only met when having $\ell_q = q\ell_1$. The observation of SBs in this case is therefore another measurement of the multiplicative rule for OAM transfer. Supplementary Fig. 5 shows that the 2ω oscillation component disappears when using $\ell_1 = 0$, for which the θ -dependent term does not cancel. The RABBIT³³ analysis of the SBs' oscillations was carried out under the assumption of fully coherent light³⁶.

Data availability. The data that support the findings of this study are available from the corresponding author (T.R.) upon request.

References

- Berestetskii, V., Lifshitz, E. & Pitaevski, L. *Quantum Electrodynamics* Vol. 4 (Butterworth-Heinemann, 1982).
- Allen, L., Beijersbergen, M. W., Spreeuw, R. J. C. & Woerdman, J. P. Orbital angular momentum of light and the transformation of Laguerre-Gaussian laser modes. *Phys. Rev. A* **45**, 8185–8189 (1992).
- Wang, J. *et al.* Terabit free-space data transmission employing orbital angular momentum multiplexing. *Nat. Photon.* **6**, 488–496 (2012).
- Fürhapter, S., Jesacher, A., Bernet, S. & Ritsch-Marte, M. Spiral interferometry. *Opt. Lett.* **30**, 1953–1955 (2005).
- Yao, A. M. & Padgett, M. J. Orbital angular momentum: origins, behavior and applications. *Adv. Opt. Photon.* **3**, 161–204 (2011).
- Toyoda, K., Miyamoto, K., Aoki, N., Morita, R. & Omatsu, T. Using optical vortex to control the chirality of twisted metal nanostructures. *Nano Lett.* **12**, 3645–3649 (2012).
- van Veenendaal, M. & McNulty, I. Prediction of strong dichroism induced by x rays carrying orbital momentum. *Phys. Rev. Lett.* **98**, 157401 (2007).
- Zambrana-Puyalto, X., Vidal, X. & Molina-Terriza, G. Angular momentum-induced circular dichroism in non-chiral nanostructures. *Nat. Commun.* **5**, 4922 (2014).
- Picón, A. *et al.* Photoionization with orbital angular momentum beams. *Opt. Express* **18**, 3660–3671 (2010).
- Scholz-Marggraf, H. M., Fritzsche, S., Serbo, V. G., Afanasev, A. & Surzhykov, A. Absorption of twisted light by hydrogenlike atoms. *Phys. Rev. A* **90**, 013425 (2014).
- Hernández-García, C., Picón, A., San Román, J. & Plaja, L. Attosecond extreme ultraviolet vortices from high-order harmonic generation. *Phys. Rev. Lett.* **111**, 083602 (2013).
- Pariante, G. & Quéré, F. Spatio-temporal light springs: extended encoding of orbital angular momentum in ultrashort pulses. *Opt. Lett.* **40**, 2037–2040 (2015).
- Takahashi, Y. *et al.* Bragg x-ray ptychography of a silicon crystal: visualization of the dislocation strain field and the production of a vortex beam. *Phys. Rev. B* **87**, 121201 (2013).
- Bahrtdt, J. *et al.* First observation of photons carrying orbital angular momentum in undulator radiation. *Phys. Rev. Lett.* **111**, 034801 (2013).
- Hemsing, E. *et al.* Coherent optical vortices from relativistic electron beams. *Nat. Phys.* **9**, 549–553 (2013).
- Rebernik-Ribic, P., Gauthier, D. & De Ninno, G. Generation of coherent extreme-ultraviolet radiation carrying orbital angular momentum. *Phys. Rev. Lett.* **112**, 203602 (2014).
- Corkum, P. B. & Krausz, F. Attosecond science. *Nat. Phys.* **3**, 381–387 (2007).
- Zürch, M., Kern, C., Hansinger, P., Dreischuh, A. & Spielmann, C. Strong-field physics with singular light beams. *Nat. Phys.* **8**, 743–746 (2012).
- Patchkovskii, S. & Spanner, M. Nonlinear optics: high harmonics with a twist. *Nat. Phys.* **8**, 707–708 (2012).
- Gariépy, G. *et al.* Creating high-harmonic beams with controlled orbital angular momentum. *Phys. Rev. Lett.* **113**, 153901 (2014).
- Uchida, M. & Tonomura, A. Generation of electron beams carrying orbital angular momentum. *Nature* **464**, 737–739 (2010).
- Verbeeck, J., Tian, H. & Schattschneider, P. Production and application of electron vortex beams. *Nature* **467**, 301–304 (2010).
- Asenjo-García, A. & García de Abajo, F. J. Dichroism in the interaction between vortex electron beams, plasmons, and molecules. *Phys. Rev. Lett.* **113**, 066102 (2014).
- Grillo, V. *et al.* Highly efficient electron vortex beams generated by nanofabricated phase holograms. *Appl. Phys. Lett.* **104**, 043109 (2014).
- Camper, A. *et al.* High-harmonic phase spectroscopy using a binary diffractive optical element. *Phys. Rev. A* **89**, 043843 (2014).
- Hernández-García, C., San Román, J., Plaja, L. & Picón, A. Quantum-path signatures in attosecond helical beams driven by optical vortices. *New J. Phys.* **17**, 093029 (2015).
- Zaïr, A. *et al.* Quantum path interferences in high-order harmonic generation. *Phys. Rev. Lett.* **100**, 143902 (2008).
- He, X. *et al.* Spatial and spectral properties of the high-order harmonic emission in argon for seeding applications. *Phys. Rev. A* **79**, 063829 (2009).
- Beijersbergen, M., Coerwinkel, R., Kristensen, M. & Woerdman, J. Helical-wavefront laser beams produced with a spiral phaseplate. *Opt. Commun.* **112**, 321–327 (1994).
- Caillat, J. *et al.* Attosecond resolved electron release in two-color near-threshold photoionization of N₂. *Phys. Rev. Lett.* **106**, 093002 (2011).
- Mauritsson, J. *et al.* Coherent electron scattering captured by an attosecond quantum stroboscope. *Phys. Rev. Lett.* **100**, 073003 (2008).
- Calegari, F. *et al.* Ultrafast electron dynamics in phenylalanine initiated by attosecond pulses. *Science* **346**, 336–339 (2014).
- Mairesse, Y. *et al.* Attosecond synchronization of high-harmonic soft X-rays. *Science* **302**, 1540–1543 (2003).
- Ammosov, M., Delone, N. & Krainov, V. Tunnel ionization of complex atoms and of atomic ions in an alternating electromagnetic field. *Soviet Physics - JETP* **64**, 1191–1194 (1986).
- Lewenstein, M., Balcou, P., Ivanov, M., L'Huillier, A. & Corkum, P. B. Theory of high-order harmonic generation by low-frequency laser fields. *Phys. Rev. A* **49**, 2117 (1994).
- Bourassin-Bouchet, C. & Couprie, M.-E. Partially coherent ultrafast spectroscopy. *Nat. Commun.* **6**, 6465 (2015).

Acknowledgements

We are particularly grateful to Vincent Gruson, Pascal Salières, Fabien Quéré and Bertrand Carré for stimulating discussions and fruitful suggestions. T.R. acknowledges Antonio Zelaquett Khoury (Univ. Fed. Fluminense, Brazil) for inviting him and introducing him to this topic. This work was supported by the French Agence Nationale de la Recherche (ANR) through XSTASE project (ANR-14-CE32-0010). A.C. acknowledges support of the US Department of Energy, Office of Science, Office of Basic Energy Sciences under contract DE-FG02-04ER15614.

Author contributions

R.G., A.C. and T.R. conceived, built and carried out the experiment and analysed the data. O.G. developed the laser system and the mode-filtering stage. T.A. did the HHG simulation. J.C. and R.T. did the RABBIT simulations. All authors contributed to the writing of the manuscript.

Additional information

Supplementary Information accompanies this paper at <http://www.nature.com/naturecommunications>

Competing financial interests: The authors declare no competing financial interests.

Reprints and permission information is available online at <http://npg.nature.com/reprintsandpermissions/>

How to cite this article: Géneaux, R. *et al.* Synthesis and characterization of attosecond light vortices in the extreme ultraviolet. *Nat. Commun.* **7**:12583 doi: 10.1038/ncomms12583 (2016).



This work is licensed under a Creative Commons Attribution 4.0 International License. The images or other third party material in this article are included in the article's Creative Commons license, unless indicated otherwise in the credit line; if the material is not included under the Creative Commons license, users will need to obtain permission from the license holder to reproduce the material. To view a copy of this license, visit <http://creativecommons.org/licenses/by/4.0/>

© The Author(s) 2016