Complete spatiotemporal and polarization characterization of ultrafast vector beams

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ABSTRACT

The use of structured ultrashort pulses with coupled spatiotemporal properties is emerging as a key tool for ultrafast manipulation. In particular, the ultrafast vector beams are opening exciting opportunities in different fields such as microscopy, time-resolved imaging, nonlinear optics, particle acceleration or attosecond science. We propose and demonstrate a technique for the full characterization of structured time-dependent polarization light waveforms with spatiotemporal resolution. We have developed and implemented a compact twofold spectral interferometer, based on in-line bulk interferometry and fibre-optic coupler assisted interferometry. We have experimentally measured structured infrared femtosecond vector beams, including radially polarized beams and complex-shaped beams exhibiting both temporal and spatial evolving polarization. Our measurements confirm that light waveforms with polarization evolving at the micrometer and femtosecond scales can be achieved through the use of s-waveplates and polarization gates. This new scale of measurement achieved will open the way to predict, check and optimize applications of structured vector beams at the ultrafast — femtosecond — and micrometer scales.

Introduction

During the last decades, the development of laser technology has boosted our ability to control the properties of ultrafast light pulses. Nowadays it is possible to routinely generate coherent radiation from the near-infrared to the soft x-rays1, that can be emitted in the form of few-cycle fs laser pulses2–4, or even attosecond pulses5. Furthermore, it is possible not only to tailor their spatiotemporal properties6–8, but also to structure ultrafast light fields in their angular momentum
properties, including both polarization and orbital angular momentum\textsuperscript{9–11}. The ultimate control of
the angular momentum properties of ultrafast laser pulses have opened new routes for the study of
chiral structures\textsuperscript{12–14}, topological systems\textsuperscript{15–17}, or magnetic materials\textsuperscript{18,19} at the ultrafast timescales.

One example of structured ultrafast light fields with tailored spatiotemporal and angular
momentum properties are the so-called vector beams, linearly polarized fields whose polarization
direction changes along the transverse spatial profile of the light beam\textsuperscript{20}. The paradigm of vector
beams is constituted by radially (RP) an azimuthally (AP) polarized beams. Interestingly, RP
beams allow to focus light below the diffraction limit\textsuperscript{21}, which enables applications in different
fields such as laser machining\textsuperscript{22,23} or particle acceleration\textsuperscript{24,25}, among others. AP beams can induce
longitudinal magnetic fields at the singularity of the electric field\textsuperscript{26} which offers potential
applications in spectroscopy and microscopy\textsuperscript{27}.

Nowadays vector beams can be routinely produced as continuum waves in the infrared (IR)
and visible regimes through the use of uniaxial and biaxial crystals\textsuperscript{28–30}, spatial light modulators\textsuperscript{31},
optical fibres\textsuperscript{32}, electrically-tuned q-plates\textsuperscript{33} or azimuthally dependent half-waveplates fabricated
by ultrafast laser nanostructuring of silica glass, also known as s-waveplates\textsuperscript{34}, among others. Recently, the generation of short pulsed, femtosecond vector beams\textsuperscript{35,36} has gained interest due to
their application in high harmonic generation and attosecond science\textsuperscript{37–39} or particle acceleration\textsuperscript{40}.
However, the advances of new laser sources and their applications is tied up to the development of
characterization techniques. Since the 1990s, different techniques have been used for the temporal
characterization of scalar —i.e., linearly polarized (LP)— ultrashort laser pulses\textsuperscript{41}. In the last two
decades, the problem of measuring spatiotemporal couplings in scalar beams has been tackled with
new strategies\textsuperscript{42–46}. In parallel, the reconstruction of time-evolving polarization pulses has also been
addressed\textsuperscript{47–51}. However, the necessity for the full spatiotemporal characterization of structured
ultrafast laser pulses, which includes both spatiotemporal and polarization properties, remained a
challenge up to now.

In this work we propose and demonstrate a technique to characterize the arbitrarily space-time
(and the space-frequency) polarization dependence of structured ultrafast light pulses. In particular
we perform the full characterization of infrared femtosecond vector beams generated through an s-
waveplate, which allow us: (i) to measure the spatiotemporal quality of RP pulses; (ii) to shape and
characterize time-dependent vector beams, structured through the use of polarization gates; and
(iii) to monitorize the focusing properties of structured vector beams. Our technique is based on
twofold spectral interferometry, both for the spatiotemporal reconstruction through spatially resolved spectral interferometry assisted by a fibre-optic coupler\textsuperscript{43} and for the polarization analysis through in-line bulk interferometry\textsuperscript{51}. The complete knowledge of the vector beam amplitude and phase allows to reconstruct the polarization state of the beam profile (including intensity, azimuth, and relative phase between the polarization components) both in the space-frequency and space-time domains. We demonstrate that spectral interferometry is a powerful technique for the characterization of ultrafast vector beams, opening the route for a new set of characterization techniques of structured light waveforms, to be used in a diverse range of applications\textsuperscript{49,52,53} that make use of spatial and temporal polarization shaping.

**Technique for complete spatial, temporal and polarization resolved characterization.**

The technique that we introduce to perform the complete spatiotemporal and polarization characterization is based on spectral interferometry. A scheme of the experimental setup is shown in Fig. 1. The laser output is divided into two replicas, one of them is used as a reference (known), while the other beam is shaped in its polarization components –using an s-waveplate and retarder waveplates–, conforming the unknown beam to be characterized.

In the unknown beam, a thick birefringent plate is placed to delay the horizontal and vertical polarization components of the beam. Afterwards, a linear polarizer (LP) is used to sample different projections of the beam. The reference beam is collected by a fixed position fibre port, while the unknown beam is spatially scanned (in the transverse XY plane) with a motorized fibre port. Both single-mode fibres are combined in a broadband fibre-optic coupler. The delay between the reference and the unknown beam is adjusted and fixed with the longitudinal position of the reference fibre. The known reference phase is measured with a standard temporal characterization technique\textsuperscript{41} (e.g., in the present case, the SPIDER technique\textsuperscript{54}).

In order to perform the complete characterization, first we measure the spatially-resolved spectral interferometry between the 0\textdegree-projection (X-component) of the unknown beam and the reference pulse, $S_{X+Ref}(\omega,x,y)$, by setting the LP horizontal. Then we obtain their relative phase using Fourier analysis\textsuperscript{55} (see details in Methods). Afterwards, we measure the spectrum profile of the same beam projection, $S_X(\omega,x,y)$. As the reference phase is known, we obtain the spatiotemporal (and spatio-spectral) amplitude and phase of the X-component of the unknown
beam. This strategy\textsuperscript{43} has been shown to be very versatile by use in the measurement of diffractive focusing, nonlinear processes and few-cycle pulse characterization\textsuperscript{8,56–58}, among others.

Second, we acquire the 90°-projection of the unknown beam, $S_Y(\omega; x, y)$ by placing the LP vertically. Finally, with the LP at 45°, we measure an intermediate projection of the X and Y components, $S_{X+Y}(\omega; x, y)$, which encodes their relative phase (see details in Methods). This allows us to accurately retrieve the phase of the Y component, and thus, the frequency-dependent and time-dependent polarization\textsuperscript{51}. The phase introduced by the birefringent plate is calibrated as described in the Methods Section. As the fibre scans the transverse profile of the unknown beam, we retrieve the full spatiotemporal (and spatiotemporal) polarization dependence of the beam.

![Experimental configuration for beam shaping and spatiotemporal polarization measurement](image)

**Figure 1 | Experimental configuration for beam shaping and spatiotemporal polarization measurement.** The X and Y components of the polarization shaped (unknown) beam are delayed with a birefringent plate. The unknown beam is spatially scanned with a fibre coupler. The X projection spectrally interferes with a known reference pulse, while the Y and X components spectrally interfere after a 45° linear polarizer (LP). Inset: definition of the polarization ellipse through the azimuthal angle $\chi$ and the ellipticity $\varepsilon = b/a$. 


Results

Characterization of radially polarized laser pulses

First, we have characterized a femtosecond RP beam created through an s-waveplate placed after the output of a chirped pulse amplification Ti:sapphire laser (see Methods for further details). In order to explore the polarization distribution of the beam along the azimuthal coordinate $\theta$, we scanned the XY plane through a circumference of radius $R = 3$ mm (corresponding to the half-maximum of the beam intensity) around the optical axis. In figure 2 we show the ellipticity ($\varepsilon$), azimuth ($\chi$) and intensity profiles of the pulse in the space-frequency (first row) and space-time (second row) domains. Whereas the ellipticity $\varepsilon$ is found to be close to zero (Figs. 2a and 2b), the azimuth $\chi$—which reflects the orientation angle of the LP field—describes the RP behaviour of the beam (Figs. 2b and 2e), corresponding to the $\chi$-varying LP beam created by the s-waveplate. Remarkably, we find the operation of the s-waveplate to imprint a RP profile can be considered homogeneous both in the spectral and temporal domains, despite the polychromatic nature of the pulse (25 nm of spectral bandwidth). The spatial dependence of the total spectral and temporal intensities (Fig. 2c and 2f) is inherent to the spatial profile of our laser beam (as shown in Supplementary Section S.I.). The full results are shown in Supplementary Section S.II.

Figure 2 | Measurement of the beam with radial polarization. a,d Ellipticity $\varepsilon$, b,e polarization azimuthal angle $\chi$, and c,f spectrum/intensity. Row 1: spatiotemporal dependence, Row 2: spatiotemporal dependence. The input laser pulses had a 10 nm spectral bandwith (FWHM) and 100 fs duration. The azimuthal angle $\theta$ was sampled in 90 points. The
magnitudes $\varepsilon$, $\chi$ and $\theta$ are defined in the inset of Fig. 1. The results presented in this and following figures are cut below the 1% of the spectral/temporal peak signal.

**Shaping and characterization of time-dependent ultrafast vector beams**

In order to show the ability of our technique to characterize vector beams whose polarization distribution varies temporally in the femtosecond timescale, we structure a laser pulse through the use of polarization gates. When combining an s-waveplate with different types and sets of waveplates, the resulting beam presents a strong spatiotemporal coupling in its intensity and polarization parameters. One set of waveplates that is particularly interesting is the so-called narrow polarization gate, which is used to effectively generate shorter pulses in certain applications as e.g. isolated attosecond pulse generation\textsuperscript{59–61}. In the Methods Section we detail the operation of the narrow gate using a multiple-order waveplate (QWM) and a zero-order waveplate (QW0).

In this experiment, instead of using a spatially-uniform LP beam, we illuminate the narrow gate setup (i.e. the two waveplates, first QWM with fast axes at $0^\circ$ and next QW0 at $-45^\circ$) using the RP described in the previous Section. The global effect of the narrow gate is that of producing two circular polarization (CP) pulses delayed 100 fs (similar to the pulse duration) with opposite helicity. When using a RP vector beam, the constant orientation angle (azimuth $\chi$) of the LP light entering the narrow gate depends on the azimuthal angle $\theta$, and thus the relative amplitude of both CP pulses depends on $\theta$. When both amplitudes are equal, this is for $\theta = \pi/4 + m\pi/2$, the narrow gate operates normally (see in Methods), producing LP in the centre and CP in the edges of the pulse in the time domain (Fig. 3b), whereas the spectral domain ellipticity (Fig. 3a) is close to $\varepsilon \approx 0$ (as measured in\textsuperscript{51}). For the azimuthal angles $\theta$ in the RP beam where the LP is parallel to the axes of the QWM (see example in Methods), i.e. $\theta = m\pi/2$, there is only one of the two CP pulses, either being the first (fast axis) or second (slow axis) pulse, which is seen in the temporal intensity (Fig. 3c), while both, the temporal and spectral ellipticities are $\varepsilon = 1$ (constant CP). For intermediate azimuthal angles $\theta$, the temporal ellipticity minimum ($\varepsilon = 0$) is shifted towards the less intense pulse, being the position where the two overlapping pulses have the same amplitude (Fig. 3b). Therefore, the whole evolution of the polarization shows a strong dependence in the space and time coordinates (Fig. 3d and Supplementary Section S.III). In Figs. 3e-h, we show the results of our numerical simulations performed as described in the Methods Section. The good
agreement between the experimental and theoretical results serves as a validation of our characterization technique. As found in the previous Section, here the small deviations of the experimental results are also due to the spatial profile of the laser beam.

**Figure 3** | Measurement and simulation of the spatiotemporal and spatiotemporal structure of a radially polarized beam followed by a narrow polarization gate. a,e Spatiotemporal ellipticity, b,f spatiotemporal ellipticity, c,g spatiotemporal intensity, and d,h selection of spatiotemporal polarization ellipses for different scanning positions. Row 1: experiment, Row 2: simulation. The azimuthal angle $\theta$ is scanned in 90 points with a radius R=3 mm. In the ellipticity, the colour scale varies from LP ($\varepsilon = 0$, blue) to CP ($\varepsilon = 1$, red).

**Focusing monitorization of structured time-dependent vector beam pulses**

Many applications of vector beams, e.g. particle trapping, microprocessing, particle acceleration or nonlinear optics, are carried out at their focus position. Our characterization technique allows us to perform the full characterization both at the far-field and at the near-field. Here we analyse and compare the spatiotemporal polarization dynamics at the focus of the two previously presented vector beams, i.e., the RP vector beam and the RP beam followed by the narrow polarization gate. In both cases, the beam was focused using an achromatic lens with a focal length of 50 cm. In these experiments the ultrafast vector beam presents a rich evolution both in the radial and azimuthal coordinates. Therefore, we did a two-dimensional spatial scan along the transverse XY plane.
First, when using a purely RP beam, the ellipticity $\varepsilon(x, y, t)$ is ideally zero for every position $x, y$ and time $t$. As shown before, although there is a spatiotemporal intensity modulation, there is no substantial temporal dependence of the polarization parameters of the beam. In Fig. 4 c1, we show the spatial profile at the focus position for a temporal instant that corresponds to the peak of the pulse. We notice that, instead of an ideally homogeneous ring shape of a RP, the measured beam presents spatial intensity modulations due to the above-mentioned inhomogeneities of the input spatial profile. On the other hand, the spatial distributions of the X and Y polarization projections (Figs. 4 a1, b1, respectively) correspond to that of a RP with the singularity at the centre (see the azimuth $\chi$ profile in Fig. 4 e1). In the Supplementary Movie 1 we show that the structure of these magnitudes of the focused RP beam is preserved in time. In the spectral domain, the dependence of the homologous magnitudes is analogous, also being wavelength independent, as shown in the Supplementary Section S.IV and in Supplementary Movie 2.

Contrarily to this case, when focusing the RP beam after illuminating the narrow gate, the beam exhibits a temporal/spectral polarization evolution together with the spatial dependence. In this second case the intensity ring is split into two lobes with time-dependent orientations, except for the centre of the pulse (the mean propagating time of the beam components passing through the fast and slow axes of the QWM), where the ring is recovered (Fig. 4 c3), as in the case of a focused RP beam (Fig. 4 c1). The X and Y projections of this temporal snapshot of the intensity corresponds to two lobes oriented at $\pm 45^\circ$ (Fig. 4 a3, b3). In the temporal leading edge of the beam – corresponding to the fast axis component at QWM– the two lobes are oriented in the x-axis for the total intensity as well as for the X and Y projections (Fig. 4 a2-c2). Contrarily, in the temporal trailing edge, the two lobes are oriented in the y-axis (Fig. 4 a4-c4), as the slow axis of the QWM is oriented vertically. The complete temporal evolution is shown in Supplementary Movie 3, where the total intensity evolves from two vertical lobes until they completely fill the ring and then they split into two horizontal lobes. Thus, the X and Y intensity projections consist in two spatial lobes evolving from vertical to horizontal but rotating in opposite direction. Regarding the ellipticity (Fig. 4 d3), it describes symmetrical CP in the $x$ and $y$ axes directions (where there is spatial superposition of the X and Y projections), while presenting LP out of those axes. The polarization azimuth (Fig. 4 e3) of the LP contributions mentioned before is oriented at $0^\circ$ and $90^\circ$ corresponding to the $\pm 45^\circ$ quadrants of the X ($+45^\circ$) and Y ($-45^\circ$) intensity projections (Fig. 4 a3-b3). We also
found that the ellipticity of the two lobes is $\varepsilon = 1$ (CP) both for the leading and trailing edges of the beam (Fig. 4 d2, d4), and there is a gradual temporal evolution (Supplementary Movie 3).

In the spatiotemporal domain, the frequency dependence presents some similarities (as shown in the Supplementary Section S.IV and in Supplementary Movie 4). The X and Y projections of the spatially resolved spectrum correspond to two lobes with different orientations in the XY-plane rotating as a function of the wavelength and forming a ring for certain intermediate wavelengths, while the total spectrum forms a ring with the cross shape in the ellipticity described for Fig. 4 d3 (CP in the x and y axes, LP elsewhere), being frequency-independent.

Figure 4 | Temporal snapshots of the spatial (x,y) profile and polarization parameters of the measured focused vector beams. a Horizontal projection of the intensity, b vertical projection of the intensity, c total intensity, d ellipticity, e azimuthal angle. Row 1:
comparison with the focus for the radially polarized beam at the pulse peak (at 20 fs). **Rows 2-4:** selections for three different times, respectively, -100, 20 and 120 fs, for the radially polarized beam followed by a narrow gate. The spatial scan was performed in a square grid with 21 x 21 points using a step of 18 μm.

**Discussion**

Tailoring light beams in full dimensionality, i.e., both spatial and temporal shaping of the individual light waveforms on a femtosecond time-scale, is nowadays possible. Our results demonstrate spectral interferometry as a suitable technique for performing a complete spatiotemporal and spatiopectral characterization of such ultrafast beams, whose polarization changes in time and space. The use of spectral interferometry is advantageous as the detection is fully linear (except for the reference measurement) and the data processing is fast, direct and univocal, as well as the acquired data being minimal for this level of measurements. The use of a birefringent plate and a fibre-optic coupler to implement a twofold interferometer avoids using multiple standard interferometers and alignment of beam recombination.

Regarding the vector beam shaping, we show that the combination of spatially varying polarization with temporal polarization shaping can produce singular spatiotemporal polarization dependences. By using a radially polarized beam to illuminate a narrow polarization gate, we create a complex vector beam with different orientations of the polarization gate or constant circular polarization, depending on the azimuthal angle in the transverse plane. We show experimentally that the temporal evolution of the focus of such a beam presents rich dynamics in contrast to focused radially polarized beams. The typical ring mode of focusing is effectively shortened in time because of the narrow polarization gate, which could be used for example to manipulate or trap nanoparticles during shorter times. This can be advantageous when using few-cycle pulses, due to the dispersion in media for ultrabroadband pulses.

In conclusion, the technique presented here constitutes the first accomplishment, to the best of our knowledge, of full spatiotemporally resolved polarization measurements at the femtosecond scale. It can be applied for far-field and near-field measurements, allowing in situ diagnostics within the region of interest in most experiments. As a consequence, a complete characterization of ultrashort vector beams can now be performed, allowing, for example, for quantitative measurements of imperfections and small deviations of spatiotemporal polarization distributions from ideal vector beams (as shown in this work). Such accurate measurements can be of capital
relevance in applications in particle trapping or acceleration, extreme ultraviolet pulse generation through high harmonic generation, or particle acceleration, where the quality of the beam mode is fundamental.

Our results pave the way for the full characterization of the most complex waves created up to now. Some examples include ultrafast beams carrying both spin and orbital angular momentum, whose quality is very important if applied to high nonlinear processes such as high harmonic generation\(^9\), light beams with time-dependent orbital angular momentum properties\(^{10}\), synthetic chiral fields\(^{14}\), or bicircular fields composed of counter rotating fields of different frequencies\(^{19,62,63}\). The emergence of these complex beams has opened opportunities, for example, to resolve the enantiomeric response of chiral molecules or to perform ultrafast studies of magnetic materials. Further advances in these new directions of ultrafast science are ultimately related to our ability to develop characterization techniques of these new laser sources.

**Methods**

**Experimental details.** The laser beam employed to perform the measurements was delivered by a chirped pulse amplification Ti:sapphire laser system (Spectra Physics Inc.), with central wavelength at 797 nm and spectral bandwidth of 10 nm FWHM (full width half maximum) at a repetition rate of 1 kHz. When compressed, the laser pulses have a duration of 100 fs FWHM. The fibre-coupler is made of broadband single-mode fibres centred at 800 nm, being both input arms almost equal-length so that their dispersion is compensated. The relative dispersion due to small difference (~1 mm) is calibrated with spectral interferometry. The thick birefringent plate used was a 3-mm calcite plate (Altechna) with the fast axis oriented vertically. The multiple-order waveplate QWM is a 3-mm quartz plate operating as quarter-wave plate for 806 nm. The zero-order waveplate QW0 is a 1.3-mm quartz plate designed for quarter-wave operation at 800 nm. The spectra were acquired with a fibre-coupled spectrometer (Avantes). The spatial scan in the XY plane was done with a two-axes motorized stage (Thorlabs). The reference pulse was characterized with a SPIDER measurement\(^{54}\). The s-waveplate for 800 nm wavelength was fabricated by ultrafast laser nanostructuring of silica glass.
Spectral interferometry and data analysis. In spectral interferometry, two delayed collinear pulses interfere in the spectral domain and their spectral fringes encode their relative phase as given in

$$S_{\text{total}}(\omega) = S_1(\omega) + S_2(\omega) + 2\sqrt{S_1(\omega)S_2(\omega)}\cos(\phi_1(\omega) - \phi_2(\omega) - \omega \tau)$$  \hspace{1cm} (1)$$

The relative phase can be extracted by Fourier-transform Spectral Interferometry (FTSI) analysis of the fringes$^{55}$. The pulse delay $\tau$ must be high enough to separate the signals in the time domain after Fourier-transform, as well as small enough to resolve the interferences in the spectrometer. The delay introduced by the birefringent plate between the X and Y components of the unknown beam is 1.8 ps, determined by the plate thickness. In the spectral interferometry between the reference and the X component of the beam, we manually introduced a 2 ps delay. The spectral bandwidth of the unknown pulse must be less or equal than the reference spectrum that interfere, and their relative amplitude must be of the same order to obtain better contrast fringes. The spatial resolution of the technique is related to the mode-field diameter of the single-mode optical fibre, being in the present case 4 $\mu$m.

As the reference pulse is characterized —the phase $\phi_{\text{Ref}}(\omega)$ is known—, the phase of the horizontal projection of the unknown vector beam, $\phi_X(\omega; x, y)$, is obtained from the spatially-resolved spectral interferences with the reference pulse, given by

$$S_{X+\text{Ref}}(\omega; x, y) = S_X(\omega; x, y) + S_{\text{Ref}}(\omega) + 2\sqrt{S_X(\omega; x, y)S_{\text{Ref}}(\omega)}\cos(\phi_X(\omega; x, y) - \phi_{\text{Ref}}(\omega) - \omega \tau)$$  \hspace{1cm} (2)$$

Then, from the interference between the projections at 45° of the X and Y components of the unknown beam, the phase of the vertical projection of the unknown vector beam, $\phi_Y(\omega; x, y)$, is obtained as

$$S_{X+Y}(\omega; x, y) = \frac{1}{2} S_X(\omega; x, y) + \frac{1}{2} S_Y(\omega; x, y) + \sqrt{S_X(\omega; x, y)S_Y(\omega; x, y)}\cos \left( \phi_Y(\omega; x, y) - \phi_X(\omega; x, y) + \phi_{YX,\text{BP}}(\omega; x, y) \right)$$  \hspace{1cm} (3)$$

where $\phi_{YX,BP}$ is the relative phase of the birefringent plate eigenaxes. The calibration of this phase is described below.

The spatial scans of the individual spectra, $S_X$ and $S_Y$, are optional as they can be obtained with the FTSI algorithm from the measurement of $S_{\text{Ref}}$ and the spectral interferometry scans given above, i.e., $S_{X+\text{Ref}}$ and $S_{X+Y}$. Nevertheless, as they can be directly measured, we acquired them, since the
performance of the FTSI algorithm is improved when subtracting the individual spectra from the interferences before data processing.

**Calibrations.** The global dispersion of the birefringent plate can be calculated from the thickness and the refractive index using Sellmeier equations. However, the accurate knowledge of the relative phase between the fast and slow axes is critical for the correct retrieval of the beam polarization. This calibration depends on the thickness and alignment of the birefringent plate. In order to calibrate the system we used a linearly polarized pulse, with no time evolving polarization, at 45° before the birefringent plate\textsuperscript{51}. From its own interferences, with the projection $S_{X+Y}$, we retrieved accurately the relative dispersion of the birefringent plate. In our measurements, we repeated this calibration after any realignment. We also found that the calibration of the birefringent plate did not depend on the transverse spatial position. Furthermore, with the same calibrating pulse (linearly polarized at 45°), we measured $S_X$ and $S_Y$ at the same sampling position, to calibrate the amplitude response of the system, which we used to correct the measurements of the individual spectra, $S_X$ and $S_Y$, of the unknown beam.

**Models for the simulations.** To simulate the shaped vector beam shown in this work, we started from a homogeneous beam, plane wave, using the experimental spectral amplitude of the laser output. To model the zero-order and multiple-order quartz waveplates, we firstly calibrated their thickness and retardation (if previously unknown) using spectral interferometry in combination with our detection (birefringent plate, LP and spectrometer). In the simulations, we calculated the dispersion of their eigenaxes from Sellmeier equations and then we imposed the known retardation for the corresponding operation wavelength. Naturally, we applied every element considering the described orientations.

For the s-waveplate, we modelled it as a half-wave plate with the fast axis orientation depending on the azimuthal angle $\theta$. As a reference, when the s-waveplate is oriented to create RP from input horizontal linear polarization, the fast axis orientation is $\theta/2$. We operated in the space-frequency domain, and at the end we obtained the space-time dependence by Fourier transformation.

**Generation of the narrow polarization gate.** The experimental implementation for the narrow polarization gate consists of using two consecutive quarter-waveplates, the first multiple-order
QWM and the second zero-order QW0, with relative eigenaxes at 45º. In the scheme of Fig. 5, the fast axes of QWM and QW0 are located at 0º and 45º, respectively. To operate as a narrow gate, the system is illuminated with LP pulses at 45º with respect to QWM (upper drawing). The thickness of the QWM introduces a delay (of the order of the pulse duration) between the X (fast) and Y (slow) polarization projections. After the QWM, there is a superposition of both delayed pulse projections, producing LP at 0º and LP at 90º at the leading and trailing edges of the pulse, respectively. Due to the dephase of the QWM, there is CP in the centre of the pulse (same amplitude of the projections). When this pulse impinges the second waveplate (QW0), the leading and trailing edges of the pulse switch to left- and right-handed CP, respectively, while the CP is converted to LP, producing a sharp minimum of ellipticity in the centre of the pulse, known as narrow polarization gate\textsuperscript{51,59–61}. Contrarily, if the waveplates are illuminated with input LP at 0º (lower scheme in Fig. 5), this is preserved after QWM (having a neutral axis at 0º). The LP is converted to constant CP after QW0 because the 45º-angle between the LP pulse and the eigenaxes of the waveplate. For other input LP pulses, the narrow gate will operate differently, as we show and discuss in the corresponding Results Sections when illuminating with a radially polarized beam.

**Figure 5 | Scheme of the operation of the narrow polarization gate implementation.** A multiple-order QWM and a second of zero-order QW0 quarter-waveplate are oriented with their fast axes at 0º and 45º, respectively. When the input pulse is LP at 45º (upper drawing), the narrow gate operates normally and produces a pulse with CP at the edges and LP at the centre. If the input pulse is LP at 0º (lower drawing), the output pulse has constant CP. The ellipticity $\varepsilon$ is represented in the given colour scale.
References
1. Popmintchev, T. et al. Bright Coherent Ultrahigh Harmonics in the keV X-ray Regime from Mid-Infrared Femtosecond Lasers. Science 336, 1287–1291 (2012).
2. Wirth, A. et al. Synthesized Light Transients. Science 334, 195–200 (2011).
3. Silva, F. et al. Strategies for achieving intense single-cycle pulses with in-line post-compression setups. Opt. Lett. 43, 337–340 (2018).
4. Butler, T. P. et al. Watt-scale 50-MHz source of single-cycle waveform-stable pulses in the molecular fingerprint region. Opt. Lett. 44, 1730–1733 (2019).
5. Brabec, T. & Krausz, F. Intense few-cycle laser fields: Frontiers of nonlinear optics. Rev. Mod. Phys. 72, 545–591 (2000).
6. Li, Y. & Lewellen, J. W. Generating a Quasiellipsoidal Electron Beam by 3D Laser-Pulse Shaping. Phys. Rev. Lett. 100, 74801 (2008).
7. Vincenti, H. & Quéré, F. Attosecond Lighthouses: How To Use Spatiotemporally Coupled Light Fields To Generate Isolated Attosecond Pulses. Phys. Rev. Lett. 108, 113904 (2012).
8. Alonso, B., Pérez-Vizcaíno, J., Mínguez-Vega, G. & Sola, Í. J. Tailoring the spatio-temporal distribution of diffractive focused ultrashort pulses through pulse shaping. Opt. Express 26, 10762–10772 (2018).
9. Dorney, K. M. et al. Controlling the polarization and vortex charge of attosecond high-harmonic beams via simultaneous spin–orbit momentum conservation. Nat. Photonics 13, 123–130 (2019).
10. Rego, L. et al. Generation of extreme-ultraviolet beams with time-varying orbital angular momentum. Science 364, eaaw9486 (2019).
11. Ghadimi Nassiri, M. & Brasselet, E. Multispectral Management of the Photon Orbital Angular Momentum. Phys. Rev. Lett. 121, 213901 (2018).
12. Cireasa, R. et al. Probing molecular chirality on a sub-femtosecond timescale. Nat. Phys. 11, 654–658 (2015).
13. Neufeld, O. et al. Ultrasensitive Chiral Spectroscopy by Dynamical Symmetry Breaking in High Harmonic Generation. Phys. Rev. X 9, 31002 (2019).
14. Ayuso, D. et al. Synthetic chiral light for efficient control of chiral light–matter interaction. Nat. Photonics 13, 866–871 (2019).
15. Spektor, G. et al. Revealing the subfemtosecond dynamics of orbital angular momentum in nanoplasmonic vortices. Science 355, 1187 (2017).
16. Shen, Y. et al. Optical vortices 30 years on: OAM manipulation from topological charge to multiple singularities. Light Sci. Appl. 8, 1–29 (2019).
17. Silva, R. E. F., Jiménez-Galán, Á., Amorim, B., Smirnova, O. & Ivanov, M. Topological strong-field physics on sub-laser-cycle timescale. Nat. Photonics 13, 849–854 (2019).
18. Kimmel, A. V. et al. Ultrafast non-thermal control of magnetization by instantaneous photomagnetic pulses. Nature 435, 655–657 (2005).
19. Fan, T. et al. Bright circularly polarized soft X-ray high harmonics for X-ray magnetic circular dichroism. Proc. Natl. Acad. Sci. U. S. A. 112, 14206–11 (2015).
20. Zhan, Q. Cylindrical vector beams: from mathematical concepts to applications. Adv. Opt. Photonics 1, 1 (2009).
21. Dorn, R., Quabis, S. & Leuchs, G. Sharper Focus for a Radially Polarized Light Beam. Phys. Rev. Lett. 91, 233901 (2003).
22. Meier, M., Romano, V. & Feurer, T. Material processing with pulsed radially and azimuthally polarized laser radiation. Appl. Phys. A 86, 329–334 (2007).
23. Hnatovsky, C., Shvedov, V., Krolikowski, W. & Rode, A. Revealing local field structure of focused ultrashort pulses. Phys. Rev. Lett. 106, (2011).
24. Salamin, Y. I., Harman, Z. & Keitel, C. H. Direct High-Power Laser Acceleration of Ions for Medical Applications. Phys. Rev. Lett. 100, 155004 (2008).
25. Marceau, V., Varin, C., Brabec, T. & Piché, M. Femtosecond 240-keV electron pulses from direct laser acceleration in a low-density gas. Phys. Rev. Lett. 111, (2013).
26. Blanco, M. et al. Ultraintense Femtosecond Magnetic Nanoprobes Induced by Azimuthally Polarized Laser Beams. ACS Photonics 6, 38–42 (2019).
27. Guclu, C., Veysi, M. & Capolino, F. Photoinduced Magnetic Nanoprobe Excited by an Azimuthally Polarized Vector Beam. ACS Photonics 3, 2049–2058 (2016).
28. Shvedov, V. G., Hnatovsky, C., Shostka, N. & Krolikowski, W. Generation of vector bottle beams with a uniaxial crystal. J. Opt. Soc. Am. B 30, 1 (2013).
29. Phelan, C. F., Donegan, J. F. & Lunney, J. G. Generation of a radially polarized light beam using internal conical diffraction. Opt. Express 19, 21793 (2011).
30. Turpin, A. et al. Polarization tailored novel vector beams based on conical refraction. Opt. Express 23, 5704 (2015).
31. Beversluis, M. R., Novotny, L. & Stranick, S. J. Programmable vector point-spread function engineering. Opt. Express 14, 2650 (2006).
32. Hirayama, T., Kozawa, Y., Nakamura, T. & Sato, S. Generation of a cylindrically symmetric, polarized laser beam with narrow linewidth and fine tunability. Opt. Express 14, 12839 (2006).
33. D’Ambrosio, V., Baccari, F., Slussarenko, S., Marrucci, L. & Sciarino, F. Arbitrary, direct and deterministic manipulation of vector beams via electrically-tuned q-plates. Sci. Rep. 5, (2015).
34. Beresna, M., Gecievicius, M., Kazansky, P. G. & Gertus, T. Radially polarized optical vortex converter created by femtosecond laser nanostructuring of glass. Appl. Phys. Lett. Phys. Lett 98, (2011).
35. Carbajo, S. et al. Efficient generation of ultra-intense few-cycle radially polarized laser pulses. Opt. Lett. 39, 2487 (2014).
36. Kong, F., Larocque, H., Karimi, E., Corkum, P. B. & Zhang, C. Generating few-cycle radially polarized pulses. Optica 6, 160 (2019).
37. Hernández-García, C. et al. Extreme ultraviolet vector beams driven by infrared lasers. Optica 4, 520–526 (2017).
38. Turpin, A., Rego, L., Picón, A., San Román, J. & Hernández-García, C. Extreme Ultraviolet Fractional Orbital Angular Momentum Beams from High Harmonic Generation. Sci. Rep. 7, (2017).
39. Kong, F. et al. Vectorizing the spatial structure of high-harmonic radiation from gas. Nat. Commun. 10, (2019).
40. Varin, C. et al. Direct Electron Acceleration with Radially Polarized Laser Beams. Appl. Sci. 3, 70–93 (2013).
41. Walmsley, I. A. & Dorrer, C. Characterization of ultrashort electromagnetic pulses. Adv. Opt. Photonics 1, 308–437 (2009).
42. Bowlan, P., Gabolde, P. & Trebino, R. Directly measuring the spatio-temporal electric field of focusing ultrashort pulses. Opt. Express 15, 10219–10230 (2007).
43. Alonso, B. et al. Spatiotemporal amplitude-and-phase reconstruction by Fourier-transform of interference spectra of high-complex-beams. J. Opt. Soc. Am. B 27, 933–940 (2010).
44. Miranda, M. et al. Spatiotemporal characterization of ultrashort laser pulses using spatially
resolved Fourier transform spectrometry. *Opt. Lett.* **39**, 5142–5145 (2014).

45. Pariente, G., Gallet, V., Borot, A., Gobert, O. & Quéré, F. Space-time characterization of ultra-intense femtosecond laser beams. *Nat. Photonics* **10**, 547–553 (2016).

46. Dorrer, C. Spatiotemporal Metrology of Broadband Optical Pulses. *IEEE J. Sel. Top. Quantum Electron.* **25**, 1–16 (2019).

47. Walecki, W. J., Fittinghoff, D. N., Smirl, A. L. & Trebino, R. Characterization of the polarization state of weak ultrashort coherent signals by dual-channel spectral interferometry. *Opt. Lett.* **22**, 81–83 (1997).

48. Schlup, P., Masihzadeh, O., Xu, L., Trebino, R. & Bartels, R. A. Tomographic retrieval of the polarization state of an ultrafast laser pulse. *Opt. Lett.* **33**, 267–269 (2008).

49. Rhee, H. *et al.* Femtosecond characterization of vibrational optical activity of chiral molecules. *Nature* **458**, 310–313 (2009).

50. Sato, M. *et al.* Terahertz polarization pulse shaping with arbitrary field control. *Nat. Photonics* **7**, 724–731 (2013).

51. Alonso, B. & Sola, I. Measurement of Ultrashort Vector Pulses From Polarization Gates by In-Line, Single-Channel Spectral Interferometry. *IEEE J. Sel. Top. Quantum Electron.* **25**, 8900307 (2019).

52. Misawa, K. Applications of polarization-shaped femtosecond laser pulses. *Adv. Phys. X* **1**, 544–569 (2016).

53. Chen, J., Wan, C. & Zhan, Q. Vectorial optical fields: recent advances and future prospects. *Sci. Bull.* **63**, 54–74 (2018).

54. Iaconis, C. & Walmsley, I. A. Spectral phase interferometry for direct electric-field reconstruction of ultrashort optical pulses. *Opt. Lett.* **23**, 792–794 (1998).

55. Lepetit, L., Chériaux, G. & Joffre, M. Linear techniques of phase measurement by femtosecond spectral interferometry for applications in spectroscopy. *J. Opt. Soc. Am. B* **12**, 2467–2474 (1995).

56. Mendoza-Yero, O. *et al.* Spatio-temporal characterization of ultrashort pulses diffracted by circularly symmetric hard-edge apertures: Theory and experiment. *Opt. Express* **18**, (2010).

57. Alonso, B., Miranda, M., Sola, I. J. & Crespo, H. Spatiotemporal characterization of few-cycle laser pulses. *Opt. Express* **20**, 17880–17893 (2012).

58. Alonso, B. *et al.* Characterization of sub-two-cycle pulses from a hollow-core fiber compressor in the spatiotemporal and spatio-spectral domains. *Appl. Phys. B* **112**, 105–114 (2013).

59. Tcherbakoff, O., Mevel, E., Descamps, D., Plumridge, J. & Constant, E. Time-gated high-order harmonic generation. *Phys. Rev. A* **68**, 043804 (2003).

60. Sansone, G. *et al.* Isolated single-cycle attosecond pulses. *Science* **314**, 443–446 (2006).

61. Sola, I. J. *et al.* Controlling attosecond electron dynamics by phase-stabilized polarization gating. *Nat. Phys.* **2**, 319–322 (2006).

62. Chen, C. *et al.* Tomographic reconstruction of circularly polarized high-harmonic fields: 3D attosecond metrology. *Sci. Adv.* **2**, e1501333 (2016).

63. Jiménez-Galán, Á. *et al.* Attosecond recorder of the polarization state of light. *Nat. Commun.* **9**, 850 (2018).
Acknowledgements
We acknowledge funding from Junta de Castilla y León (SA287P18) and FEDER Funds, and from Spanish Ministerio de Economía y Competitividad (FIS2016-75652-P, FIS2017-87970-R, EQC2018-004117-P). C.H.-G. acknowledges support Ministerio de Ciencia, Innovación y Universidades for a Ramón y Cajal contract (RYC-2017-22745), cofunded by the European Social Fund. B.A. acknowledges funding from the European Union’s Horizon 2020 research and innovation programme under the Marie Skłodowska-Curie Individual Fellowship grant agreement No. 798264. PGK acknowledges assistance of Dr. R. Drevinskas and support of ERC project ENIGMA. This project has received funding from the European Research Council (ERC) under the European Union’s Horizon 2020 research and innovation programme (grant agreement No. 851201).

Author contributions statement
IJS and BA conceived the idea and designed the experiments. BA conducted the experiments. IJS and ILQ participated in the experiments. WH collaborated in the experimental setup. PGK fabricated and provided the s-waveplate. BA analysed the experimental results and performed the simulations and the graphics. IJS, BA and CHG discussed the results and wrote the first manuscript. All authors revised the manuscript.

Additional information
Supplementary information is available in the online version of the paper.

Competing financial interests
BA declares past employment with Sphere Ultrafast Photonics SA (Portugal), a company that sells devices for the temporal measurement and compression of ultrashort laser pulses.
Complete spatiotemporal and polarization characterization of ultrafast vector beams

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Summary

In this Supplementary material we show enlarged and detailed information regarding the results presented in the main manuscript.

S.I. Characterization of the input laser beam

In this Section we show the measurement of the laser beam used for the experiments. As in the first part of the manuscript, we perform a circular scan with radius R=3 mm.

Figure S1 | Measurement of the input laser beam. a Spatially resolved spectrum, and b spatiotemporal intensity.
S.II. Characterization and simulation of radially polarized laser pulses

In this Section we show the experimental measurements compared to the theoretical simulations of a radially polarized beam, corresponding to the manuscript section: Results || Characterization of radially polarized laser pulses.

Figure S2 | Measurement (panel 1) and simulation (panel 2) of the radially polarized beam in the spati spectral domain. a horizontal projection of the spectrum, b vertical projection of the spectrum, c total spectrum, d dephase, e ellipticity, f azimuthal angle.
Figure S3 | Measurement (panel 1) and simulation (panel 2) of the radially polarized beam in the spatiotemporal domain. a horizontal projection of the intensity, b vertical projection of the intensity, c total intensity, d dephase, e ellipticity, f azimuthal angle.
S.III. Shaping and characterization of time-dependent ultrafast vector beams

In this Section we show the experimental measurements compared to the theoretical simulations of a radially polarized beam followed by a narrow polarization gate, corresponding to the manuscript section: Results || Shaping and characterization of time-dependent ultrafast vector beams.

Figure S4 | Measurement (panel 1) and simulation (panel 2) of the radially polarized beam followed by a narrow polarization gate in the spatio-spectral domain. a horizontal projection of the spectrum, b vertical projection of the spectrum, c total spectrum, d dephase, e ellipticity, f azimuthal angle.
Figure S5 | Measurement (panel 1) and simulation (panel 2) of the radially polarized beam followed by a narrow polarization gate in the spatiotemporal domain. a horizontal projection of the intensity, b vertical projection of the intensity, c total intensity, d dephase, e ellipticity, f azimuthal angle.
S.IV. Focusing monitorization of structured time-dependent vector beam pulses

In this Section we show the spectral results for the experimental measurements of the focusing of a radially polarized beam followed by a narrow polarization gate (Fig. S5, rows 2-4), corresponding to the manuscript section: Results || Focusing monitorization of structured time-dependent vector beam pulses. The results are compared to the focusing of a radially polarized beam (Fig. S5, row 1). The full spatio-spectral results are expanded to see the wavelength-dependent evolution in Supplementary Movies 2 and 4, already described in the manuscript.

Figure S5 | Spectral snapshots of the measurement of the radially polarized beam followed by a narrow polarization gate in the spatial (x,y) domain. a Horizontal projection of the spectrum, b vertical projection of the spectrum, c total spectrum, d ellipticity, e azimuthal angle. **Row 1**: comparison with the focus for the radially polarized beam for 800 nm. **Rows 2-4**: selections for three different wavelengths, respectively, 795, 800 and 803 nm.