Supplementary Materials for

Synthesizing ultrafast optical pulses with arbitrary spatiotemporal control

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The PDF file includes:

Texts S1 to S6
Figs. S1 to S11
Legends for movies S1 and S2
References

Other Supplementary Material for this manuscript includes the following:

Movies S1 and S2
S1. Spatiotemporal shaping of ultrafast pulses: separating spatial and temporal masking functions

An input femtosecond pulse train in the time-domain is equivalent to a series of equally spaced spectral lines at frequencies $\nu_j$ ($j$ is an integer) in the frequency domain. The corresponding angular frequencies are expressed as, $\omega_j = 2\pi \nu_j = 2\pi (j \nu_{\text{rep}} + \nu_0)$, where $\nu_{\text{rep}}$ is the repetition rate of the pulse train and $\nu_0$ is the offset frequency. Assuming an input beam with Gaussian spatial distribution, the transform-limited electric field $\vec{E}_{\text{in}}$ of a $p$-polarized (electric-field oscillating along the $x$-direction) input pulse train can be expressed by its Fourier series:

$$\vec{E}_{\text{in}}(x, y, t) = \sum_j E(x)E(y)E(\omega_j, t) \left[ \begin{array}{c} 1 \\ 0 \end{array} \right] = \sum_j e^{-\frac{x^2}{\sigma^2}}e^{-\frac{y^2}{\sigma^2}}a_j e^{-i\omega_j t} \left[ \begin{array}{c} 1 \\ 0 \end{array} \right], \quad \text{(S1)}$$

where spatial components $E(x), E(y)$ and temporal components $E(\omega_j, t)$ are treated as separable variables, $\sigma$ is the spatial beam-waist radius, $a_j$ is the spectral amplitude, and $\left[ \begin{array}{c} 1 \\ 0 \end{array} \right]$ is the Jones vector that represents the input polarization state.

In the Fourier transform pulse shaper (Fig. 3), the input grating diffracts each frequency component $\omega_j$ into a specific angle exiting the $x$-$y$ plane. Without losing generality, the effect of the grating is to add a linear phase to the input field along the $x$-direction, resulting in a field $\vec{E}_{G1}$ at the exit of the input grating expressed as:

$$\vec{E}_{G1}(x, y, t) = G\{\vec{E}_{\text{in}}(x, y, t)\} = \sum_j E(x)e^{\frac{i2\pi x}{\Lambda_0}}E(y)a_j e^{-i\omega_j t} \left[ \begin{array}{c} 1 \\ 0 \end{array} \right], \quad \text{(S2)}$$

where $G$ represents the grating function and $\Lambda_0$ is the grating pitch. A comprehensive derivation leading to the same results can be performed by following the approach presented in ref. 39.

Next, the angularly dispersed spectral components are focused by an off-axis parabolic mirror onto the metasurface placed at the Fourier plane (defined as the $\xi$-$\eta$ plane) in a 2-$f$ configuration. The electric field in the $\xi$-$\eta$ plane at the input of the metasurface, $\vec{E}_-(\xi, \eta, t)$, is the spatial Fourier transform of $\vec{E}_{G1}(x, y, t)$:

$$\vec{E}_-(\xi, \eta, t) = \mathcal{F}_{xy \rightarrow \xi \eta}\{\vec{E}_{G1}(x, y, t)\} = \sum_j u_j(j - \xi_j)u_j(\eta)j e^{-i\omega_j t} \left[ \begin{array}{c} 1 \\ 0 \end{array} \right]. \quad \text{(S3)}$$

Here, $\xi_j = (\lambda_j - \lambda_0)f/\Lambda_0$, and $u_j(j)$ is the one-dimensional (1D) spatial Fourier transform of the Gaussian spot for the spectral line $\nu_j$, given by:

$$u_j(\xi) = \mathcal{F}_{x \rightarrow \xi}\left\{ e^{-\frac{x^2}{\sigma^2}} \right\} = e^{-\left(\frac{\xi}{\nu_j}\right)^2}, \quad \text{(S4)}$$
where $w_j = \lambda_j f / \pi \sigma$ is the diffraction-limited beam waist of the spectral line $\nu_j$ at the Fourier plane.

In the following, we show that polarization-multiplexed, spatiotemporal pulse shaping can be achieved by using the metasurface design scheme described below. Note that $E_-(\xi, \eta, t)$ is the summation of a 1D array of diffraction-limited, quasi continuous-wave (cw) Gaussian spots, representing a beam consisting of different spectral lines spatially dispersed along the $\xi$-axis at the Fourier plane. The Jones matrix of the metasurface $M(\xi, \eta)$ can be written as a complex $2 \times 2$ matrix; hence, the electric field distribution of the beam after passing through the metasurface $\tilde{E}_+(\xi, \eta, t)$ is given by:

$$\tilde{E}_+(\xi, \eta, t) = \left[ \begin{array}{c} E_+^p(\xi, \eta, t) \\ E_+^s(\xi, \eta, t) \end{array} \right] = M(\xi, \eta) E_-(\xi, \eta, t)$$

$$= \left[ \begin{array}{cc} M_{11} & M_{12} \\ M_{21} & M_{22} \end{array} \right] \left[ \begin{array}{c} E_-(\xi, \eta, t) \\ 1 \end{array} \right]$$

$$= \left[ \begin{array}{c} M_p(\xi, \eta) E_-(\xi, \eta, t) \\ M_s(\xi, \eta) E_-(\xi, \eta, t) \end{array} \right].$$

(S5)

The detailed implementation of the four elements of $M(\xi, \eta)$, representing a polarization multiplexing functionality, is described in section S3. Here, we take $M_p(\xi, \eta)$ as an example. Note that $\xi_j \propto \lambda_j$; we design a metasurface consisting of $Q = 201$ superpixels, each nominally manipulating a $\Delta \lambda \approx 1$ nm bandwidth of the pulse spectrum ranging from 700 nm to 900 nm. For each $j \in [1, Q]$, the $j$-th superpixel, centered at $\xi_j = (\lambda_j - \lambda_c) f / \Lambda_c$, simultaneously imparts a targeted spectral modulation $Y_j^p(\omega_j)$, which is spatially constant across each superpixel, and a targeted spatial modulation $\Gamma_j^p(\xi, \eta)$, varying within each superpixel, on the subgroup of spectral lines centered at wavelength $\lambda_j = (699 + j)$ nm. The targeted masking function of the metasurface $M_p(\xi, \eta)$ can be expressed as a two-dimensional (2D) concatenated function:

$$M_p(\xi, \eta) = \sum_{j=1}^{Q} Y_j^p(\omega_j) \Gamma_j^p(\xi - \xi_j, \eta) \Pi(\xi - \xi_j).$$

(S6)

Here, $\Pi(\xi)$ is a rectangular function defined as:

$$\Pi(\xi) = \begin{cases} 1, & -\frac{D}{2} \leq \xi < \frac{D}{2}, \\ 0, & \text{else} \end{cases}$$

(S7)

where $D = f \Delta \lambda / \Lambda_c$ is the width of the superpixel.

To further simplify $E_+^p(\xi, \eta, t)$, noting $\Delta \xi_j \ll w_j$, given $Q$ is large and the input spectrum is slow-varying, $E_-(\xi, \eta, t)$ can be resampled with $Q$ spectral lines and expressed as:
\[ E_-(\xi, \eta, t) = \sum_j u_j(\xi - \xi_j)u_j(\eta)a_je^{-i\omega_jt} \]
\[ = \sum_{j=1}^{Q} \left( \sum_{\lambda_j \Delta \lambda_j \leq \lambda_j \Delta \lambda_j + \frac{\Delta \lambda_j}{2}} u_j(\xi - \xi_j)u_j(\eta)a_je^{-i\omega_jt} \right) \approx \sum_{j=1}^{Q} a_j\Pi(\xi - \xi_j)u_j(\eta)e^{-i\omega_jt}, \]

With this approximation, we have:

\[ E^P_+(\xi, \eta, t) = M_p(\xi, \eta)E_-(\xi, \eta, t) \]
\[ = \left\{ \sum_{j=1}^{Q} Y_j^p(\omega_j)\Gamma_j^p(\xi - \xi_j, \eta)\Pi(\xi - \xi_j) \right\}\left\{ \sum_{j=1}^{Q} a_j\Pi(\xi - \xi_j)u_j(\eta)e^{-i\omega_jt} \right\} \approx \sum_{j=1}^{Q} \left\{ a_jY_j^p(\omega_j)e^{-i\omega_jt}\right\}\left\{ \Gamma_j^p(\xi - \xi_j, \eta)\Pi(\xi - \xi_j)u_j(\eta) \right\} \]

Going back to the 4-f configuration, the second identical off-axis parabolic mirror performs the inverse spatial Fourier transform on the modulated \( E^P_+(\xi, \eta, t) \), resulting in the field \( \vec{E}_{G2} \) at the input of the second grating, expressed as:

\[ \vec{E}_{G2}(x, y, t) = \mathcal{F}^{-1}|_{\xi \eta \rightarrow xy}\{ E^P_+(\xi, \eta, t) \} \]
\[ = \sum_{j=1}^{Q} a_jY_j^p(\omega_j)e^{-i\omega_jt}\mathcal{F}^{-1}\left\{ \Gamma_j^p(\xi - \xi_j, \eta)\Pi(\xi - \xi_j)u_j(\eta) \right\} e^{\frac{i2\pi jx}{\lambda_j \eta}}. \]

Finally, the second grating recombines the angularly dispersed electric fields in Eq. S10, resulting in a train of spatiotemporally shaped output pulses, with \( E^P_{\text{out}}(x, y, t) \) expressed as:

\[ E^P_{\text{out}}(x, y, t) = \sum_{j=1}^{Q} a_jY_j^p(\omega_j)e^{-i\omega_jt}\mathcal{F}^{-1}\left\{ \Gamma_j^p(\xi, \eta)\Pi(\xi)u_j(\eta) \right\} e^{\frac{i2\pi jx}{\lambda_j \eta}} e^{-\frac{2\pi}{\wG}x}. \]
\[ \sum_{j=1}^{Q} a_j Y_j^p(\omega_j) e^{-i\omega_j t} F^{-1}\{ \Gamma_j^p(\xi, \eta) \Pi(\xi) u_j(\eta) \} \]

\[ \triangleq \sum_{j=1}^{Q} E_{\text{out}, j}^p(\omega_j, t) E_{\text{out}, j}^p(x, y). \]

Following the same procedure for the s-polarization, the full output electric field can be expressed as:

\[ \bar{E}_{\text{out}}(x, y, t) = \begin{bmatrix} E_{\text{out}}^p(x, y, t) \\ E_{\text{out}}^s(x, y, t) \end{bmatrix} = \sum_{j=1}^{Q} E_{\text{out}, j}^p(\omega_j, t) E_{\text{out}, j}^p(x, y) \]

\[ = \sum_{j=1}^{Q} E_{\text{out}, j}^s(\omega_j, t) E_{\text{out}, j}^s(x, y) \]  \hspace{1cm} (S12)

### S2. Jones Calculus for Polarization Multiplexing

In section S1, we denote the Jones matrix of each polarization-multiplexed metasurface \(M(\xi, \eta)\) to be a complex 2×2 term, given by \(M(\xi, \eta) = \begin{bmatrix} M_p(\xi, \eta) & M_{12} \\ M_{12}^* & M_{22} \end{bmatrix}\) (Eq. S5). Here, given a targeted \(M_p(\xi, \eta)\) and \(M_s(\xi, \eta)\), we describe the metasurface design process for each pixel. The polarization multiplexing function at the single pixel level is achieved via a dielectric nanopillar of rectangular cross-section acting as a birefringent waveplate providing phase-shifts \(\Phi_1\) and \(\Phi_2\) for light polarized along its major and minor axes. When rotated counter-clockwise with respect to the x-direction by an angle \(\theta\), the Jones matrix of the birefringent nanopillar can be expressed as:

\[ M(\Phi_1, \Phi_2, \theta) = \begin{bmatrix} \cos \theta & -\sin \theta \\ \sin \theta & \cos \theta \end{bmatrix} \begin{bmatrix} e^{i\Phi_1} & 0 \\ 0 & e^{i\Phi_2} \end{bmatrix} \begin{bmatrix} \cos \theta & \sin \theta \\ -\sin \theta & \cos \theta \end{bmatrix} \]

\[ = \begin{bmatrix} e^{i\Phi_1} \cos^2 \theta + e^{i\Phi_2} \sin^2 \theta & (e^{i\Phi_1} - e^{i\Phi_2}) \cos \theta \sin \theta \\ (e^{i\Phi_1} - e^{i\Phi_2}) \cos \theta \sin \theta & e^{i\Phi_1} \sin^2 \theta + e^{i\Phi_2} \cos^2 \theta \end{bmatrix}. \]  \hspace{1cm} (S13)

For \(\theta = \frac{\pi}{4}\), Eq. S13 simplifies to:

\[ M \left( \Phi_1, \Phi_2, \frac{\pi}{4} \right) = \frac{1}{2} \begin{bmatrix} e^{i\Phi_1} + e^{i\Phi_2} & e^{i\Phi_1} - e^{i\Phi_2} \\ e^{i\Phi_1} - e^{i\Phi_2} & e^{i\Phi_1} + e^{i\Phi_2} \end{bmatrix}. \]  \hspace{1cm} (S14)

Hence, the values of \(\Phi_1\) and \(\Phi_2\) can be obtained from:
\[
\begin{align*}
\{ e^{i\Phi_1} &= M_p(\xi, \eta) + M_s(\xi, \eta) \\
\{ e^{i\Phi_2} &= M_p(\xi, \eta) - M_s(\xi, \eta). 
\end{align*}
\] (S15)

Finally, the nanopillar in-plane dimensions \( L_1(\xi, \eta), L_2(\xi, \eta) \) for each metasurface pixel are calculated using the FOM minimization procedure described in the Material and Methods section.

S3. Design of spectral phases for polarization shaping:

To generate the time-varying polarization-swept pulses (Fig. 2A), the spectral phases \( \varphi_1 \) and \( \varphi_2 \) are designed to take the following form:

\[
\begin{align*}
\varphi_1(\omega_j) &= \frac{b_{II}}{2} (\omega_j - \omega_c)^2 + b_I (\omega_j - \omega_c) \\
\varphi_2(\omega_j) &= \frac{b_{II}}{2} (\omega_j - \omega_c)^2 - b_I (\omega_j - \omega_c),
\end{align*}
\] (S16)

where \( b_I \) and \( b_{II} \) are the group delay and group delay dispersion, respectively. Here, we only focus on the time-varying polarization shaping, thus ignore the spatial part of the masking function. Substituting Eq. S16 into Eq. S15 and comparing with Eqs. S11 and S12 yields:

\[
\bar{E}_{\text{out}}(t) = \sum_{j=1}^{Q} a_j e^{-i\omega_j t} e^{i\frac{b_{II}}{2}(\omega_j - \omega_c)^2} \begin{bmatrix} \cos\left(b_I (\omega_j - \omega_c)\right) \\ i \sin\left(b_I (\omega_j - \omega_c)\right) \end{bmatrix}.
\] (S17)

A quarter-wave plate (QWP) with its fast axis aligned along the x-direction (\( \theta = 0\pi \)), placed after the shaper (Fig. 3), transforms the polarization state into:

\[
\bar{E}_{\text{out QWP}}(t) = \sum_{j=1}^{Q} a_j e^{-i\omega_j t} e^{i\frac{b_{II}}{2}(\omega_j - \omega_c)^2} \begin{bmatrix} \cos\left(b_I (\omega_j - \omega_c)\right) \\ -\sin\left(b_I (\omega_j - \omega_c)\right) \end{bmatrix}.
\] (S18)

where the corresponding \( \bar{E}_{\text{out}}(t) \) and \( \bar{E}_{\text{out QWP}}(t) \) are shown in Fig. S7.

In addition to the two representative cases with and without the QWP shown in Fig. S7, the overall pulse envelope, plotted as a curve on the surface of the Poincaré sphere for \( \bar{E}_{\text{out QWP}}(t) \) can be readily reoriented by simply rotating the fast axis of the QWP with respect to the x-axis by an angle \( \theta \) (Fig. S8).
S4. Effect of the superpixel boundaries

To examine the effect of the superpixel boundaries on the synthesis of OAM beams, we consider three superpixels (Fig. S9A), centered at $\lambda_j=101 = \lambda_c = 800$ nm, within the metasurface. Again, we first analyze the $p$-polarization; the corresponding spatial masking function term in Eq. S6 can be expressed as:

$$\Gamma_j^p(\xi - \xi_j, \eta) = \begin{cases} 
e^{-\ell \tan^{-1} \frac{\eta}{\xi - \xi_j}}, & 100 \leq J \leq 102, \\
0, & \text{else} \end{cases}$$

(S19)

where $\ell = -1$. The output electric field resulting from superpixel at $J = 101$ is simulated by only considering spectral lines incident on that superpixel. Five representative input cw beams (wavelengths $\lambda_j$ varying from 799.50 nm to 800.50 nm in 0.25 nm steps) are depicted, including two (positions a and e) at the boundaries between superpixels, two (positions b and d) at off-center positions and one (position c) at the center of the superpixel at $J = 101$ (Fig. S9B). Their electric fields are assumed to have a Gaussian input spatial distribution:

$$E_{-j}(\xi, \eta) = u_j(\xi - \xi_j)u_j(\eta),$$

(S20)

with $w_j \approx 25 \mu m$. The spatial distribution $E_j^p(u, v, z)$ of the beam at a $uv$-plane after the metasurface superpixel (before the second parabolic mirror) can be calculated under Fresnel approximation as:

$$E_j^p(u, v, z) = \frac{e^{\frac{2\pi z}{\lambda_j}}}{i\lambda_j} e^{\frac{2i\pi}{\lambda_j}}(u^2+v^2) \int_{-\infty}^{\infty} E_{-j}(\xi, \eta)M_p(\xi, \eta)e^{\frac{i\pi}{\lambda_j^2}((\xi^2+\eta^2)} e^{-\frac{i2\pi}{\lambda_j^2}(u\xi+v\eta)} d\xi d\eta.$$

(S21)

The calculated intensity and phase distributions of $E_j^p(u, v, z)$, at a representative distance of $z = (1250 \cdot \lambda_c) = 1$ mm after the metasurface, are shown in Figs. S9C and S9D, respectively. These results are experimentally confirmed by directly measuring the transmission intensity, at $z = 1$ mm after the metasurface, upon illumination of the superpixel at $J = 101$ at five spatially shifted locations using a Gaussian cw laser beam at the wavelength of $\lambda_c$ and a nominal waist of $\approx 25 \mu m$ (Fig. S9E).

Next, we numerically simulate the experimental scenario by placing the three superpixels (Fig. S10A) at the Fourier plane of the pulse shaper and calculating the far-field electric-field distribution. The summation of all the spectral lines incident on the superpixel $S_j$, given by $\sum_{\lambda_j - \frac{\lambda_j}{2} \leq \lambda_j < \lambda_j + \frac{\lambda_j}{2}} u_j(\xi - \xi_j)u_j(\eta)a_j e^{-i\omega_j t}$ in Eq. S8, is represented by an effective input beam at $\lambda_j$ (Fig. S10B). The resulting output intensity and phase distribution of $E_{out,101}(x, y, t = 0)$ at $\lambda_j = 800$ nm, along with equivalent calculations performed for superpixels $J = 2$ (at $\lambda_j = 701$ nm) and $J = 200$ (at $\lambda_j = 899$ nm) are shown in Figs. S8C and S8D, respectively. The collective $I_{out}^p$ from all the 201 superpixels, shown in Fig. 4D, closely matches the experimentally measured $I_{out}^{(s)}(x, y)$ (Fig. 4A, insets).
We further examine the impact of approximation in Eq. S8, where the effective input beam is approximated by $a_J \Pi (\xi - \xi_J) \cdot u_J (\eta) e^{-i \omega_J t}$ (Fig. S10E). The resulting $\bar{I}^p_{\text{out},J}$ (Fig. S10F) and $\Psi^p_{\text{out},J}$ (Fig. S10G) corresponding to the same three superpixels show a similar intensity null and phase singularity at the beam center as Figs. S10C and S10D, respectively, confirming that the approximation is valid.

These simulations illustrate that the deviation of the far-field intensity profile $\bar{I}^p_{\text{out}}$ from an ideal circularly symmetric Laguerre-Gaussian mode originates from spectral lines illuminating the boundaries between adjacent superpixels, introducing vertical fringes in $\bar{I}^p_{\text{out}}$ (Fig. S10C). We perform a set of far-field numerical simulations with varying superpixel widths and an optimal width of $\approx 115 \ \mu m$ is identified as a compromise between minimizing boundary effects while maintaining high spectral resolution.

**S5. Extended control of spatiotemporal properties of ultrafast pulses**

Similar to how an ultrafast pulse train can be decomposed into a series of frequency components, each with a corresponding amplitude and phase, any static spatial mode profile can also be decomposed into appropriately weighted spatial modes from a complete modal basis. Hence, by coherently superimposing multiple frequencies spatially, where each frequency is carrying a different spatial mode, a dynamic spatiotemporal wave packet can be realized (25, 26). We leverage this capability using the superpixel approach to demonstrate independent and parallel control over the spatial and temporal properties of the frequencies constituting an ultrafast pulse train, allowing arbitrary synthesis of spatiotemporal wave packets. One set of azimuthal spatial modes that form a complete and orthogonal basis is a vortex beam carrying integer topological orders. As an illustrative example, here, we demonstrate extended spatiotemporal control of femtosecond pulses by assigning five different OAM orders, one each to a preassigned set of superpixels. Two categories of spatiotemporal wave packets are realized, as discussed below.

**Spatiotemporal intensity control: light-coil**

A light coil, as shown in Fig. 5B, represents a spatiotemporal intensity distribution that revolves around a central axis (26). Along the revolving trajectory, at each azimuthal angle, the light beam remains a transform-limited ultrafast pulse, but this pulse arrives at a later time for increasing azimuthal angles, forming a helical intensity distribution.

First, we consider the coherent superposition of the constituent frequencies, sampled by $Q$ superpixels, after passing through the pulse shaper. Here, we only consider the s-polarized output component and take advantage of the minimal unmodulated light in this polarization. To simplify the analysis without losing its generality, here, we ignore the boundary effects discussed in section S1 and S2, and assume that the superpixels impart ideal OAM modes to the constituent frequency lines. Rewriting Eqs. S11 and S12 into:
\[ E_{\text{out}}^s(x, y, t) = \sum_{j=1}^{Q} a_j Y_j^s(\omega_j) e^{-i\omega_j t} \mathcal{F}^{-1}\{ R_j^s(\xi, \eta) u_j(\xi) u_j(\eta) \} \]

\[ = \sum_{j=1}^{Q} a_j Y_j^s(\omega_j) e^{-i\omega_j t} \mathcal{F}^{-1}\left\{ e^{i\ell_j \tan^{-1} \eta / \xi} e^{-\xi^2 + \eta^2 / w_j^2} \right\}. \]  

Converting Eq. S22 from the Cartesian coordinate to the polar coordinate system and expressing the analytical solution of the 2D Fourier transform in the polar coordinate (30):

\[ E_{\text{out}}^s(\rho, \Omega, t) = \sum_{j=1}^{Q} a_j Y_j^s(\omega_j) e^{-i\omega_j t} 2\pi i \ell_j e^{i\ell_j \Omega} H_{\ell_j}(\rho), \]

where \( \Omega = \tan^{-1} \frac{\eta}{\xi} \), \( H_{\ell_j}(\rho) = \int_0^\infty r e^{-r^2 / w_j^2} B_{\ell_j}(2\pi \rho r) dr \) is the \( \ell_j \)-th order Hankel transform with \( B_{\ell_j} \) being the \( \ell_j \)-th order Bessel function of the first kind, \( r = \sqrt{\xi^2 + \eta^2} \), and \( \rho = \sqrt{x^2 + y^2} \).

To achieve the light-coil in Fig. 5B, we design \( Y_j^s(\omega_j) = 1 \) and \( \ell_j = \begin{cases} 2 - \left\lfloor \frac{J - 1}{40} \right\rfloor, & 1 \leq J \leq 200 \\ -2, & J = 201 \end{cases} \). The floor function in \( \ell_j \) indicates that the 201 superpixels are divided into five groups: each group of frequencies acquires a designed vortex spatial phase front upon exiting the pulse shaper. The effective central frequencies \( \nu_{\text{eff}} \) for the five groups are 416.4 THz, 394.5 THz, 374.7 THz, 356.9 THz, and 340.7 THz, corresponding to groups carrying a topological order \( \ell \) of +2, +1, 0, -1, and -2, respectively. At a given time \( t \), all the frequencies coherently interfere in space, leading to an azimuthally localized wave packet, with its azimuthal location determined by the phase relationship among constituting OAM orders (40). Because different OAM orders are carried by different groups of frequencies, this phase relationship varies as a function of time, producing a spatiotemporal light-coil. The effective revolving period can be estimated by considering that the effective time-varying phase relationship \( 2\pi \Delta \nu_{\text{eff}} t \) goes through \( 2\pi |\Delta \ell| \) within one period, which gives an effective period of \( |\Delta \ell| / \Delta \nu_{\text{eff}} \), where the sign of \( \Delta \ell / \Delta \nu_{\text{eff}} \) determines the revolving direction. Here, \( \Delta \ell = 1 \) for all neighboring groups, but \( \nu_{\text{eff}} \) has a non-constant spacing. The generated light-coil thus may have time-variant speed (25). We estimate an average revolving period of \( \approx 53 \) fs, which agrees well with the numerical simulation result shown in Fig. 5B.

The shaped light-coil is then characterized by performing spatial interference with a tilted reference Gaussian pulse at different time delay \( \tau \):

\[ I_{\text{interf}}(\rho, \Omega, \tau) = \left( E_{\text{out}}^s(\rho, \Omega, t) + E_{\text{ref}}(\rho, \Omega, t + \tau) \right) \left( E_{\text{out}}^s(\rho, \Omega, t) + E_{\text{ref}}(\rho, \Omega, t + \tau) \right)^*, \]

where \( E_{\text{ref}}(\rho, \Omega, t + \tau) = a_{\text{ref}}(\rho) e^{-i\omega_\text{ref}(t + \tau) + ik_x x} \) with \( a_{\text{ref}}(\rho) = \sqrt{\frac{\rho^2}{w_{\text{ref}}^2}} \), \( w_{\text{ref}} \) and \( k_x \) are the waist and wave-vector of the tilted reference beam, respectively, and \( x = \rho \cos \Omega \). The interferogram from Eq. S24 contains a cross term that exhibits vertical parallel fringes coming
from the $e^{-i k x^2}$ term and the fringe contrast is modulated by the revolving electric field amplitude represented in $E_{\text{out}}^i$ (numerical simulations in Supplementary Movie S1). To experimentally verify the azimuthally revolving intensity profile of the shaped pulse, we reconstruct its time-dependent intensity $I_{\text{meas}}(x_1, y_1, \tau)$ by calculating a moving contrast from the interferograms recorded at each time delay $\tau$. This contrast is defined by the difference between the maximum and minimum intensity of a $10 \times 10$ pixel region (determined by the interference fringe spacing) centered at a spatial location $(x_1, y_1)$. The contrast extraction is repeated for all $(x_1 + 2n_x, y_1 + 2n_y)$ coordinates, with $n_x = 1, 2, \ldots, 291$ and $n_y = 1, 2, \ldots, 236$, to generate $I_{\text{meas}}(x, y, \tau)$. The resulted $I_{\text{meas}}(x, y, \tau)$ clearly shows a revolving peak intensity as a function of time and is consistent with the numerical simulations.

**Spatiotemporal phase front control: time-varying OAM**

A light beam carries a time-varying OAM can be achieve by designing $Y_j^\pm(\omega_j) = e^{i b_n (\omega_j - \omega_c)^2}$ while keep the same $\ell_j = \left\{ \begin{array}{ll} 2 - \lfloor \frac{j - 1}{40} \rfloor, & 1 \leq j \leq 200 \\ -2, & j = 201 \end{array} \right.$. Here, the spectral phase term $Y_j^\pm(\omega_j)$ provides a chirp to the pulse. Groups of frequency comb lines carrying different OAM orders are thus engineered to arrive at different time delays, forming a spatiotemporal pulse carrying time-varying OAM (Fig. 5C).

Experimentally, we insert a 5 mm thick plate of glass after the pulse shaper, providing $b_\| = 180$ fs$^2$/rad, to positively chirp the pulse. Both numerical simulations and experimental results show vortex spatial structures varying from a topological order of $-1$ to 0 (Gaussian) then to $+1$ as time varies from $-26$ fs to $+46$ fs within the spatiotemporally shaped pulse (Supplementary Movie S2).

**S6. Instantaneous Stokes parameters**

The time-varying electric field for $p$- and $s$- polarized components can be expressed in terms of amplitude and phase as a function of time:

$$\begin{align*}
E_p(t) &= A_p(t)e^{i \Psi_p(t)} \\
E_s(t) &= A_s(t)e^{i \Psi_s(t)}
\end{align*}$$

At each time instant $t$, the normalized instantaneous Stokes parameters are then calculated using the following equations:

$$\begin{align*}
I(t) &= A_p^2(t) + A_s^2(t) \\
Q(t) &= \frac{(A_p^2(t) - A_s^2(t))}{I(t)} \\
U(t) &= 2A_p(t)A_s(t) \cos(\Delta \Psi(t))/I(t) \\
V(t) &= 2A_p(t)A_s(t) \sin(\Delta \Psi(t))/I(t)
\end{align*}$$

where $\Delta \Psi(t) = \Psi_s(t) - \Psi_p(t)$ is the relative phase between $s$- and $p$-polarized components.
Fig. S1. Spatial dispersion of $\lambda$ vs. $\xi$. (A) Normalized spectrum of an input pulse transmitted through a reference mask containing nine half-wave plate metasurface superpixels used for spectral calibration, and alignment of the metasurface at the Fourier plane. (B) The calibrated quasi-linear relationship between $\xi$ and $\lambda$ at the Fourier plane. The red circles correspond to the peaks in (A) and the blue line is calculated using $\xi_j = (\lambda_j - \lambda_c) f / \Lambda_G$, where $f = 381$ mm is the focal length of the parabolic mirror and $\Lambda_G = (1/300)$ mm is the grating pitch.
Fig. S2. Refractive index of a-Si. The real and imaginary part of the refractive index $n$ vs. $\lambda$, for a-Si, measured using spectroscopic ellipsometry. The a-Si film is deposited on an oxide coated reference Si substrate (thermal oxide thickness = 300 nm) using PECVD.
Fig. S3. Raman spectra of a-Si before and after exposure to electron beam. (A) The Raman spectra from a polished crystalline Si (c-Si) wafer (blue), PECVD deposited 650 nm thick a-Si film on a fused-silica substrate before (orange) and after (yellow) exposure to 100 keV electron beam. (B) Zoom-in plot of the Raman spectra corresponding to the pink dashed box in (A). The measurements are performed at room temperature using an excitation wavelength of 532 nm (incident power: ≈3 mW) focused onto the sample with a 50× objective (numerical aperture = 0.35, FWHM spot size: 760 nm). The measured spectra show no observable change between the a-Si samples before and after electron-beam exposure, indicating that any morphological change of the a-Si film after exposure is minimal, and the film remains amorphous.
Fig. S4. Representative nanopillar library to achieve the targeted masking function. (A and B) Targeted phase topographies $\Phi_1$ and $\Phi_2$, at $\lambda_{35} = 734$ nm, to achieve the masking function in Fig. 2. (C and D) Simulated nanopillar in-plane dimensions $L_1$ and $L_2$ that can deliver any combination of $\psi_1$ and $\psi_2$ covering the full $[0, 2\pi]$ range along the two birefringent axes. (E and F) Simulated transmission coefficients corresponding to the choice of $L_1$ and $L_2$ shown in (C) and (D). The colored dots in (C) to (F) correspond to the designs for eight representative targeted ($\Phi_1, \Phi_2$) combinations in (A) and (B).
Fig. S5. Simulated phase and intensity transmission maps for the eight representative superpixels shown in Fig. 2. (A and B) the simulated phase maps to achieve the $\Phi_1$ and $\Phi_2$ designs shown in Fig. 2D. (C and D) The simulated transmission maps $t_1^2$ and $t_2^2$ for the corresponding superpixels in (A) and (B). Scale bar: 100 μm. (E) The average $\bar{t}_1^2$ and $\bar{t}_2^2$ for each superpixel $S_J$. 
Fig. S6. SPIDER measurement of the reference pulse. (A-B) The temporal intensity (A) and spectral phase (B) of the reference pulse measured using the SPIDER setup.
Fig. S7. Time-varying polarization control. Simulated electric-field waveforms and corresponding time-varying polarization states (section S4) with $b_{\|} = 100 \text{ fs}^2/\text{rad}$ and $b_{\perp}/b_{\|} = 0.1 \text{ rad/fs}$. (A) With no QWP at the output of the pulse shaper, the polarization state of $\mathbf{E}_{\text{out}}(t)$, denoted by a curve on the Poincaré sphere, varies adiabatically from $-45^\circ$ linear $\rightarrow$ elliptical $\rightarrow$ horizontal linear $\rightarrow$ elliptical $\rightarrow$ $+45^\circ$ linear. (B) When a QWP, with its fast axis aligned parallel to the $x$-direction ($\vartheta = 0\pi$), is added after the pulse shaper, the corresponding polarization state of $\mathbf{E}^{\text{QWP}}_{\text{out}}(t)$ varies from left circular $\rightarrow$ elliptical $\rightarrow$ horizontal linear $\rightarrow$ elliptical $\rightarrow$ right circular. $Q(t), U(t), V(t)$ are the corresponding Stokes parameters (defined in Supplementary Text, S5).
Fig. S8. Ready time-varying polarization control using a QWP. (A-I) The instantaneous polarization states of simulated $\mathbf{E}_{\text{out}}(t)$ plotted on the surface of the Poincaré sphere as the fast-axis of the QWP at the output of the pulse shaper is rotated from $\vartheta = 0\pi$ to $\vartheta = \pi/2$ with respect to the $x$-direction.
Fig. S9. Superpixel boundary effect on the Fresnel far-field intensity distribution from a superpixel designed to impart a topological charge of $-1$. (A) $\Phi_p^J(\xi, \eta)$ for three adjacent superpixels $S_{j-1}$, $S_j$, and $S_{j+1}$ for $J = 101$, centered at $\lambda_c$, each designed to impart a 2D spiral phase with $\ell_j = -1$ to the corresponding $\omega_j$. Red trapezoids: five representative spatially shifted excitation positions (marked a-e). (B) Superpixel at $J = 101$ illuminated at the corresponding spatial locations marked a-e in (A). Dotted white lines indicate the boundary between adjacent superpixels. The excitation beam waist is $\approx 25 \mu m$. Numerically simulated far-field intensity (C) and phase (D) distributions, at $z = 1$ mm, upon transmission through the metasurface superpixel (each panel corresponds to the excitation condition in (B)). (E) Experimentally measured transmitted intensity distribution, at $z = 1$ mm, upon illumination of the superpixel at $J = 101$ with a cw-laser at 800 nm at the nominal locations indicated in (B). Scale bars: 100 $\mu$m.
Fig. S10. Superpixel boundary effect on the spatial term of the shaped output pulse $E^p_{\text{out},j}(x,y)$ from a superpixel designed to impart a topological charge of $-1$. 

(A) Representative $\Phi^p_j(\xi, \eta)$ for three adjacent superpixels $S_{j-1}$, $S_j$, and $S_{j+1}$. Scale bar: 100 $\mu$m. 

(B) The effective incident intensity distribution illuminating the superpixel $J = 101$ at $\lambda_J = 800$ nm. Scale bar: 100 $\mu$m. Calculated $\bar{I}^p_{\text{out},j}$ (C) and $\Psi^p_{\text{out},j}$ (D) for superpixels $J = 2$ (at $\lambda_J = 701$ nm), $J = 101$ (at $\lambda_J = 800$ nm), and $J = 200$ (at $\lambda_J = 899$ nm). Scale bars: 5 mm. 

(E) The approximated incident intensity distribution following Eq. S8 (scale bar: 100 $\mu$m), along with the corresponding $\bar{I}^p_{\text{out},j}$ (F) and $\Psi^p_{\text{out},j}$ (G) (scale bars: 5 mm).
Fig. S11. Mode-purity of the time-varying OAM pulse. Colored lines represent the retrieved mode-purity of OAM orders $\ell = -2$ (yellow), $-1$ (orange), 0 (blue), $+1$ (purple), $+2$ (green), respectively for the simulated time varying OAM pulse shown in Fig. 5C. Colored stars denote the retrieved mode-purity of the three experimental snapshots (Fig. 5C, Inset) for OAM orders $\ell = -1$ (orange), 0 (blue), $+1$ (purple), respectively.
**Movie S1. Spatiotemporal intensity control: light-coil.** An ultrafast pulse engineered to encode a time-varying spatial intensity distribution that revolves around a central axis. Numerically simulated $\Psi_{\text{sim}}(x,y,t)$ (top left) and $I_{\text{sim}}(x,y,t)$ (top right) of the designed light-coil exhibiting an azimuthally revolving intensity $I_{\text{sim}}$, within which the spatial phase front $\Psi_{\text{sim}}$ remains flat. Experimentally measured interferogram $I_{\text{intf}}(x,y,\tau)$ (bottom left), acquired by interfering $E_{\text{out}}$ with a reference $E_{\text{in}}$, shows parallel fringes with a revolving fringe contrast, confirming a flat phase front in the synthesized pulse. The reconstructed $I_{\text{meas}}(x,y,\tau)$, shown in the bottom right, closely matches $I_{\text{sim}}(x,y,t)$.

**Movie S2. Spatiotemporal phase control: time-varying OAM.** By chirping the frequency groups carrying different OAM orders, an ultrafast pulse is engineered to carry time-varying OAM. Top: numerically simulated $\Psi_{\text{sim}}(x,y,t)$ shows three different vortex phase topographies, varying from a topological order of $-1$ to 0 then to $+1$ as a function of time. Bottom: experimentally measured $I_{\text{intf}}(x,y,\tau)$ changes from fringes with a down-opening fork-dislocation to parallel fringes to fringes with an up-opening dislocation, as $\tau$ increases.
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