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Nonlinear Nanophotonic Chip-space Interfaces Enabled Multi-dimensional Engineering of Structured Optical Vortices

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Miniaturized and reconfigurable interfaces between confined optical modes within integrated photonic chips and structured light propagating in free space would serve as a cornerstone for fundamental optical science and modern photonic technology. In this work, we present nonlinear nanophotonic chip-space interfaces capable of flexibly generating and multi-dimensionally engineering structured light via injections of photons to on-chip waveguides. By harnessing the nonlinear Čerenkov radiation in integrated nonlinear microring resonators, we successfully tailor the spatial profile, polarization state, emission wavelength, topological charge and temporal wave packet of structured optical vortices, exhibiting reconfigurabilities and tuning ranges far beyond the state-of-the-art. To further showcase the capabilities of our platform, we use a single pump to generate multi-state integrated vortex microcombs in the visible range via synergistic $\chi^{(2)}$ and $\chi^{(3)}$ nonlinear optical processes, potentially boosting applications in biochemical sensing, optical clocks, and astronomical spectroscopy. Our work bridges the research fields of structured light and integrated optics, providing unprecedented opportunities for spatiotemporal light generation, on-chip nonlinear optics and integrated quantum photonics.

Structured light, whose characteristics can be arbitrarily tailored in all degrees of freedom, including frequency/wavelength, time, phase, amplitude and polarization etc, has profoundly reshaped the landscape of modern optical science and photonic technology [1, 2]. As a prominent example, optical vortices carrying orbital angular momentum (OAM) of light feature a spiral phase front of $e^{il\varphi}$, where topological charge (TC) l is an unbounded integer and φ is the azimuthal angle ranging from 0 to 2π [3]. These optical vortices allow the exploration of new concepts such as phase singularities, light topology, optical chirality in fundamental optical physics [4] and bring unprecedented opportunities to dramatically enlarge the capacities of optical communication and quantum information processing [5–7]. Beyond modern information technology, optical vortices have also revolutionized numerous applications including optical manipulation, microscopy, and holography [8–11]. Driven by the tremendously growing aggregated data traffic in the

big data era, a variety of chip-scale devices have emerged as promising solutions for creating optical vortices, featuring advantages in terms of small footprint, low-power consumption, high stability and compatibility with modern nanofabrication techniques [12–16]. Despite plausible progress in on-chip control of optical vortices [17–20], the tuning range and engineerable dimensionality for the integrated devices remain rather limited due to the fact that the underlying physical mechanism is mainly based on conventional linear optics. To date, optical vortices with controlled radiation characteristics such as spatial profile, polarization state, TC and temporal wave packet can only be generated via multiple bulk optical elements including forked holograms, spatial light modulators, spiral phase plates, and q-plates, etc [21]. Therefore, new operation principles and device architectures are urgently needed to fully explore the advantage of configurability and multi-dimensional engineering associated with integrated photonic platforms.

In this work, we theoretically propose and experimentally demonstrate universal chip-space interfaces capable of flexibly generating and on-demand engineering structured optical vortices in full dimensions by manipulating the propagating/confined optical modes and their nonlinear interactions in integrated photonic chips. Different from the widely employed integrated OAM emitters consisting of microring resonators with embedded angular gratings as linear light scatterers [12, 13], we take advantage of the nonlinear $\chi^{(2)}$ interactions between the counter-propagating whispering gallery modes (WGMs) confined by nonlinear microring resonators, leading to the out-of-plane sum-frequency generation (SFG) carrying OAM of light assisted by nonlinear Čerenkov radiations (NCR). Though the NCR has been widely employed in light generations at different directions in bulk materials [22–25], its implications in integrated photonics are rather limited [26], lacking the realization in microresonator systems. Based on the microring resonators made of x -cut and z -cut thin film lithium niobate (TFLN), nonlinear structured optical vortices without the need for complete phase matching are experimentally demonstrated, enabling independent engineering of the spatial profile, polarization state, emission wavelength and TC of the sum-frequency (SF) wave in a reconfigurable manner. Furthermore, by combining the excellent $\chi^{(2)}$ and $\chi^{(3)}$ effects in the TFLN platform, multi-state integrated vortex microcombs with engineerable temporal wave packets are demonstrated, for the first time, to the best of our knowledge. Our work not only provides unexplored paradigm to produce and engineer structured light with integrated optics but also endows new functionalities to cutting-edge TFLN photonic devices and similar thin-film platforms with $\chi^{(2)}$ nonlinearities, particularly offering exciting opportunities for integrated quantum/nonlinear photonics.

ON-CHIP NONLINEAR ČERENKOV RADIATION FOR GENERATING OPTICAL VORTICES

Figure 1 shows the working principle of integrated nonlinear chip-space interfaces. In a straight waveguide with a $\chi^{(2)}$ nonlinearity, the nonlinear interaction between two counter-propagating guided modes at different fundamental frequencies produces a strong SF wave through NCR [26], which requires phase matching along the propagation direction of fundamental waves, i.e., $\Delta k_x = k_m - k_n - |\vec{k}_{\text{SF}}|\sin(\theta) = 0$, but allows phase mismatch in the plane perpendicular to the propagation direction, i.e., $\Delta k_z = |\vec{k}_{\text{SF}}|\cos(\theta) \neq 0$ (Fig. 1a). Here, k_m and k_n refer to moduli of the propagation constants of the two fundamental waves injected from the left and right sides, respectively, and $\vec{k}_{\text{SF}}$ is the wavevector of the produced SF wave emitting towards the free space.

Therefore, the radiated SF wave presents itself as a plane wavefront tilted at an angle of θ , governed by the NCR condition. If the waveguide is curved to form a loop, the guided fundamental waves will become confined WGMs satisfying the resonant conditions (Fig. 1b). Both k_m of the counterclockwise (CCW) wave and k_n of the clockwise (CW) wave circulate within the plane of the microring. The NCR requires phase matching along the azimuthal direction of $\hat{\varphi}$, so that the wavefront of the radiated SF wave should skew in the azimuthal direction, suggesting the creation of optical radiations with helical phase fronts, i.e., optical vortices. The azimuthal component of $\vec{k}_{\text{SF}}$ is labeled as $k_{\text{SF},\varphi}$, meeting the condition of $k_{\text{SF},\varphi} = k_m - k_n$, which produces an azimuthal momentum of $\hbar k_{\text{SF},\varphi}$ for the radiated SF wave. Considering the circular symmetry of the microring with a radius of R , this momentum leads to a z -axis OAM of $J = R\hbar k_{\text{SF},\varphi}$ [27]. Using the resonant conditions of $k_m R = m$ for the CCW mode and $k_n R = n$ for the CW mode, the obtained TC of OAM equals $J = (m - n)\hbar$. Here, m and n are positive integers, referring to azimuthal mode numbers. This result shows that the TC of the SF wave equals to the modal difference between the interacting CCW and CW WGMs, revealing the NCR condition in the microring governed by the conservation of azimuthal numbers, i.e., $l_{\text{SF}} = m - n$. Denoting the fundamental wavelengths as λ_m and λ_n , the SF wavelength can be expressed as $\lambda_{\text{SF}} = (\lambda_m \lambda_n) / (\lambda_n + \lambda_m)$. The NCR condition of $l_{\text{SF}} = m - n$ in the microring can be strictly proven using the nonlinear coupling wave equations in the cylindrical coordinate system, where WGM modes are the eigenmodes of the microring resonator (see Methods for theory on NCR in a microring resonator).

The aforementioned discussion provides an intuitive but simplified physical picture of the SF optical vortices generated by the on-chip NCR. In a real on-chip nonlinear optical frequency mixing process, several factors should be taken into consideration as follows: polarization states of WGMs, i.e., transverse electric (TE) modes or transverse magnetic (TM) modes; nonlinear polarization dependence on the polarization states of the fundamental waves and the $\chi^{(2)}$ tensor of the resonator material; and the diffraction of SF wave induced by sub-wavelength scale of the microring. Figure 1c illustrates the schematic of our device which consists of a microring resonator and a side-coupled bus waveguide. Such a standard and practical configuration not only enables simple excitations of CCW modes from the left port and CW modes from the right port but is also compatible with the well-established nanofabrication process for large-scale integration. The electric fields of the fundamental waves are labeled as $E_{\text{P},m}$ for CCW modes and $E_{\text{P},-n}$ for CW modes, respectively. The first subscript “P” refers to a TE-like or TM-like polarization state. The minus before the second subscript “ n ” in

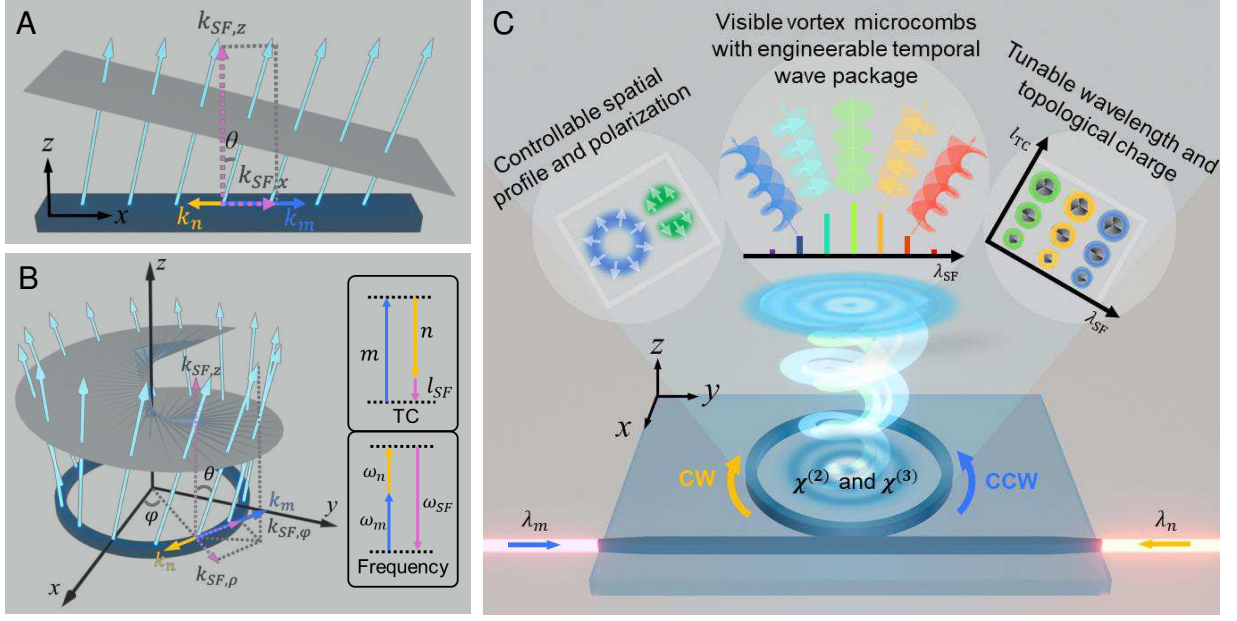


FIG. 1. **Working principle of the nonlinear nanophotonic chip-space interfaces based on NCR.** (a) Out-of-plane radiated SF waves from a waveguide at a certain angle determined by the NCR produced via the second-order nonlinear optical interaction between two counter-propagating fundamental waves with wavevectors of k_m and k_n . The radiation angle θ is determined by $\Delta k_x = k_m - k_n - |\vec{k}_{SF}|\sin(\theta) = 0$, in which $\vec{k}_{SF}$ is the wavevector of the produced SF wave. (b) The parallel wavevectors of the SF wave in a straight waveguide turn into twisted wavevectors in a microring geometry, leading to the out-of-plane radiation carrying OAM of light. The TC and frequency of the emitted vortex are $l_{SF} = m - n$ and $\omega_{SF} = \omega_m + \omega_n$, in which m (ω_m) and n (ω_n) are the mode numbers (frequencies) of the microring. (c) Schematic of engineering SF optical vortices in multiple degree-of-freedom via on-chip NCR, including controllable spatial profile and polarization state, independently tunable wavelength and TC, and the generation of visible vortex microcombs with controlled temporal profiles by a synergistic $\chi^{(2)}$ and $\chi^{(3)}$ nonlinear optical process.

indicates the CW mode, distinguishing it from the CCW one. Thus, the fundamental waves producing NCR can be expressed as $E_{P,m} = u_{P,m}(\rho, z) e^{i(\omega_m t + m\varphi)} \hat{e}_P$ and $E_{P,-n} = u_{P,-n}(\rho, z) e^{i(\omega_n t - n\varphi)} \hat{e}_P$. Here, $u_{P,m}(\rho, z)$ and $u_{P,-n}(\rho, z)$ are the electric field distributions in the cross-section of the microring, and $\hat{e}_P$ is a unit vector of the dominant electric field for TE or TM modes. Therefore, the evolution of the SF field can be written as a function of the two fundamental fields and the second-order optical nonlinear coefficient under the classical slow varying envelope approximation [28]:

$$\begin{aligned} \vec{k}_{SF} \cdot \nabla \vec{E}_{SF} = & -i \frac{(\omega_m + \omega_n)^2}{c^2} \overleftrightarrow{\chi^{(2)}} : \hat{e}_P \hat{e}_P u_{P,m}(\rho, z) \\ & \times u_{P,-n}(\rho, z) e^{i(m-n)\varphi} e^{-i\Delta\vec{k} \cdot \vec{r}} e^{i(\omega_m + \omega_n)t} \end{aligned} \quad (1)$$

where $\Delta\vec{k}$ is the wavevector mismatch in the NCR process. Due to the in-plane confinement of the fundamental waves and the satisfaction of the NCR condition, the wavevector mismatch is mainly determined by the out-of-plane radiated SF wave, given by $\Delta\vec{k} = -k_{SF,\rho}\hat{\rho} - k_{SF,z}\hat{z}$, where $k_{SF,\rho}$ and $k_{SF,z}$ are wavevector components along the radial direction $\hat{\rho}$ and z -axis direction $\hat{z}$, respec-

tively. $\overleftrightarrow{\chi^{(2)}} : \hat{e}_P \hat{e}_P$ denotes a double contraction of the nonlinear tensor under the interaction of two unit vectors, leading to the vector effective nonlinear coefficient $\vec{\chi}_{eff} = [\chi_{eff,x} \ \chi_{eff,y} \ \chi_{eff,z}]^T$. Assuming that $u_{P,m}(\rho, z)$ and $u_{P,-n}(\rho, z)$ are an approximately constant within the microring, and the thickness Δd and the width ΔR are sufficiently small for the microring with a radius of R , the far field of the SF wave normal to the z -axis can be written as

$$\begin{aligned} \vec{E}_{SF-far} = & -i \frac{(\omega_m + \omega_n)^2}{k_{SF,z} c^2} u_{P,m} u_{P,-n} R \Delta d \Delta R \\ & \times \int_0^{2\pi} \vec{\chi}_{eff} e^{i(m-n)\varphi} e^{ik_{SF,\rho} R \cos(\phi-\varphi)} d\varphi \end{aligned} \quad (2)$$

where ϕ is the azimuthal angle in the wavevector space (see Methods for calculation of the radiated SF fields). Each component in $\vec{\chi}_{eff}$, labeled as $\chi_{eff,j}$ ($j = x, y, z$), possibly depends on the azimuthal angle φ , so that it can be expressed as Fourier series, i.e., $\chi_{eff,j} = \sum_{l_\chi} C_{l_\chi} e^{il_\chi \varphi}$, where the integer l_χ refers to additional TC induced by the spatially varying nonlinear polarization and C_{l_χ} is the corresponding Fourier coefficient. Therefore, the total TC of the radiated SF wave is determined by the sum of

modal-difference-induced TC l_{WGM} and χ_{eff} -induced TC l_{χ} , resulting in the general NCR condition:

$$l_{\text{SF}} = l_{\text{WGM}} + l_{\chi} = m - n + l_{\chi}. \quad (3)$$

In general, the NCR process in microring can produce structured optical vortices with a wide range of spatial profiles and polarization states by combining the two pump beams in different polarizations and mode numbers. As a proof of concept, we only show the interactions between fundamental TE and TE modes or between fundamental TM and TM modes in our study. Nonlinear microrings made of both x -cut and z -cut TFLN are employed to highlight the universality of the NCR process. Therefore, four representative SF processes assisted by NCR are presented: case 1 for interaction between $E_{\text{TM},m}$ and $E_{\text{TM},-n}$ modes in x -cut TFLN, case 2 for interaction between $E_{\text{TE},m}$ and $E_{\text{TE},-n}$ modes in x -cut TFLN, case 3 for interaction between $E_{\text{TE},m}$ and $E_{\text{TE},-n}$ modes in z -cut TFLN, and case 4 for interaction between $E_{\text{TM},m}$ and $E_{\text{TM},-n}$ modes in z -cut TFLN. The dominant effective nonlinear coefficients are rather distinct in these four NCR cases, with some of them linked to the azimuthal angle, leading to SF optical vortices with different spatial profiles and polarization states as summarized in Table 1 (see Extended Data Fig. E1 and formulas for polarization-dependent χ_{eff} and SF radiation in Methods). It should be noticed that the physical coordinate system of z -cut TFLN is aligned with the lab coordinate system, while that of x -cut TFLN has a CCW rotation of 90° around the y -axis relative to the lab coordinate system. The dominant χ_{31} and $\chi_{33} \cos^2 \varphi$ in cases 1 and 2 resulting in the main x -polarization state in the $x - y$ plane. Moreover, the superposition of nonlinear polarization in case 3 produces a cylindrical polarization, while the diffraction of the subwavelength microring in case 4 leads to a radial polarization (Extended Data Fig. E2). According to Eqs. (1) to (3), these distinct nonlinear polarizations and radiation together with the NCR condition allow us to engineer the optical vortices in multi-dimensions including the spatial profile, polarization state, emission wavelength, and TC by carefully choosing the resonant modes and effective nonlinear coefficients involved in the on-chip optical frequency mixing process, as listed by the last column using ket notation displayed in Table 1. We fabricate a variety of devices and customize the characterization system to demonstrate the proposed functionalities, as described in Methods of TFLN microring fabrication and experimental characterization of nonlinear optical vortices, along with Extended Data Figs. E3-E7 and Table E2.

ENGINEERING THE SPATIAL PROFILE AND POLARIZATION STATE OF NONLINEAR OPTICAL VORTICES

In the experiment, we firstly demonstrate the controllability of spatial profile and polarization state for on-chip nonlinear radiation by setting $\lambda_n = \lambda_m$, leading to $l_{\text{WGM}} = 0$, in order to avoid complications induced by modal difference. In such a scenario, the on-chip SF process degenerates into the second harmonic generation (SHG). Although previous studies have explored on-chip out-of-plane SHG [29, 30], the underlying principle of NCR remains unrevealed, thereby restricting further investigation into SFG with structured wavefronts. Figure 2a shows the spectra of the fundamental and second-harmonic (SH) waves for the four cases. The degenerated wavelengths of the fundamental waves are 1562.04 nm, 1561.30 nm, 1563.37 nm, and 1563.91 nm, leading to the exactly frequency-doubled emission at 781.02 nm, 780.65 nm, 781.69 nm, and 781.96 nm, respectively. In case 1, a homogeneous SH intensity distribution on the ring is observed owing to the $\chi_{\text{eff},x}$ and $\chi_{\text{eff},y}$ being independent on the azimuthal angle (Fig. 2b). The Fourier transformation of this distribution is a zero-order Bessel function, which coincides with the recorded far-field intensity pattern and the simulated one (Fig. 2c). Since χ_{31} is generally two times larger than χ_{22} , the x -polarization component of the SH wave dominates in case 1, resulting in an NCR state of $|V, 0\rangle$ (Fig. 2d). In case 2, although the non-zero $\chi_{\text{eff},x}$ and $\chi_{\text{eff},y}$ contribute to the radiated SH waves, the near-field intensity distribution presents as two circular arcs due to the dominant $\chi_{33} \cos^2 \varphi$ component within $\chi_{\text{eff},x}$ (Fig. 2e). This is attributed to the largest χ_{33} of LN crystals that is an order of magnitude larger than the other coefficients. Because $\cos^2 \varphi$ can be rewritten as $\frac{1}{2} + \frac{1}{4}e^{i2\varphi} + \frac{1}{4}e^{-i2\varphi}$, such a circular amplitude modulation produced non-circular symmetric far-field patterns with OAM sidebands encoded by angular diffraction (Fig. 2f,g) [31]. These two sidebands correspond to two OAM states with $l_{\chi} = \pm 2$, leading to a superposition state of $\frac{1}{2}|V, 0\rangle + \frac{1}{4}|V, 2\rangle + \frac{1}{4}|V, -2\rangle$. In case 3, the two orthogonal polarizations in the $x - y$ plane form a cylindrical vector polarization state expressed by a Jones matrix of $[\sin 2\varphi \ \cos 2\varphi]^T$, which rotates around the z -axis and repeats twice in one round trip. Therefore, a homogeneous SH intensity distribution in the near field is observed (Fig. 2h), but the far-field intensity pattern presents as a donut shape owing to the polarization singularity at the center (Fig. 2i) [32]. The cylindrical polarization is revealed by the intensity patterns of x -polarization and y -polarization components (Fig. 2j), exhibiting a rotational angle of 45° in between. Generally, the non-zero $\chi_{\text{eff},z}$ in case 4 cannot produce an out-of-plane SH wave. However, the microring with a subwavelength width acts as a circularly symmetric diffraction

Table 1. Summary of effective nonlinear coefficients and the corresponding intensity distributions for the four different nonlinear interaction processes. The dominating effective nonlinear coefficients in each process are highlighted in red. The ket notation is used to express generated states, where the first and second parameter refer to a polarization state and a total TC, respectively. V, vertical (x) polarization; H, horizontal (y) polarization; R, right-handed circular polarization; L, left-handed circular polarization.

NCR processes	Effective nonlinear coefficients in the lab coordinate system $\chi_{\text{eff}} = \overleftrightarrow{\chi}^{(2)}: \hat{e}_p \hat{e}_p$			Dominated angle-dependent polarization state in the x - y using Jones matrix	Far-field SF intensity distribution in the x - y plane
	$\chi_{\text{eff},x}$	$\chi_{\text{eff},y}$	$\chi_{\text{eff},z}$		
case 1 x-cut TM+TM	$-\chi_{31}$	$-\chi_{22}$	0	$\begin{bmatrix} 1 \\ 0 \end{bmatrix}$	$ V, m-n\rangle$
case 2 x-cut TE+TE	$-\chi_{31}\sin^2\varphi$ $-\chi_{33}\cos^2\varphi$	$\chi_{22}\sin^2\varphi$ $-\chi_{15}\sin 2\varphi$	0	$\cos^2\varphi \begin{bmatrix} 1 \\ 0 \end{bmatrix}$ $= \left(\frac{1}{2} + \frac{1}{4}e^{i2\varphi} + \frac{1}{4}e^{-i2\varphi}\right) \begin{bmatrix} 1 \\ 0 \end{bmatrix}$	$\frac{1}{2} V, m-n\rangle$ $+\frac{1}{4} V, m-n+2\rangle$ $+\frac{1}{4} V, m-n-2\rangle$
case 3 z-cut TE+TE	$\chi_{22}\sin 2\varphi$	$\chi_{22}\cos 2\varphi$	0	$\begin{bmatrix} \sin 2\varphi \\ \cos 2\varphi \end{bmatrix}$ $= \frac{-i}{2} \left(e^{i2\varphi} \begin{bmatrix} 1 \\ i \end{bmatrix} - e^{-i2\varphi} \begin{bmatrix} 1 \\ -i \end{bmatrix} \right)$	$\frac{1}{2} L, m-n+2\rangle$ $-\frac{1}{2} R, m-n-2\rangle$
case 4 z-cut TM+TM	0	0	χ_{33}	$\begin{bmatrix} \cos \varphi \\ \sin \varphi \end{bmatrix}$ $= \frac{1}{2} \left(e^{-i\varphi} \begin{bmatrix} 1 \\ i \end{bmatrix} + e^{i\varphi} \begin{bmatrix} 1 \\ -i \end{bmatrix} \right)$	$\frac{1}{2} L, m-n-1\rangle$ $+\frac{1}{2} R, m-n+1\rangle$

structure, scattering the SH field produced by χ_{33} in the $x-y$ plane to the far field (Extended Data Fig. E2). This diffracted feature resembles what occurs in a microring addressed with angular gratings, which results in a radial polarization of radiated SH wave, given by $[\cos \varphi \ \sin \varphi]^T$, as shown by the homogeneous SH intensity distribution on the ring (Fig. 2k) and the far-field cylindrical vector SH beam (Fig. 2l,m). Both cylindrical vector beams in cases 3 and 4 can be decoupled into two scalar OAM states with left-handed and right-handed circular polarizations, i.e., $|L, l_\chi = 2\rangle$ and $|R, l_\chi = -2\rangle$ for case 3 and $|L, l_\chi = -1\rangle$ and $|R, l_\chi = 1\rangle$ for case 4, based on spin-to-orbital angular momentum coupling [33]. The simulated far-field intensity pattern in Fig. 2l, calculated using the theoretical result of Eq. (2) with the $[\cos \varphi \ \sin \varphi]^T$ component incorporated into $\chi_{\text{eff},z}$, are in very good agreement with the measured ones.

GENERATING THE OPTICAL VORTICES WITH ON-DEMAND WAVELENGTH AND TC VIA NCR

In addition to the flexible engineering of the spatial profile and polarization state, we can on-demand load helical phase fronts to the NCR by employing two non-degenerated fundamental waves, i.e. $l_{\text{WGM}} \neq 0$, leading to the generation of optical vortices with an on-demand wavelength and TC. Based on Eq. (3), the radiated SF will present different TCs of l_{SF} when the CCW and CW WGMs are at different orders. As a demonstration, $l_{\text{WGM}} = m-n = \pm 4, \pm 2, \pm 1$ are set in cases 1 to 4, respectively. As shown in Fig. 3a, the fundamental wavelengths for the four cases are fixed at $\lambda_m = 1556.55$ nm, 1558.43 nm, 1557.91 nm, and 1561.30 nm for CCW modes and $\lambda_n = 1562.02$ nm, 1561.30 nm, 1563.37 nm, and 1563.88 nm for CW modes to get a pos-

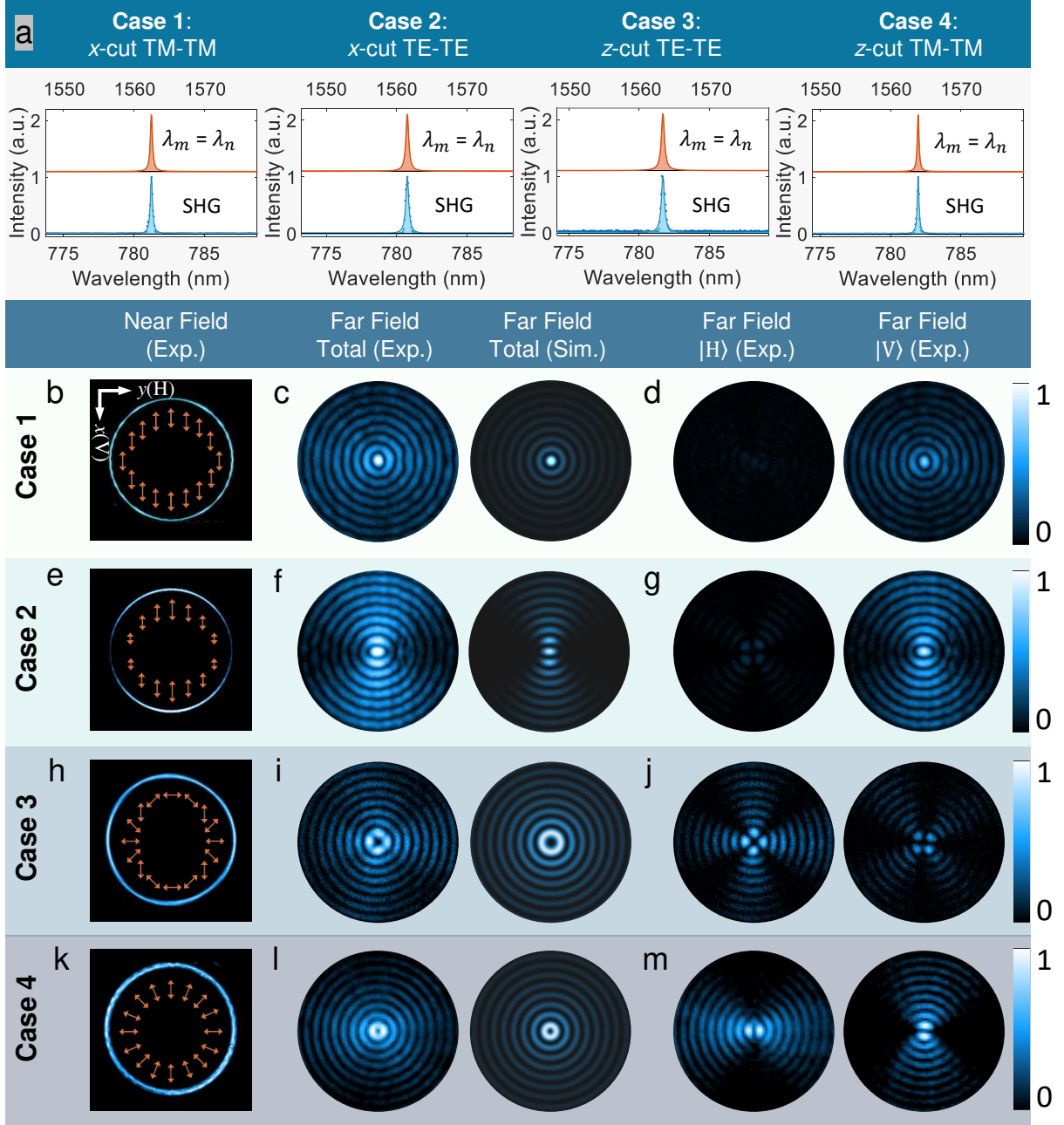


FIG. 2. **Engineering the spatial profile and polarization state of nonlinear optical vortices in frequency doubling processes.** (a) Spectra of the fundamental waves and the SHG ($l_{\text{WGM}}=0$) in four different cases. (b-d) Measured and simulated near- and far-field patterns of the SHG in case 1. Due to the absence of intensity modulation and spin-orbital angular momentum coupling, the emitted beam doesn't carry any OAM and therefore exhibits a solid center in the far-field pattern. The x -polarization component of the SH wave dominates due to the fact that χ_{31} is generally two times larger than χ_{22} . (e-g) Measured and simulated near- and far-field patterns of the SHG in case 2. The near-field amplitude modulation with two circular arcs leads to the SHG in a superposition state $\frac{1}{2}|V, 0\rangle + \frac{1}{4}|V, 2\rangle + \frac{1}{4}|V, -2\rangle$, forming a solid center and two lobes in the far-field pattern as demonstrated in both the experiment and the simulation. The dominated x -polarization component in the far-field pattern can be attributed to the largest χ_{33} of LN crystals that is an order of magnitude larger than the other coefficients. (h-j) Measured and simulated near- and far-field patterns of the SHG in case 3. The far-field pattern of the cylindrically polarized SHG shows a hollow center, corresponding to a polarization singularity. There is a 45° rotation between the H-polarized/V-polarized far-field patterns because the polarization state rotates twice in 360° . (k-m) Measured and simulated near- and far-field patterns of the SHG in case 4. The far-field pattern of the cylindrically polarized SHG shows a hollow center, corresponding to a polarization singularity. H-polarized/V-polarized far-field patterns are shifted by 90° due to fact that the polarization state rotates only once in 360° .

itive l_{WGM} , while swap them to obtain a negative l_{WGM} . Clearly, strong SF wave assisted by NCR is located at 779.64 nm, 779.93 nm, 780.31 nm, and 781.29 nm, respectively. These results demonstrate that the prominent out-of-plane radiation comes from nonlinear interactions of the counter-propagating two fundamental waves. The near-field profiles of the SF waves inserted in Fig. 3a are consistent with those in Fig. 2b,e,h,k. To reveal the OAM states, their far-field intensity patterns are characterized as shown in Fig. 3b-e. In case 1, the scalar SF far fields with $l_{\text{WGM}} = \pm 4$ show a typical OAM feature of hollow beams which is in contrast to SHG without OAM as presented in Fig. 2c. According to the theoretical framework in Table 1, these two SF vortices carry total on-demand OAM states of $|V, 4\rangle$ and $|V, -4\rangle$, respectively. To verify the TC of the generated OAM, we interfere these two optical vortices (see Methods and Extended Data Fig. E7), and the number of the anti-nodes in the far-field pattern equals $2|l_{\text{WGM}}| = 8$, confirming the theoretical prediction of $l_{\text{SF}} = \pm 4$ (Fig. 3b) [34, 35]. Figure 3c shows the capability of simultaneously modulating the intensity profile and controlling the TC of the emitted optical vortices in case 2 by setting $l_{\text{WGM}} = \pm 2$. The recorded far-field intensity pattern relates to a superposition of multiple OAM states, i.e., $\frac{1}{2}|V, 2\rangle + \frac{1}{4}|V, 4\rangle + \frac{1}{4}|V, 0\rangle$ for $l_{\text{WGM}} = 2$ and $\frac{1}{2}|V, -2\rangle + \frac{1}{4}|V, 0\rangle + \frac{1}{4}|V, -4\rangle$ for $l_{\text{WGM}} = -2$, respectively. The bright centers at the far field reveal the state of $|V, 0\rangle$ within those two superposition states resulted from the counteraction of the modal-difference-induced TC and the χ_{eff} -induced TC, i.e., $l_{\text{SF}} = l_{\text{WGM}} \pm 2 = 0$. It also provides evidence that the OAM sidebands in Fig. 2f carry TC of $l_{\chi} = \pm 2$. Their interfering pattern in Fig. 3c features non-uniform four nodes.

Microrings on z -cut TFLN are used to load additional OAM onto the cylindrical vector beams in cases 3 or 4, which exhibit couplings between spin angular momentum (SAM) and OAM [14, 36]. In case 3, due to the SAM and OAM coupling, the produced OAM states will become into $\frac{1}{2}|L, m - n + 2\rangle - \frac{1}{2}|R, m - n - 2\rangle$ according to Table 1. Setting $l_{\text{WGM}} = 2$ (or -2) in the experiment, the generated SF vortices turn into a superposition of $|L, 0\rangle$ and $|R, -4\rangle$ (or $|L, 4\rangle$ and $|R, 0\rangle$), confirmed by far-field SF intensity patterns in Fig. 3d. The bright spot in the center of the intensity distribution for the left-handed (or right-handed) circularly polarized beam reveals the total OAM of $l_{\text{WGM}} + l_{\chi} = 0$. A similar effect can be observed in case 4. According to Table 1, vortex states with $\frac{1}{2}|L, -2\rangle + \frac{1}{2}|R, 0\rangle$ and $\frac{1}{2}|L, 0\rangle + \frac{1}{2}|R, 2\rangle$ are obtained by setting $l_{\text{WGM}} = -1$ and $l_{\text{WGM}} = 1$, respectively, as shown in Fig. 3e. Those radiated SF intensity patterns in cases 1 to 4 are in excellent agreement with the numerical simulations.

INDEPENDENT AND WIDE-RANGE TUNING OF THE WAVELENGTH AND TC ASSOCIATED WITH OPTICAL VORTICES

We now present the ability to independently tune the wavelength and TC of the nonlinear optical vortices with unprecedented ranges, which significantly outperforms the state-of-the-art [18, 19]. In case 1 with $l_{\chi} = 0$, the correlation between wavelength and TC could be decoupled by simultaneously changing the orders of both fundamental waves with several free spectrum ranges (FSRs), labeled as Δl , along the same direction, i.e. m to $m + \Delta l$ and n to $n + \Delta l$, or opposite direction, i.e. $m + \Delta l$ and $n - \Delta l$. The former could produce optical vortices with a pre-designed TC, i.e., $|V, m - n\rangle$, but varied emission wavelengths (Fig. 4a), while the latter can tune the TC of OAM beams, i.e., $|V, m - n + 2\Delta l\rangle$, with a targeted emission wavelength (Fig. 4b). We set $\lambda_m = 1562.02$ nm and $\lambda_n = 1556.55$ nm to produce OAM with $l_{\text{SF}} = 4$ at first, and then tune the pump wavelengths to shorter and longer values, achieving the lowest $\Delta l = -17$ and highest $\Delta l = 20$, respectively. This tuning process covers a tuning bandwidth of about 51.8 nm, corresponding to 37 times of the FSR (1.4 nm for each FSR). As a result, the unchanged far-field SF intensity distribution indicates the invariable TC while the emission wavelength spans from 767.77 nm to 793.62 nm, as shown in Fig. 4c. To demonstrate the independently tunable TC, the fundamental wavelengths are firstly fixed at $\lambda_m = \lambda_n = 1562.02$ nm, and then they are tuned to obtain $\Delta l = -5$ to 5, respectively. In this configuration, the TC changes with a step of $2\Delta l$ at the same SF wavelength of 781.01 nm, as the blue spectra and patterns shown in Fig. 4d. Setting $\lambda_m = 1560.65$ nm and $\lambda_n = 1562.03$ nm to get $m - n = 1$ with an SF wavelength of 780.66 nm, the TC follows the law of $2\Delta l + 1$ using the same controlling scheme. It should be noticed that there is a slight difference between the SF wavelengths of odd and even OAMs. We use temperature to tune the resonant wavelength of the microring by half FSR when producing the optical vortices with odd ($l_{\text{SF}} = 2\Delta l + 1$) TCs (Extended Data Fig. E8) so that the SF wavelength keeps the same as that with even ($l_{\text{SF}} = 2\Delta l$) TCs (orange spectra and patterns in Fig. 4d).

VISIBLE MULTI-STATE VORTEX MICROCOMBS WITH ENGINEERABLE WAVE PACKETS

Finally, we employ the NCR to generate multi-state vortex microcombs at visible range in a synergistic $\chi^{(2)}$ and $\chi^{(3)}$ process by using a z -cut TFLN microring (see Methods and Extended Data Fig. E5). Integrated vortex microcombs at telecom bands have been very re-

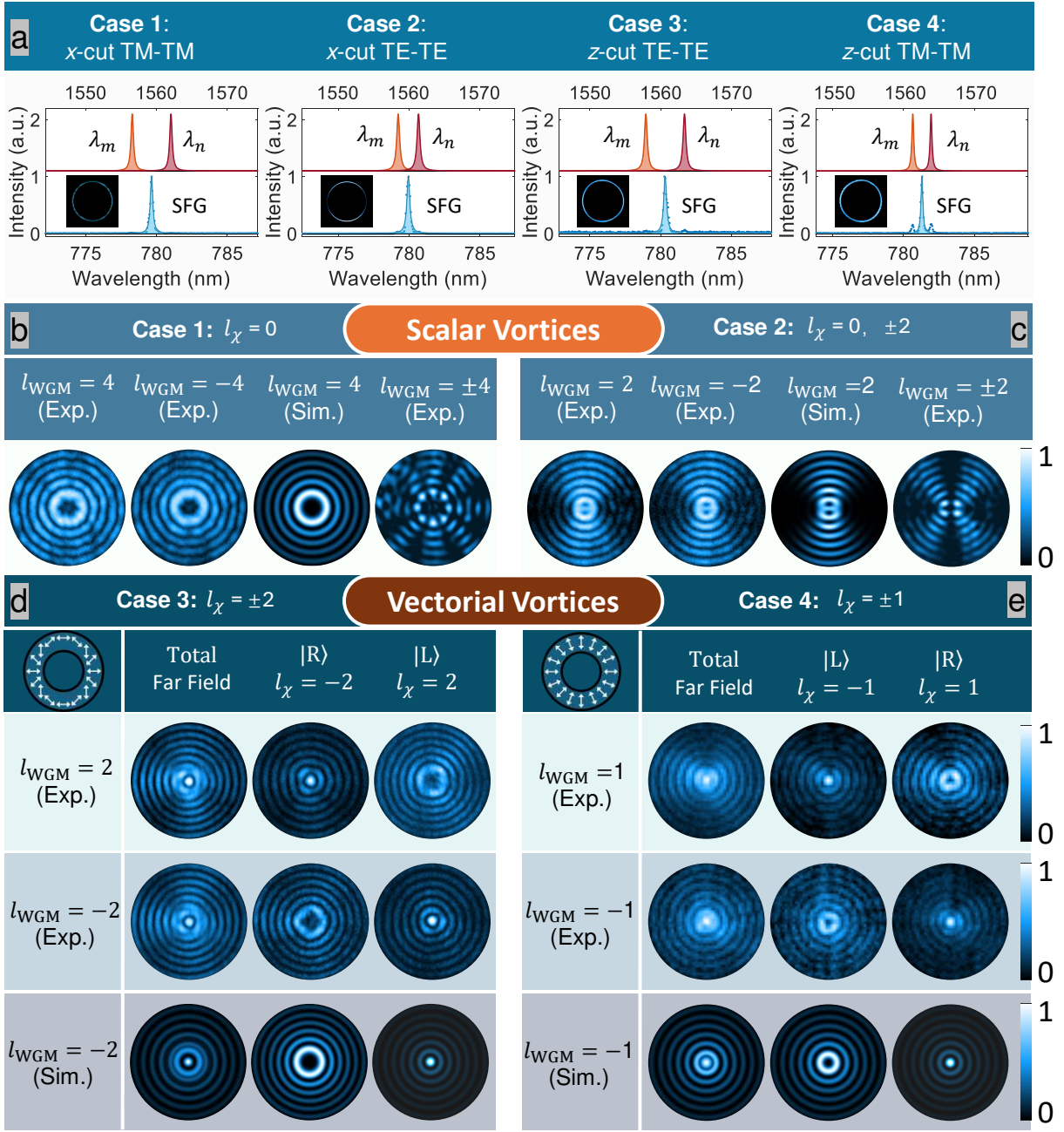


FIG. 3. **Encoding OAM with on-demand wavelength and TC to NCR.** (a) Spectra of the fundamental wave and SF and near-field SF intensity distributions for cases 1 to 4, respectively. (b) Measured and simulated far-field patterns for $l_{WGM} = \pm 4$ in case 1 with $l_\chi = 0$. The TC of the SF $l_{SF} = \pm 4$ results in an interference pattern with an anti-node number of $2|l_{WGM}| = 8$. (c) Measured and simulated far-field patterns for $l_{WGM} = \pm 2$ in case 2 with $l_\chi = 0, \pm 2$. Due to the amplitude modulation, the generated vortex states are $\frac{1}{2}|V, 2\rangle + \frac{1}{4}|V, 4\rangle + \frac{1}{4}|V, 0\rangle$ for $l_{WGM} = 2$ and $\frac{1}{2}|V, -2\rangle + \frac{1}{4}|V, 0\rangle + \frac{1}{4}|V, -4\rangle$ for $l_{WGM} = -2$, respectively. Therefore, both far-field patterns for $l_{WGM} = \pm 2$ exhibit a solid center and their interference pattern features 4 anti-nodes. (d) Measured and simulated far-field patterns for $l_{WGM} = \pm 2$ in case 3 with $l_\chi = \pm 2$. Due to the spin-orbital coupling with polarization rotating twice in one round trip, the generated vortex state is $\frac{1}{2}|L, m - n + 2\rangle - \frac{1}{2}|R, m - n - 2\rangle$. Thus, the right-handed polarization component for the cylindrically polarized optical vortex with $l_{WGM} = 2$ exhibits $l_{SF} = 0$, leading to a solid center in the far-field pattern. Similarly, the far-field pattern of the left-handed polarization component for the cylindrically polarized optical vortex with $l_{WGM} = -2$ also experiences a solid center due to $l_{SF} = 0$. (e) Measured and simulated far-field patterns for $l_{WGM} = \pm 1$ in case 4. Due to the spin-orbital coupling with polarization rotating only once in one round trip, the generated vortex state is $\frac{1}{2}|L, m - n - 1\rangle + \frac{1}{2}|R, m - n + 1\rangle$. Thus, the left-handed polarization component for the cylindrically polarized optical vortex with $l_{WGM} = 1$ exhibits TC of 0, leading to a solid center in the far-field pattern. Similarly, the far-field pattern of the right-handed polarization component for the cylindrically polarized optical vortex with $l_{WGM} = -1$ also experiences a solid center.

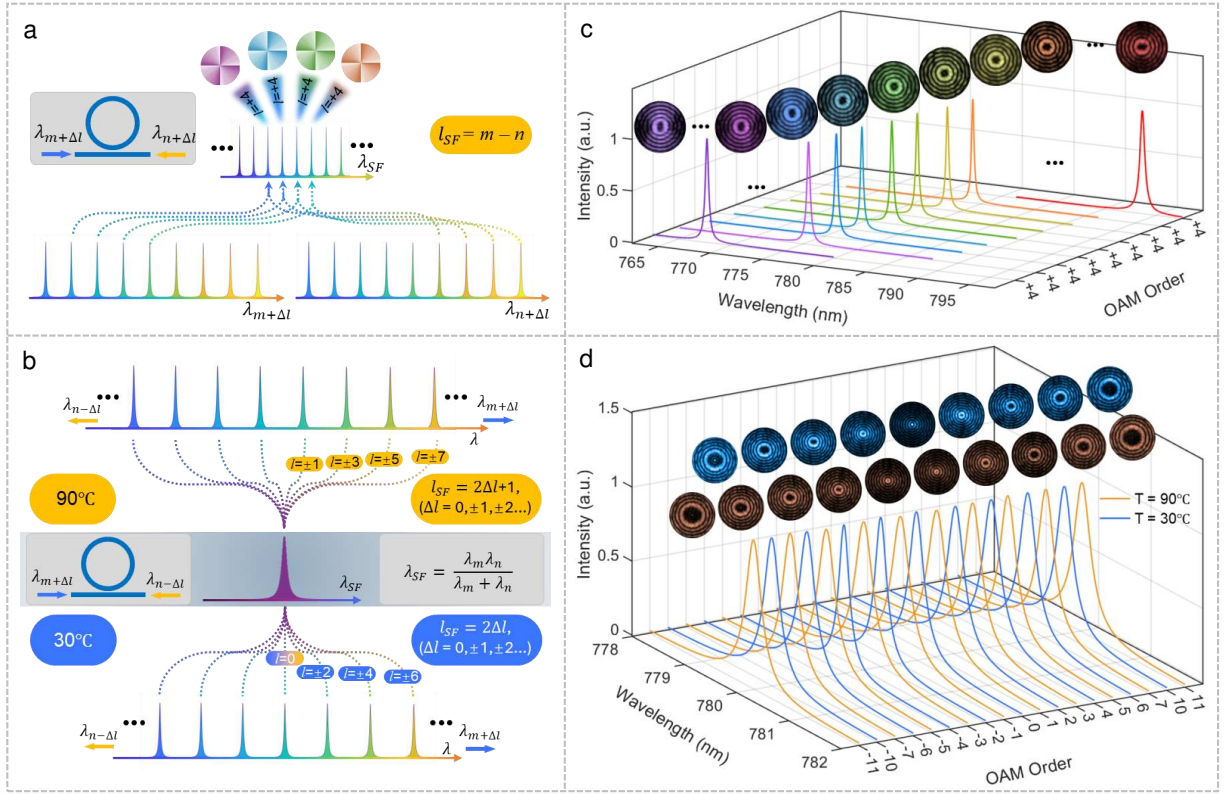


FIG. 4. **Independent and wide-range tuning of the wavelength and TC of the nonlinear optical vortices.** (a) The scheme for achieving optical vortices with tunable wavelengths but a fixed TC of $l_{SF} = m - n$ by simultaneously changing the orders of two fundamental waves along the same direction. (b) The scheme for achieving SF vortices with tunable TCs following the laws of $l_{SF} = 2\Delta l$ and $l_{SF} = 2\Delta l + 1$ but at a fixed wavelength by simultaneously changing the orders of two fundamental waves along the opposite direction. Temperature control on the microring is employed to fine-tune the resonant wavelength by half of the FSR, compensating for the wavelength difference between SF vortices with odd and even TCs. (c) Experimental demonstrations of optical vortices with tunable wavelengths without changing the TC by keeping $m - n = 4$ in case 1 based on the scheme in (a). (d) Experimental demonstrations of optical vortices with tunable TCs without changing the wavelength in case 1 based on the scheme in (b). The TC ranges from -11 to 11 continuously and the wavelength difference between the odd and even TCs is compensated by temperature control.

cently realized in angular-grating-addressed microrings, exhibiting technological advantages in the applications of OAM spectroscopy and spatiotemporal light generation [37, 38]. However, the angular gratings employed in previous work impose an inevitable compromise between the Q-factors of microring and the vortex emission efficiencies. In addition, it has been extremely challenging to directly transplant such a strategy from the telecom band to the visible range due to the very strong material dispersion and higher Raman gain at the visible wavelength. Although the frequency doubling technique can translate telecom microcombs to the visible range but the operation bandwidth is limited by the phase matching condition. The NCR employed in our scheme simultaneously addresses these three issues and therefore successfully enables the first demonstration of the visible vortex microcombs. Figure 5a shows the principle for the generation of visible vortex microcombs. A TE fundamental

pump at $\lambda_m = 1554.45$ nm is injected from the left port into the waveguide with an on-chip optical power of 326 mW and excite the CCW optical soliton [39–42]. The same TE fundamental beam is injected from the opposite direction of the coupling waveguide (from the right side) and experiences SF processes with each of the CCW modes associated with the comb lines, leading to the formation of SF vortex microcombs emitting toward free space (see Extended Data Fig. E9 for the experimental setup). Figure 5b shows the output comb spectrum from the right port of the coupling waveguide, in which the smooth envelope of the produced comb lines indicates the generation of a Kerr soliton along with two dispersive waves with a repetition rate of 250 GHz (2 nm in wavelength). The SF spectrum of emitted vortex combs with the same repetition rate of 250 GHz (0.5 nm in wavelength) is collected by a free space setup, as presented on the top of Fig. 5b. The soliton feature together with two

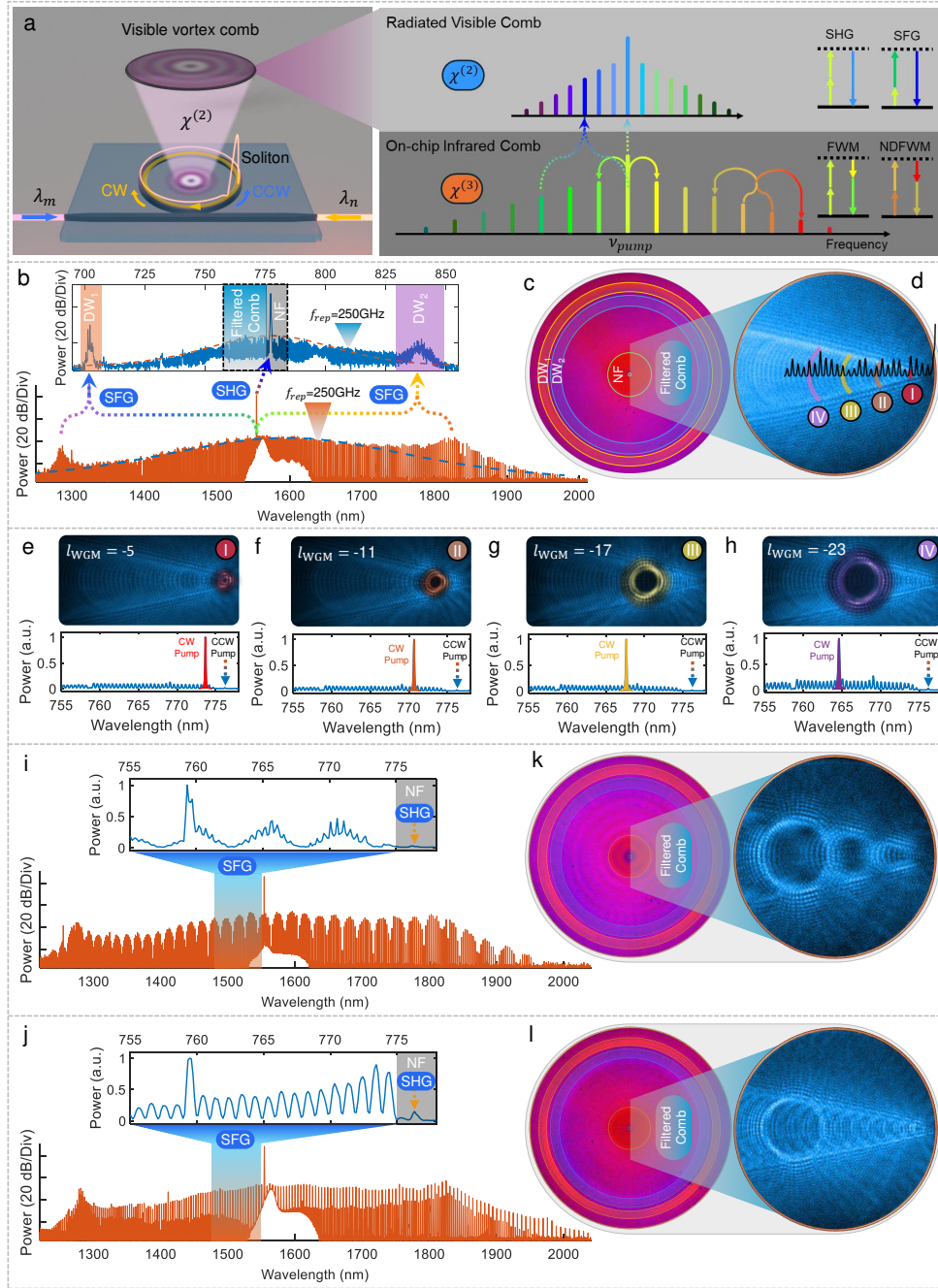


FIG. 5. Visible multi-state vortex microcombs with engineerable temporal wave packet. (a) A schematic illustrating the principles of $\chi^{(3)}$ and $\chi^{(2)}$ nonlinear interactions that produce telecom-band Kerr soliton combs and visible vortex soliton combs. The infrared combs are generated by the CCW pump via four-wave mixing processes while the visible vortex combs are produced by the NCR process between the CCW soliton and the CW pump. (b) Spectra of a Kerr soliton comb with the repetition rate of 250 GHz (2 nm in wavelength) in the telecom band and spectra of the corresponding vortex soliton comb with the same repetition rate of 250 GHz (0.5 nm in wavelength) at visible. A notch filter (gray area denoted as NF) is used to suppress the SH spectrum from the CCW and CW pumps and a bandpass filter (blue area denoted Filtered Comb) is employed to spectrally select the optical vortices of interest (with low TCs). The dispersive waves at short (orange area) and long (purple area) wavelengths are marked by DW₁ and DW₂, respectively. (c) Vertically radiated intensity profiles of the visible vortex combs. The far-field patterns associated with the vortices with different OAMs selected by the bandpass filter and dispersive waves correspond to the green, blue and orange rings, respectively. (d) The filtered vortex comb radiation is dispersed by a blazed grating, resulting in different diffraction angles for the comb lines at different wavelengths. The zoomed-in comb lines at different wavelengths in the spectrum overlay the corresponding diffractive rings (I, II, III, IV) at different angles. (e-h) Amplified single vortex comb lines and corresponding vortex far-field patterns (I, II, III, IV) by tuning an extra laser to the targeted comb line wavelengths. (i) and (j) The spectra of the dual-soliton state and soliton-crystal state generated in the telecom band. Above them are the zoomed-in spectra of the up-converted dual-soliton combs and soliton-crystal combs in the visible range. (k) and (l) are the diffracted far-field patterns associated with the visible dual-soliton combs and soliton-crystal combs. The clear intensity modulations respective to (d) reveal the characteristics of a dual-soliton state with a modulated envelope and a soliton-crystal state with doubled mode spacing.

dispersive waves are faithfully converted to the vortex combs in the visible range. To reveal OAM information encoded in each visible comb line, a notch filter (NF) is used to suppress the up-converted pump (grey area) while a bandpass (BP) filter is used to spectrally select the comb lines with small TCs (blue area) as marked in Fig. 5b. The far-field intensity distribution of vortex combs corresponding to the visible spectrum in Fig. 5b is presented in Fig. 5c, in which the dispersive waves and filtered comb lines are marked by circles with varied diameters due to their significant differences in the TC of the OAM states. The filtered vortex comb emission, denoted by the filtered comb region in Fig. 5c is further dispersed by a blazed grating in order to spatially separate the different comb lines, as shown in Fig. 5d, overlaying with the spectrum of the filtered combs. The diameter of each ring increases from right to left, indicating a larger TC for the shorter wavelength comb line, as exemplified by I to IV. Such a wavelength and TC correlation is further verified by introducing an additional weak fundamental beam with a power of 5 mW that mimics the single comb line in the telecom band. Its wavelength can be finely tuned to align with any of the Kerr comb lines (e.g., I to IV), thereby amplifying the corresponding SF spectra and the OAM states. As shown in Fig. 5e-h, the additional fundamental beam is set to match higher resonant modes with 5 (I), 11 (II), 17 (III), and 23 (IV) FSRs from the $\lambda_m = 1554.45$ nm, resulting in the enhanced SF signals with $l_{\text{WGM}} = -5, -11, -17, \text{ and } -23$ at different SF wavelengths in both spectra and the corresponding far-field patterns. In addition, we also demonstrate a dual-soliton state featuring a modulated envelope (Fig. 5i) and a soliton crystal state (Fig. 5j) exhibiting a doubled FSR at telecom band via a $\chi^{(3)}$ process [43, 44], and faithfully transfer these distinct comb states to the visible ranges via SF processes, as shown on the top of Fig. 5i,j. As clearly demonstrated in Fig. 5k,l, the modulated envelope in the dual-soliton state and the doubled mode spacing in the soliton crystal state are revealed in the far-field intensity distribution and the mode space associated with the emitted vortices in the visible range. The stable access to visible vortex microcombs, which overcomes the challenges of strong material dispersion and higher Raman gain in the visible wavelength, may immediately find applications in biochemical sensing [45], optical clocks [46], astronomical spectroscopy [47] and beyond [48].

CONCLUSION

We've successfully demonstrated universal chip-space interfaces capable of generating and engineering structured optical vortices by manipulating WGMs in TFLN microring resonators. The nonlinear optical interactions in the nanophotonic chip-space interface enable the

multi-dimensional engineering of structured optical vortices in terms of spatial profile, polarization state, emission wavelength, TC and temporal wave packet. Independently wide-range tunings of the TC and emission wavelength are achieved—an advantage not available in microrings with embedded gratings. This capability enables the reconfiguration of both the wavelength and TC by adjusting the wavelength of the excitation lasers in the NCR process, offering significantly greater flexibility and reconfigurability compared to the state-of-the-art. The power of our method is fully released with the first realization of visible integrated vortex microcombs with engineerable temporal wave packets by taking advantage of NCR in a synergistic $\chi^{(2)}$ and $\chi^{(3)}$ nonlinear optical process. The advantages associated with the NCR presented in our work can be directly transplanted to other popular nonlinear optical platforms with $\chi^{(2)}$ nonlinearities, such as AlN, SiC, GaP and AlGaAs for integrated nonlinear photonics [49–54]. Moving forward, programmable nanophotonic chip-space interfaces with single injection waveguides can be pursued by incorporating additional integrated linear optical components on the same chip [55], offering advantages in compactness, reconfigurability and operation speed over the nonlinear metasurfaces [56, 57]. From a fundamental physics perspective, it would also be intriguing to investigate the reversal process of SF in which optical vortices are loaded from the top of the microring to generate correlated/entangled photon pairs propagating in the waveguides or the difference-frequency processes assisted by NCR in periodically-poling microrings for generating optical vortices at mid-infrared or terahertz. Our work bridges structure light and integrated optics, bringing an entirely unexplored paradigm for chip-scale nonlinear optics and integrated quantum photonics to enable structured light-matter interaction and high-dimensional optical information processing.

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METHODS

Theory on NCR in a microring resonator

In a dielectric microring resonator with $\mu = \mu_r \mu_0 = u_0$, the two fundamental whispering gallery modes (WGMs) are denoted as E_m and E_{-n} . The subscripts “ m ” and “ n ” are positive integers representing the azimuthal mode numbers of the WGMs, where the negative sign “ $-$ ” before “ n ” indicates the clockwise (CW) propagation direction, distinguishing it from the counterclockwise (CCW) direction. These modes in the cylindrical coordinate system (ρ, φ, z) can be expressed as

$$\begin{cases} E_m = u_m(\rho, z) e^{i(\omega_m t + m\phi)} \\ E_{-n} = u_{-n}(\rho, z) e^{i(\omega_n t - n\phi)} \end{cases} \quad (S1)$$

Here, $u_m(\rho, z)$ and $u_{-n}(\rho, z)$ represent the electric field distributions in the cross-section of the microring. The radiated sum-frequency (SF) wave, generated by second-order nonlinear polarization, can be seen as a linear combination of the WGMs of the unperturbed microring,

with angular amplitude modulation A_q , expressed as:

$$E_{\text{SF}} = \sum_q A_q(\varphi) u_q(\rho, z) e^{i[(\omega_m + \omega_n)t + q\varphi]} \quad (S2)$$

where q is an integer, the coefficients A_q depend on φ , and the summation includes all eigenmodes. These eigenmodes in Eq.(S2) encompass both guided modes and radiated modes, which satisfy the linear wave equation in the cylindrical coordinate system [58]:

$$\left[\frac{\partial^2}{\partial \rho^2} + \frac{1}{\rho} \frac{\partial}{\partial \rho} + \frac{\partial^2}{\partial z^2} + \frac{1}{\rho^2} \frac{\partial^2}{\partial \varphi^2} - \frac{\partial^2}{\partial t^2} \mu_0 \varepsilon_{\text{SF}}(\rho, z) \right] u_q(\rho, z) e^{iq\varphi} = 0 \quad (S3)$$

Using a non-depletion condition of the two fundamental beams, the total SF field in Eq.(S2) satisfies the nonlinear wave equation [59], given by

$$\left[\frac{\partial^2}{\partial \rho^2} + \frac{1}{\rho} \frac{\partial}{\partial \rho} + \frac{\partial^2}{\partial z^2} + \frac{1}{\rho^2} \frac{\partial^2}{\partial \varphi^2} - \frac{\partial^2}{\partial t^2} \mu_0 \varepsilon_{\text{SF}}(\rho, z) \right] E_{\text{SF}} = \mu_0 \frac{\partial^2}{\partial t^2} P^{(2)} \quad (S4)$$

where $P^{(2)}$ is the second-order nonlinear polarization induced by the interaction between E_m and E_{-n} , mathematically expressed as

$$\begin{aligned} P^{(2)} &= 2\varepsilon_0 \chi_{\text{eff}} E_m E_{-n} \\ &= 2\varepsilon_0 \chi_{\text{eff}} u_m(\rho, z) u_{-n}(\rho, z) e^{i[(\omega_m + \omega_n)t + (m-n)\varphi]} \end{aligned} \quad (S5)$$

where χ_{eff} refers to the effective second-order nonlinear coefficient. Substituting Eq.(S2) and (S5) into Eq.(S4) and using the slow varying amplitude approximation (i.e., $\frac{\partial^2 A_q(\varphi)}{\partial \varphi^2} \approx 0$) and the result of Eq.(S3), one obtains

$$\begin{aligned} \sum_q \frac{2iq}{\rho^2} \frac{dA_q}{d\varphi} u_q(\rho, z) e^{iq\varphi} &= -2\mu_0 \varepsilon_0 (\omega_m + \omega_n)^2 \\ &\times \chi_{\text{eff}} u_m(\rho, z) u_{-n}(\rho, z) e^{i(m-n)\varphi} \end{aligned} \quad (S6)$$

Taking the scalar product of Eq. (S6) with $u_s^*(\rho, z)$ (the complex conjugate of the s -order mode) and integrating over the $\rho-z$ plane, the result, using the orthogonality of the eigenmodes (i.e., $\langle s|q \rangle = \int u_s^*(\rho, z) u_q(\rho, z) d\rho dz = 0$ for $s \neq q$), is

$$\begin{aligned} \frac{dA_s}{d\varphi} &= -\frac{(\omega_m + \omega_n)^2}{isc^2 \langle s|s \rangle} \chi_{\text{eff}} e^{i(m-n-s)\varphi} \\ &\times \iint u_s^*(\rho, z) \rho^2 u_m(\rho, z) u_{-n}(\rho, z) d\rho dz \end{aligned} \quad (S7)$$

with $\langle s|s \rangle = \int u_s^*(\rho, z) u_s(\rho, z) d\rho dz$. It should be noted that χ_{eff} may be a function of φ due to the anisotropic nature of the $\chi^{(2)}$ tensor in thin film lithium niobate (TFLN). The integration on the right-hand side of Eq. (S7) is confined to the microring region where the $\chi^{(2)}$ process occurs. Without loss of generality, χ_{eff} can be expanded using a Fourier series: $\chi_{\text{eff}} = \sum_{l_x} C_{l_x} e^{il_x \varphi}$, where l_x is an integer and C_{l_x} is the Fourier coefficient determined by the χ_{eff} distribution around the ring. Eq. (S7) can then be rewritten as

$$\begin{aligned} \frac{dA_s}{d\varphi} &= -\frac{(\omega_m + \omega_n)^2}{isc^2 \langle s|s \rangle} \sum_{l_x} C_{l_x} e^{i(m-n-s+l_x)\varphi} \\ &\times \iint u_s^*(\rho, z) \rho^2 u_m(\rho, z) u_{-n}(\rho, z) d\rho dz \end{aligned} \quad (S8)$$

By performing the integration over the azimuthal angle in Eq. (S8), we obtain

$$\Delta A_s = -\frac{(\omega_m + \omega_n)^2}{isc^2 \langle s|s \rangle} \sum_{l_\chi} C_{l_\chi} \int_0^{2\pi} e^{i(m-n-s+l_\chi)\varphi} d\varphi \times \iint u_s^*(\rho, z) \rho^2 u_m(\rho, z) u_{-n}(\rho, z) d\rho dz \quad (S9)$$

According to Eq.(S9), a non-zero ΔA_s requires conservation of azimuthal mode numbers, i.e.,

$$s = m - n + l_\chi \quad (S10)$$

If χ_{eff} is independent of the azimuthal angle, Eq. (S10) can be simplified as $s = m - n = l_{\text{WGM}}$, corresponding to the momentum conservation law along the azimuthal direction, i.e., nonlinear Čerenkov radiation (NCR), given by $k_{\text{SF},\varphi} = k_m - k_n$, where k_m and k_n refer to moduli of the propagation constants of the two fundamental waves injected from the left and right sides, respectively, and $k_{\text{SF},\varphi}$ is the azimuthal wavevector of the produced SF wave. Therefore, Eq. (S10) is the general NCR law in a microring resonator.

In our configuration, the confined WGM has an azimuthal order of $2\pi R n_{\text{eff}}/\lambda$. With $\lambda = 775$ nm and $R = 100$ μm , this order is estimated to be approximately 10^3 . However, the SF waves generated by the interaction of the two fundamental beams at the C/L band have a much smaller azimuthal order governed by Eq. (S10), resulting in the production of a radiated SF wave. Due to the existence of the phase term $e^{i(m-n+l_\chi)\varphi}$, the radiated SF wave features an optical vortex, carrying orbital angular momentum (OAM) of light with topological charge (TC) of $l_{\text{SF}} = m - n + l_\chi = l_{\text{WGM}} + l_\chi$. Here, l_{WGM} and l_χ can be also called modal-difference-induced TC and χ_{eff} -induced TC.

Calculation of the radiated SF fields in NCR

In above discussion, the vector nature of the fundamental modes and generated SF waves is neglected to simplify the derivation of the NCR condition. However, in real on-chip second-order nonlinear processes, several factors must be considered: polarization states of WGMs, i.e. transverse electric (TE) modes or transverse magnetic (TM) modes; nonlinear polarization dependence on the polarization states of the fundamental waves and the $\chi^{(2)}$ tensor of the materials used in microcavities; and the diffraction of SF wave induced by subwavelength scale of the microring. Here, the two fundamental modes are denoted as $E_{\text{P},m}$ for the CCW mode and $E_{\text{P},-n}$ for the CW mode, respectively. The added subscript ‘‘P’’ refers to TE or TM modes. These two modes can be expressed

as follows:

$$\begin{cases} \vec{E}_{\text{P},m} = u_{\text{P},m}(\rho, z) e^{i(\omega_m t + m\varphi)} \hat{e}_{\text{P}} \\ \vec{E}_{\text{P},-n} = u_{\text{P},-n}(\rho, z) e^{i(\omega_n t - n\varphi)} \hat{e}_{\text{P}} \end{cases} \quad (S11)$$

where $\hat{e}_{\text{P}}$ represents the unit vector of the dominant electric field for TE or TM modes, given by

$$\begin{cases} \hat{e}_{\text{TM}} = [0 \ 0 \ 1]^T \\ \hat{e}_{\text{TE}} = [\cos \varphi \ \sin \varphi \ 0]^T \end{cases} \quad (S12)$$

The evolution of the SF field satisfies the vector nonlinear wave equation under the classical slowly varying envelope approximation [28], i.e.,

$$\vec{k}_{\text{SF}} \cdot \nabla \vec{E}_{\text{SF}} = -i \frac{(\omega_m + \omega_n)^2}{c^2} \overleftrightarrow{\chi^{(2)}} : \vec{E}_{\text{P},m} \vec{E}_{\text{P},-n} e^{-i\Delta\vec{k} \cdot \vec{r}} \quad (S13)$$

Here, the colon ‘‘:’’ indicates a double contraction between the tensors, meaning that the second-order polarization $\vec{P}^{(2)}$ is obtained by summing over the products of the components in $\overleftrightarrow{\chi^{(2)}}$ tensor and the electric-field components in $\vec{E}_{\text{P},m}$ and $\vec{E}_{\text{P},-n}$. $\Delta\vec{k}$ is the wavevector mismatch in the NCR process, which can be directly written as

$$\begin{aligned} \Delta\vec{k} &= -k_{\text{SF},\rho} \hat{\rho} - k_{\text{SF},z} \hat{z} \\ &= -k_{\text{SF},\rho} \cos \phi \hat{x} - k_{\text{SF},\rho} \sin \phi \hat{y} - k_{\text{SF},z} \hat{z} \end{aligned} \quad (S14)$$

where $k_{\text{SF},\rho}$ and $k_{\text{SF},z}$ are wavevector components along the radial direction $\hat{\rho}$ and the z -axis direction $\hat{z}$, and ϕ is the azimuthal angle in wavevector space. The disappearance of the azimuthal component in Eq. (S14) is attributed to resonant nature along the azimuthal direction and satisfaction of the NCR condition. Considering $\vec{r} = \rho \cos \varphi \hat{x} + \rho \sin \varphi \hat{y} + z \hat{z}$, the phase mismatch in the cylindrical coordinator system is given by

$$\Delta\vec{k} \cdot \vec{r} = -k_{\text{SF},\rho} \rho \cos(\phi - \varphi) - k_{\text{SF},z} z \quad (S15)$$

By introducing Eq. (S11) into Eq. (S13), we obtain

$$\begin{aligned} \vec{k}_{\text{SF}} \cdot \nabla \vec{E}_{\text{SF}} &= -i \frac{(\omega_m + \omega_n)^2}{c^2} \overleftrightarrow{\chi^{(2)}} : \hat{e}_{\text{P}} \hat{e}_{\text{P}} u_{\text{P},m}(\rho, z) \\ &\times u_{\text{P},-n}(\rho, z) e^{i(m-n)\varphi} e^{-i\Delta\vec{k} \cdot \vec{r}} e^{i(\omega_m + \omega_n)t} \end{aligned} \quad (S16)$$

Based on Eq. (S16), it is possible to engineer optical vortices in multi-dimensions, including the TC, emission wavelength, polarization state and spatial profile, by carefully choosing excitation of resonant modes (m and n) and the nonlinear coefficients $\overleftrightarrow{\chi^{(2)}} : \hat{e}_{\text{P}} \hat{e}_{\text{P}}$. In Eq. (S16), $\overleftrightarrow{\chi^{(2)}} : \hat{e}_{\text{P}} \hat{e}_{\text{P}}$ can be replaced by the vector effective nonlinear coefficient $\vec{\chi}_{\text{eff}} = [\chi_{\text{eff},x} \ \chi_{\text{eff},y} \ \chi_{\text{eff},z}]^T$, which is determined by the polarization states of the interacted modes and the $\chi^{(2)}$ tensor of the materials used in microring.

We firstly focus on the near-field SF radiation on the microring. Since the interacted fundamental beams are confined to the $x-y$ plane and the SF waves radiates upward and downward, each segment of the microring can be modeled as a one-dimensional system where the SF wave propagates along the z -axis. Therefore, the phase mismatch in Eq. (S15) for the upward radiated SF wave becomes $\vec{\Delta k} \cdot \vec{r} = -k_{\text{SF},z}z$, so that Eq. (S16) can be simplified as

$$\begin{aligned} \frac{d\vec{E}_{\text{SF-near}}}{dz} = & -i \frac{(\omega_m + \omega_n)^2}{k_{\text{SF},z}c^2} \vec{\chi}_{\text{eff}} u_{\text{P},m}(\rho, z) \\ & \times u_{\text{P},-n}(\rho, z) e^{i(m-n)\varphi} e^{ik_{\text{SF},z}z} e^{i(\omega_m + \omega_n)t} \end{aligned} \quad (\text{S17})$$

Assuming the thickness Δd of the microring is sufficiently small and that $u_{\text{P},m}(\rho, z)$ and $u_{\text{P},-n}(\rho, z)$ are approximately constant within the microring, equal to $u_{\text{P},m}$ and $u_{\text{P},-n}$, respectively, the integration form of Eq. (S17) can be written as

$$\begin{aligned} \vec{E}_{\text{SF-near}} = & -i \frac{(\omega_m + \omega_n)^2}{k_{\text{SF},z}c^2} \vec{\chi}_{\text{eff}} u_{\text{P},m} u_{\text{P},-n} \\ & \times e^{i(m-n)\varphi} e^{i(\omega_m + \omega_n)t} \Delta d e^{i \frac{k_{\text{SF},z} \Delta d}{2}} \end{aligned} \quad (\text{S18})$$

Obviously, the near field of SFG contains the azimuthal phase term of $e^{i(m-n)\varphi}$ carrying OAM of light and its intensity distribution is determined by $\vec{\chi}_{\text{eff}}$. The far-field SF radiation in the $x-y$ plane can be further obtained by performing the Fourier transform of Eq. (S18), which should only account for the phase mismatch term of $\vec{\Delta k} \cdot \vec{r} = -k_{\text{SF},\rho} \rho \cos(\phi - \varphi)$. Ignoring the terms of $e^{i(\omega_m + \omega_n)t}$ and $e^{i \frac{k_{\text{SF},z} \Delta d}{2}}$, the result is

$$\begin{aligned} \vec{E}_{\text{SF-far}} = & \mathcal{F} \left\{ \vec{E}_{\text{SF-near}} \right\}_{\Delta k} \\ = & -i \frac{(\omega_m + \omega_n)^2}{k_{\text{SF},z}c^2} \Delta d u_{\text{P},m} u_{\text{P},-n} \\ & \times \iint \vec{\chi}_{\text{eff}} e^{i(m-n)\varphi} e^{ik_{\text{SF},\rho} \rho \cos(\phi - \varphi)} \rho d\varphi d\rho \end{aligned} \quad (\text{S19})$$

Further assuming that the width ΔR is also small for the microring with a radius of R , Eq. (S19) can be rewritten as

$$\begin{aligned} \vec{E}_{\text{SF-far}} = & -i \frac{(\omega_m + \omega_n)^2}{k_{\text{SF},z}c^2} u_{\text{P},m} u_{\text{P},-n} R \Delta d \Delta R \\ & \times \int \vec{\chi}_{\text{eff}} e^{i(m-n)\varphi} e^{ik_{\text{SF},\rho} R \cos(\phi - \varphi)} d\varphi \end{aligned} \quad (\text{S20})$$

Eqs.(S18) and (S20) can be used to calculate the TC and the intensity patterns for the SFG by setting $m \neq n$ or the second harmonic generation (SHG) by setting $m = n$ at the near and far fields, respectively.

Formulas for polarization-dependent χ_{eff} and SF radiation

In our configuration, the physical coordinate system of the z -cut TFLN aligns with the lab coordinate system, so that the second-order nonlinear coefficient tensor of the z -cut TFLN in the lab coordinate system can be written as

$$\chi_{z\text{-cut}}^{(2)} = \begin{bmatrix} 0 & 0 & 0 & 0 & \chi_{15} & -\chi_{22} \\ -\chi_{22} & \chi_{22} & 0 & \chi_{15} & 0 & 0 \\ \chi_{31} & \chi_{31} & \chi_{33} & 0 & 0 & 0 \end{bmatrix} \quad (\text{S21})$$

Because the physical coordinate system of the x -cut TFLN is rotated counterclockwise by 90° around the y -axis relative to the lab coordinate system, the second-order coefficient tensor of the x -cut TFLN in the lab coordinate system should be expressed as

$$\chi_{x\text{-cut}}^{(2)} = \begin{bmatrix} -\chi_{33} & -\chi_{31} & -\chi_{31} & 0 & 0 & 0 \\ 0 & \chi_{22} & -\chi_{22} & 0 & 0 & \chi_{15} \\ 0 & 0 & 0 & -\chi_{22} & \chi_{15} & 0 \end{bmatrix} \quad (\text{S22})$$

Only considering the dominant polarization components of the TE and TM WGMs given by Eq. (S12), the nonlinearly interacted electric-field components for TE modes or TM modes are

$$\begin{cases} \hat{e}_{\text{TE}} \hat{e}_{\text{TE}} = [\cos^2 \varphi & \sin^2 \varphi & 0 & 0 & 0 & \sin 2\varphi]^T \\ \hat{e}_{\text{TM}} \hat{e}_{\text{TM}} = [0 & 0 & 1 & 0 & 0 & 0]^T \end{cases} \quad (\text{S23})$$

For the four NCR processes: case 1 involves the interaction between $E_{\text{TM},m}$ and $E_{\text{TM},-n}$ in x -cut TFLN, case 2 involves the interaction between $E_{\text{TE},m}$ and $E_{\text{TE},-n}$ in x -cut TFLN, case 3 involves the interaction between $E_{\text{TE},m}$ and $E_{\text{TE},-n}$ in z -cut TFLN, and case 4 involves the interaction between $E_{\text{TM},m}$ and $E_{\text{TM},-n}$ in z -cut TFLN. The involved effective nonlinear coefficients are

$$\begin{cases} \chi_{x\text{-cut}}^{(2)} : \hat{e}_{\text{TM}} \hat{e}_{\text{TM}} = [-\chi_{31} & -\chi_{22} & 0]^T \\ \chi_{x\text{-cut}}^{(2)} : \hat{e}_{\text{TE}} \hat{e}_{\text{TE}} = [-\chi_{31} \sin^2 \varphi & -\chi_{33} \cos^2 \varphi & \chi_{22} \sin^2 \varphi & -\chi_{15} \sin 2\varphi & 0]^T \\ \chi_{z\text{-cut}}^{(2)} : \hat{e}_{\text{TE}} \hat{e}_{\text{TE}} = [\chi_{22} \sin 2\varphi & \chi_{22} \cos 2\varphi & 0]^T \\ \chi_{z\text{-cut}}^{(2)} : \hat{e}_{\text{TM}} \hat{e}_{\text{TM}} = [0 & 0 & \chi_{33}]^T \end{cases} \quad (\text{S24})$$

Extended Data Figure E1 illustrate effective nonlinear coefficients as functions of the azimuthal angle, with $\chi_{22} = 4$ pm/V, $\chi_{31} = \chi_{15} = 10$ pm/V, and $\chi_{33} = 40$ pm/V [60]. In cases 1 and 2, the dominant polarization components are χ_{31} and $\chi_{33} \cos^2 \varphi$, respectively, leading to the main x -polarized SF wave, denoted by the Jones matrix of $\begin{bmatrix} 1 & 0 \end{bmatrix}^T$. Case 3, described by Eq. (S24), produces spatially varying polarized beam in the $x-y$ plane, represented by a Jones matrix of $[\sin 2\varphi \quad \cos 2\varphi]^T$, due to the superposition of x -polarized and y -polarized states. This cylindrical vector beam can be decoupled into two scalar OAM states with left-handed and right-handed circular

polarizations: $e^{i2\varphi} [1 \ i]^T$ and $e^{-i2\varphi} [1 \ -i]^T$. In case 4, the microring with a subwavelength width acts as a circularly symmetric diffraction structure, scattering the SF or SH fields produced by the z -component of χ_{33} into the far field perpendicular to the z -axis (see Extended Data Fig. E2). This results in radial polarization represented by $[\cos \varphi \ \sin \varphi]^T$, consisting of $e^{-i\varphi} [1 \ i]^T$ and $e^{i\varphi} [1 \ -i]^T$. The Jones matrixes of $[1 \ i]^T$ and $[1 \ -i]^T$ are used to denote the left-handed and right-handed circular polarization states.

Considering the dominant x -polarized state in cases 1 and 2 and the spin-orbital coupling in cases 3 and 4, the near-field SF waves in the $x - y$ plane by substituting Eq.(S24) into Eq.(S18) could be expressed as

$$\left\{ \begin{array}{l} \vec{E}_{\text{SF-N-1}} \propto \chi_{31} \begin{bmatrix} 1 \\ 0 \end{bmatrix} e^{i(m-n)\varphi} \\ \vec{E}_{\text{SF-N-2}} \propto \chi_{33} \begin{bmatrix} 1 \\ 0 \end{bmatrix} \cos^2 \varphi e^{i(m-n)\varphi} \\ \quad = \chi_{33} \begin{bmatrix} 1 \\ 0 \end{bmatrix} \left(\frac{1}{2} + \frac{1}{4} e^{i2\varphi} + \frac{1}{4} e^{-i2\varphi} \right) e^{i(m-n)\varphi} \\ \vec{E}_{\text{SF-N-3}} \propto \chi_{22} \begin{bmatrix} \sin 2\varphi \\ \cos 2\varphi \end{bmatrix} e^{i(m-n)\varphi} \\ \quad = \frac{-i}{2} \chi_{22} \left(e^{i2\varphi} \begin{bmatrix} 1 \\ i \end{bmatrix} - e^{-i2\varphi} \begin{bmatrix} 1 \\ -i \end{bmatrix} \right) e^{i(m-n)\varphi} \\ \vec{E}_{\text{SF-N-4}} \propto \chi_{33} \begin{bmatrix} \cos \varphi \\ \sin \varphi \end{bmatrix} e^{i(m-n)\varphi} \\ \quad = \frac{1}{2} \chi_{33} \left(e^{-i\varphi} \begin{bmatrix} 1 \\ i \end{bmatrix} + e^{i\varphi} \begin{bmatrix} 1 \\ -i \end{bmatrix} \right) e^{i(m-n)\varphi} \end{array} \right. \quad (\text{S25})$$

The corresponding far-field SF waves based on Eqs.(S20) and (S24) are

$$\left\{ \begin{array}{l} \vec{E}_{\text{SF-F-1}} \propto \chi_{31} \begin{bmatrix} 1 \\ 0 \end{bmatrix} e^{i(m-n)\phi} J_{m-n} \\ \vec{E}_{\text{SF-F-2}} \propto \chi_{33} \begin{bmatrix} 1 \\ 0 \end{bmatrix} \left\{ \frac{1}{2} e^{i(m-n)\phi} J_{m-n} \right. \right. \\ \quad \left. \left. + \frac{1}{4} e^{i(m-n+2)\phi} J_{m-n+2} + \frac{1}{4} e^{i(m-n-2)\phi} J_{m-n-2} \right\} \\ \vec{E}_{\text{SF-F-3}} \propto \frac{1}{2} \chi_{22} \left\{ \begin{bmatrix} 1 \\ i \end{bmatrix} e^{i(m-n+2)\phi} J_{m-n+2} - \begin{bmatrix} 1 \\ -i \end{bmatrix} e^{i(m-n-2)\phi} J_{m-n-2} \right\} \\ \vec{E}_{\text{SF-F-4}} \propto \frac{1}{2} \chi_{33} \left\{ \begin{bmatrix} 1 \\ i \end{bmatrix} e^{i(m-n-1)\phi} J_{m-n-1} + \begin{bmatrix} 1 \\ -i \end{bmatrix} e^{i(m-n+1)\phi} J_{m-n+1} \right\} \end{array} \right. \quad (\text{S26})$$

where J_q is the q -order of Bessel beam, defined as

$$J_q(k_{\text{SF},\rho} R) = \int_0^{2\pi} e^{iq\varphi} e^{ik_{\text{SF},\rho} R \cos(\phi-\varphi)} d\varphi \quad (\text{S27})$$

The results of Eqs.(S24) to (S26) are summarized in Extended Data Table E1.

TFLN microring fabrication

General devices fabrication. In the experiment, microring devices are fabricated on both x -cut and z -cut TFLN to perform generation and multidimensional control of structured optical vortices. Extended Data Figure E3a displays the fabrication processes. The devices are prepared from commercial x -cut and z -cut TFLN wafers (NANOLN inc.). These wafers consist of a 2 μm silicon oxide interlayer and a 500 μm silicon substrate. Electron-beam lithography patterns the microring and waveguide using HSQ resist. After developing the pattern, the TFLN is partially etched by argon-ion-based reactive ion etching to create trapezoidal cross sections for the microring and the coupling waveguide. The chip is then cleaned with a buffered HF solution and RCA1 cleaning solution to remove any remaining HSQ resist and etching redeposits. Finally, the chip facets are diced to ensure optimal fiber-to-chip light coupling. Extended Data Figure E3b,c show the SEM images of the fabricated microring. The device parameters used in this work are summarized in Extended Data Table E2. Generation of optical vortices via NCR can be observed in most of the fabricated microrings (e.g. Samples 1 and 3) with varying geometric parameters and Q-factors, while generation of visible vortex combs requires carefully designed dispersion and much higher Q-factors (e.g. Sample 3). Extended Data Figure E4 shows the transmission spectra of the three microring along with the measured quality factors for the TE and TM modes. All the three devices have quality factors up to 10^5 .

Details on fabrication of the z -cut microring for Kerr soliton generation. To achieve the Kerr soliton in the LN microring resonators, the anomalous dispersion is crucial [41]. Since the material dispersion of LN is normal in the communication band, it is necessary to design the cross-section of the microring to make the structural dispersion abnormal. The primary parameters affecting the structural dispersion are the top width, the etching depth, and the radius of the microring. To mitigate bending loss while expanding the spectral width of the soliton microcomb, we set the radius of the microring at 80 μm . Subsequently, to achieve broadband soliton microcomb, the existence of the dispersive wave (DW) is important. Both the width and the etching depth of the microring can effectively change the location of DWs, while they respectively have a different influence on DWs at high and low frequencies. In our experiment, due to the nanoscale uncertainty of the etching depth during the etching process, engineering the dispersion by changing the microring width is more convenient and feasible. Therefore, we fabricate a set of microrings of different widths on a chip to ensure that the position of the DWs is relatively controllable. In the main text, the width of the microring we use is 1.45 μm and the etching depth is 340 nm. As shown

in Extended Data Fig. E5, the measured integrated dispersion (the orange dots) for Sample 3 in Extended Data Table E2 is well consistent with the simulated one (the blue curve), presenting zero integrated dispersion near the wavelength of 1554 nm.

Experimental characterization of nonlinear optical vortices

Experimental setup for characterization of single-wavelength optical vortices. The experimental setup for recording the SF OAM and its spectrum is schematically shown in Extended Data Fig. E6. Two tunable continuous-wave lasers at the C\band are amplified by Erbium Doped Fiber Amplifiers (EDFAs), with their polarization states independently controlled by fiber polarization controllers (FPC). The fundamental beams from these lasers are coupled into the same straight waveguide through lensed fibers from the left and right ports, respectively, exciting the CCW and CW resonator modes. The vertically radiated SF optical vortices, produced by the NCR-assisted nonlinear interaction between the CCW and CW fundamental modes, are captured by a CMOS camera to record the intensity patterns and collected by a spectrometer to record spectra, respectively. A quarter waveplate (WP) and a polarizer placed behind the objective are used to select different polarization states.

On-chip interference for measuring TC. An on-chip interference method is proposed to measure the TC of the SF optical vortex in cases 1 and 2. As shown in Extended Data Fig. E7, laser beams pass through a 2×2 fiber beam splitter (BS), that splits each input into two outputs, and then are coupled into the waveguide from the right and left ports. Consequently, the two fundamental beams with wavelengths λ_m and λ_n are simultaneously coupled into the right and left ports, respectively, so that the CCW and CW modes contained both of the two resonant wavelengths, leading to simultaneous generation of two SF optical vortices with the same wavelength and polarization but reverse TCs. The radiated SF intensity patterns presents as circularly distributed petals due to interference between optical vortices with $l_{\text{WGM}} = \pm(m - n)$.

Characterizing the visible vortex microcombs. The generation of infrared soliton microcombs in high-Q TFLN resonators is a key process in creating broadband visible OAM frequency combs. The generation process of soliton microcomb in z -cut TFLN is simple by leveraging the photorefractive effect, which is typically considered a limiting factor but here is used to extend the soliton existence range [42]. The process begins with a tunable continuous-wave laser, whose power is increased until it

reaches a threshold that allows for the formation of solitons. As the laser frequency is adjusted to the soliton step range, solitons can spontaneously form and stabilize in the microring, exhibiting a broad spectral bandwidth. To observe the SF vortex comb, a blazed grating, a single-photon camera and an optical spectrum analyzer (OSA) are added to the former experimental setup, as illustrated in Extended Data Fig. E9. The formation of the different types of solitons are coupled out by the fiber lens and monitored by the OSA. The radiated SF vortex combs, generated by the CCW infrared combs and the additional CW fundamental beam, are collected by the objective and directed into a spectrometer to measure the spectrum. The vortex combs are then analyzed by the spatial dispersion modulus, where the vortex comb lines are spatially separated by the blazed grating before being captured by the single-photon camera.

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Author Contributions

J.L. and D.Z.W. conceived the project. D.Z.W., C.C. and J.L. developed the theory. B.C., D.Z.W., S.W. and Y.X.W. performed the numerical simulations. S.W., P.Y.W., Z.L.T. and Y.C. fabricated the devices. D.Z.W., B.C., S.W., Y.X.W. and J.L. built the setup and characterized the devices. D.Z.W., B.C., S.W., Y.X.W., G.X.Q., X.S.H., T.J., S.N.F., C.H.D. and J.L. analyzed the data. D.Z.W., X.H.W., C.H.D., and J.L. wrote the manuscript with inputs from all authors. D.Z.W., S.N.F., Q.W.Z., F.B., X.H.W., C.H.D. and J.L. supervised the project.

Competing interests The authors declare no competing financial interests.

Data and materials availability The data sets will be available upon reasonable request.

EXTENDED DATA

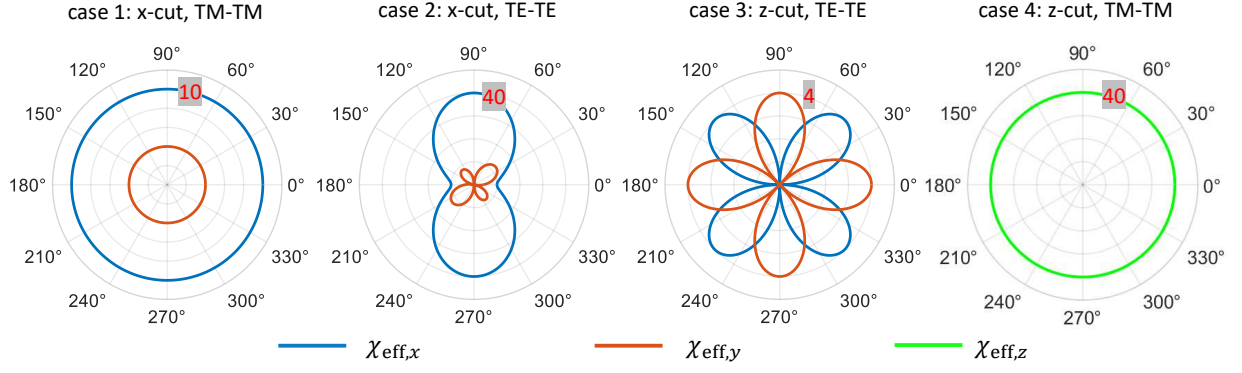


FIG. E1. **Effective nonlinear coefficients dependent on the azimuthal angle for the four cases.** The non-zero nonlinear coefficients of the TFLN are set to with $\chi_{22} = 4$ pm/V, $\chi_{31} = \chi_{15} = 10$ pm/V, and $\chi_{33} = 40$ pm/V.

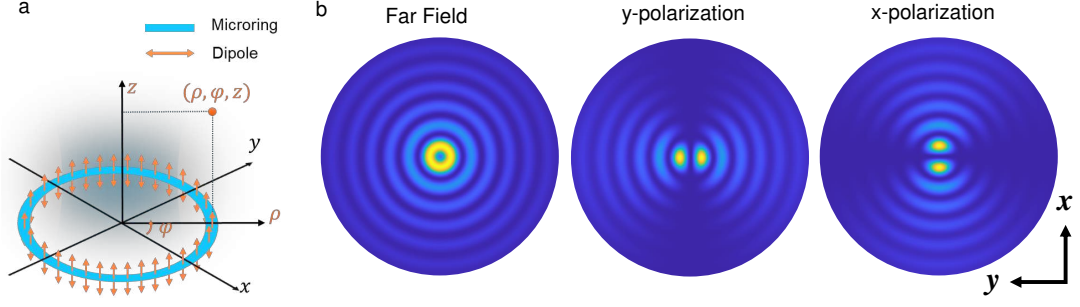


FIG. E2. **Simulation on extraction of nonlinear emissions induced by z -component of the nonlinear polarization into the $x - y$ plane by using finite-different time-domain (FDTD).** (a) Schematic diagram of the dipole model representing the SH sources in the cylindrical coordinate for far-field analysis. z -polarized dipoles azimuthally distributing around a circle in the cylindrical coordinate (ρ, φ, z) represent the sources of generated SH waves. Here, ρ and φ are the polar radius and azimuthal angle, respectively, while z denotes the distance from microring plane. A monitor is placed above the ring to collect the radiated SH field in the plane perpendicular to the z -axis. (b) The simulated SH far-field intensity distribution, horizontal (y -) polarized intensity distribution and vertical (x -) polarized intensity distribution, respectively. The rotation of 90° between x -polarization and y -polarization reveals a cylindrical vector beam $[\cos \varphi \ \sin \varphi]^T$, agree excellently with the experiment results in Fig. 2l,m.

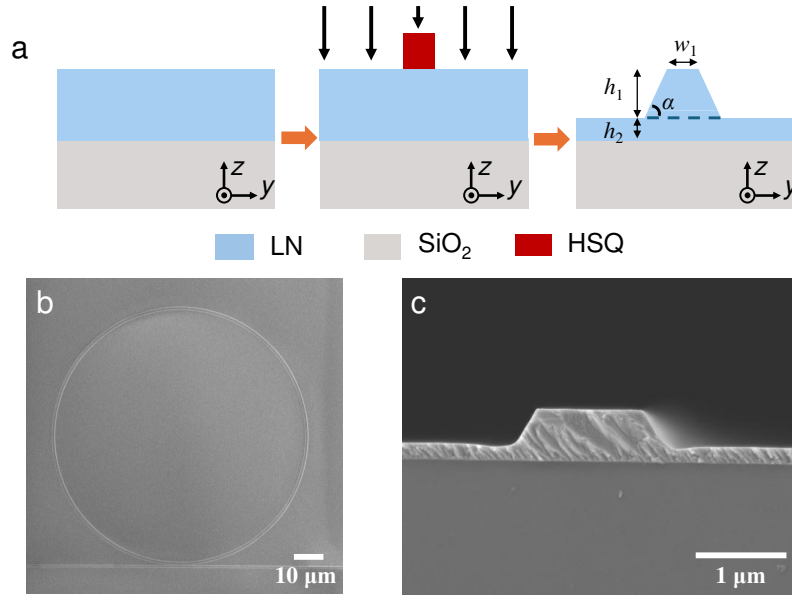


FIG. E3. **Fabrication processes of the LN microring and their SEM images.** (A) Fabrication process; (B-C) Top-view and cross-section SEM images of a typical LN microring.

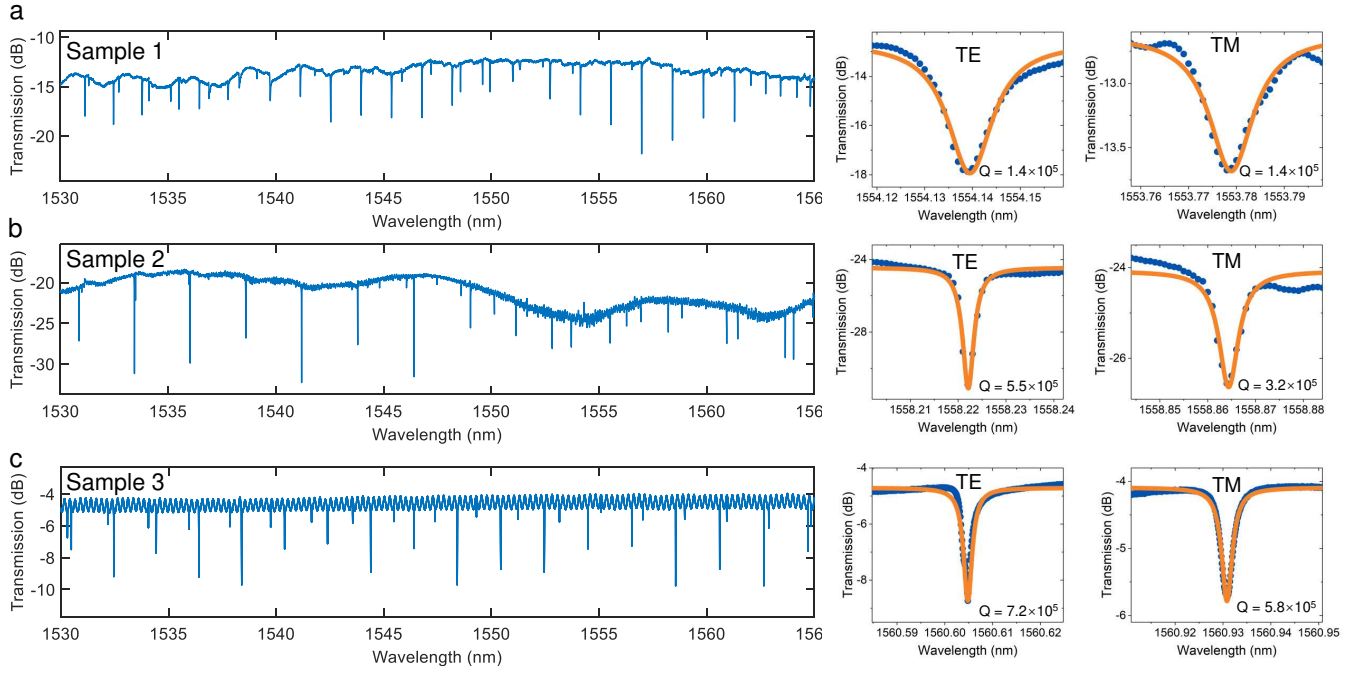


FIG. E4. **Transmission spectra and quality factors of the devices.** (a) Sample 1, *x*-cut microring for generation of scalar SF vortices in cases 1 and 2; (b) Sample 2 *z*-cut microring for generation of vector SF vortices in cases 3 and 4; (c) Sample 3 for generation of visible vortex microcombs.

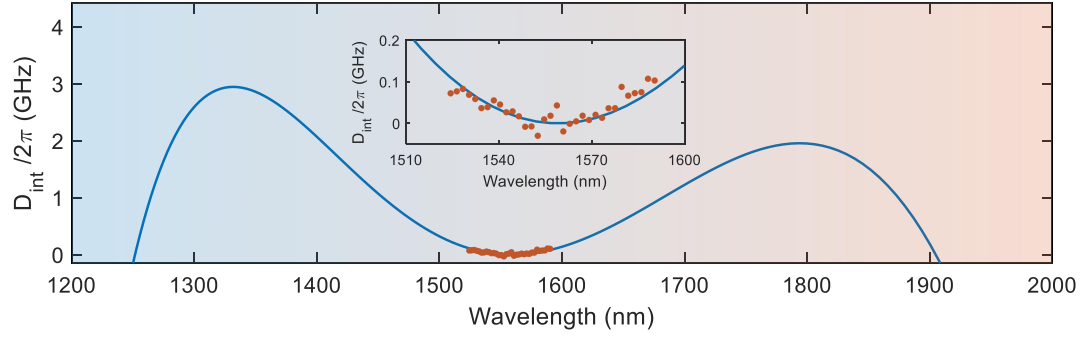


FIG. E5. Simulated and measured integrated dispersions of the z -cut microring (Sample 3) for generation of Kerr soliton.

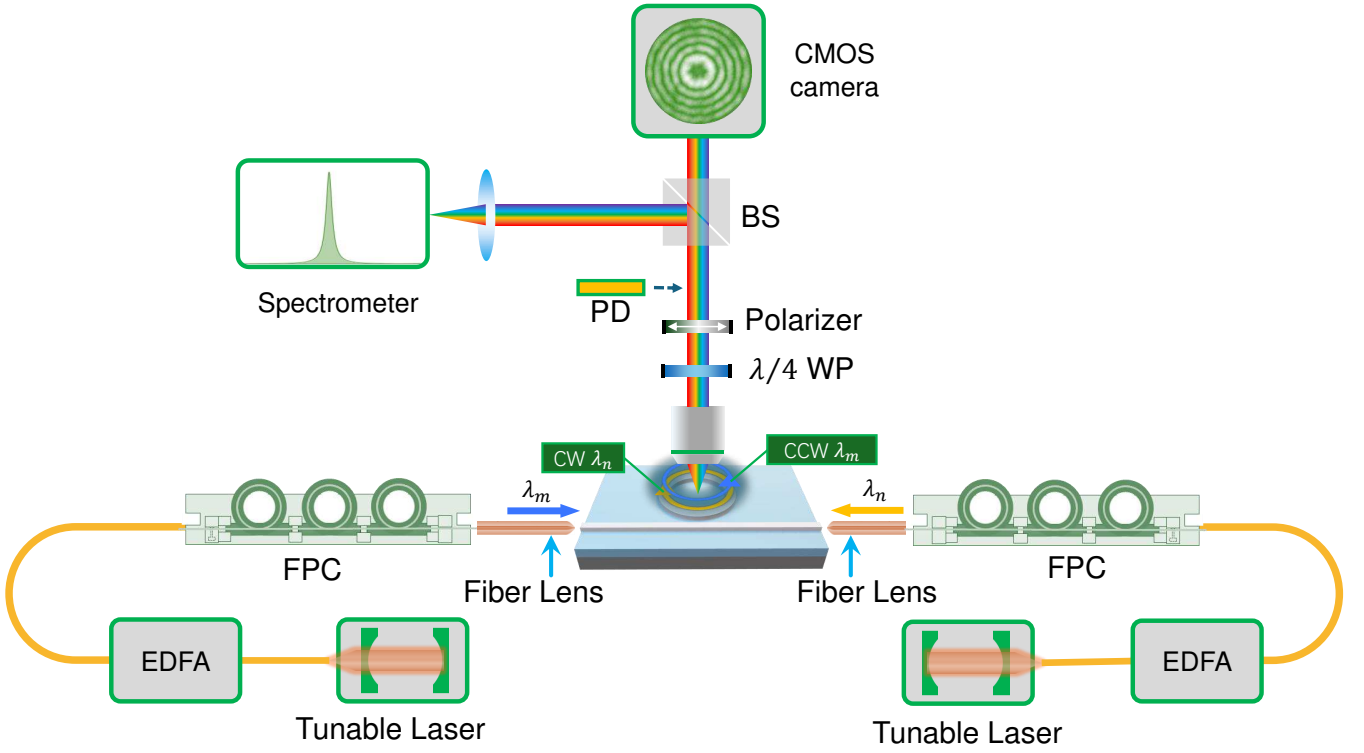


FIG. E6. **Schematic of the experimental setup for characterizing the radiated SH fields or SF vortex fields.** WP: waveplate; EDFA: Erbium Doped Fiber Amplifier; BS: beam splitter; FPC: fiber polarization controllers.

FIG. E7. **Schematic of TC measurement in cases 1 and 2 via on-chip interference.** Fiber BS: 2×2 Fiber beam splitter.

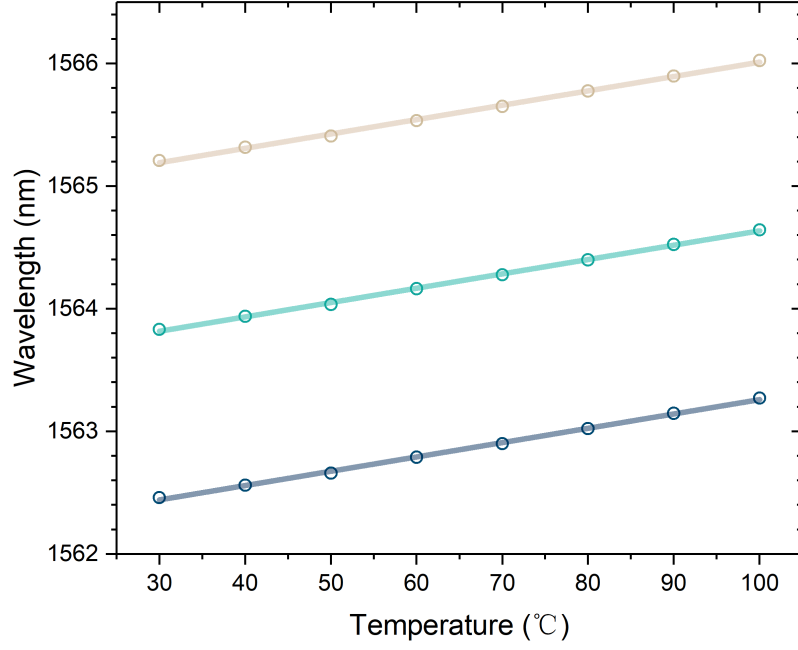


FIG. E8. **Thermal tuning of three resonant TM modes for the x -cut microring (Sample 1).** The microring is placed on the platform with a temperature controller in the fiber-waveguide coupling system. As the temperature increased from 30°C to 100°C, the resonant wavelength for the same mode order exhibits a redshift of approximately 0.8 nm. This shift is larger than half of the free spectral range (1.4 nm for Sample 1), allowing for compensation of the SF wavelength difference for SF optical vortices with even and odd TCs, as shown in Fig. 4d.

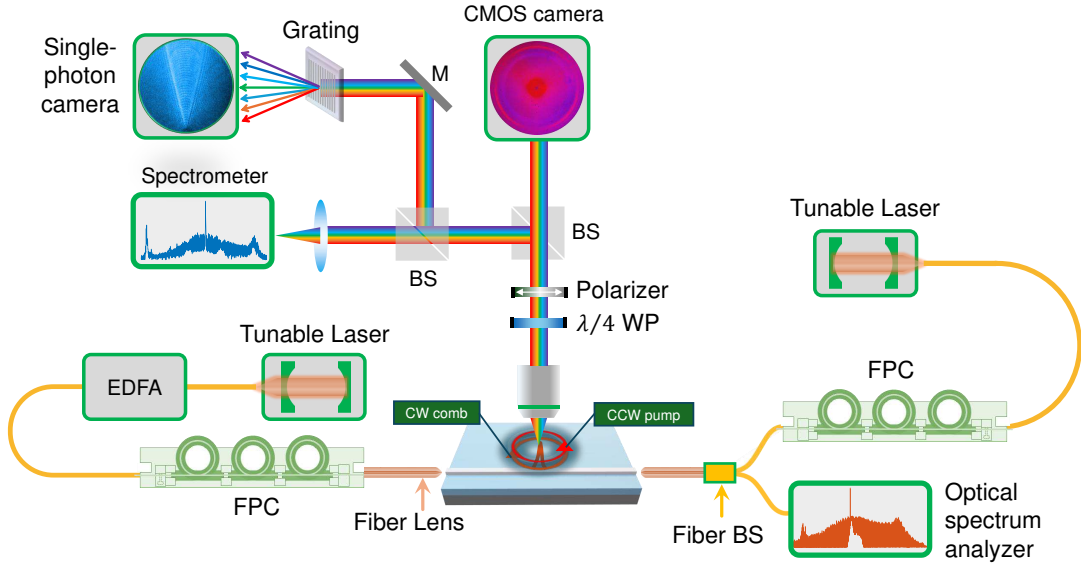


FIG. E9. **Experimental setup for producing and characterizing the visible vortex microcombs.** WP: waveplate; EDFA: Erbium Doped Fiber Amplifier; BS: beam splitter; FPC: fiber polarization controllers; M: mirror.

Table E1. Second-order nonlinear polarization and corresponding radiated SF vortices.

NCR processes	Effective nonlinear coefficients $\vec{\chi}_{\text{eff}} = \overrightarrow{\chi^{(2)}} : \hat{e}_p \hat{e}_p$			Dominant angle-dependent nonlinear polarization in the x-y plane using Jones matrix	Far-field SF radiation in x-y plane	Generated OAM states using ket notation
	$\chi_{\text{eff},x}$	$\chi_{\text{eff},y}$	$\chi_{\text{eff},z}$			
Case 1 x-cut, TM+TM	$-\chi_{31}$	$-\chi_{22}$	/	x-polarization $[1 \ 0]^T$	$\chi_{31} \begin{bmatrix} 1 \\ 0 \end{bmatrix} e^{i(m-n)\phi} J_{m-n}$	$ V, m-n\rangle$
Case 2 x-cut, TE+TE	$-\chi_{31} \sin^2 \varphi$ $-\chi_{33} \cos^2 \varphi$	$\chi_{22} \sin^2 \varphi$ $-\chi_{15} \sin 2\varphi$	/	x-polarization $[1 \ 0]^T$	$\chi_{33} \begin{bmatrix} 1 \\ 0 \end{bmatrix} \left\{ \frac{1}{2} e^{i(m-n)\phi} J_{m-n} + \frac{1}{4} e^{i(m-n+2)\phi} J_{m-n+2} + \frac{1}{4} e^{i(m-n-2)\phi} J_{m-n-2} \right\}$	$\frac{1}{2} V, m-n\rangle$ $\frac{1}{4} V, m-n+2\rangle$ $\frac{1}{4} V, m-n-2\rangle$
Case 3 z-cut, TE+TE	$\chi_{22} \sin 2\varphi$	$\chi_{22} \cos 2\varphi$	/	Cylindrical vector beam $[\sin 2\varphi \ \cos 2\varphi]^T$	$\frac{1}{2} \chi_{22} \left\{ \begin{bmatrix} 1 \\ i \end{bmatrix} e^{i(m-n+2)\phi} J_{m-n+2} - \begin{bmatrix} 1 \\ -i \end{bmatrix} e^{i(m-n-2)\phi} J_{m-n-2} \right\}$	$\frac{1}{2} L, m-n+2\rangle$ $\frac{1}{2} R, m-n-2\rangle$
Case 4 z-cut, TM+TM	/	/	χ_{33}	Cylindrical vector beam $[\cos \varphi \ \sin \varphi]^T$	$\frac{1}{2} \chi_{33} \left\{ \begin{bmatrix} 1 \\ i \end{bmatrix} e^{i(m-n-1)\phi} J_{m-n-1} + \begin{bmatrix} 1 \\ -i \end{bmatrix} e^{i(m-n+1)\phi} J_{m-n+1} \right\}$	$\frac{1}{2} L, m-n-1\rangle$ $\frac{1}{2} R, m-n+1\rangle$

Table E2. Key parameters of the microring devices used in the experiments.

Sample No.		Sample 1	Sample 2	Sample 3
TFLN		<i>x</i> -cut	<i>z</i> -cut	<i>z</i> -cut
Microring	Geometric parameters of the cross-section	$w_{\text{ring}} = 1.9 \text{ } \mu\text{m}$ $h_1 = 500 \text{ nm}$ $h_2 = 100 \text{ nm}$ $\alpha = 75^\circ$	$w_{\text{ring}} = 1.45 \text{ } \mu\text{m}$ $h_1 = 360 \text{ nm}$ $h_2 = 220 \text{ nm}$ $\alpha = 55^\circ$	$w_{\text{ring}} = 1.45 \text{ } \mu\text{m}$ $h_1 = 360 \text{ nm}$ $h_2 = 220 \text{ nm}$ $\alpha = 57^\circ$
	Radius	$R = 115 \text{ } \mu\text{m}$	$R = 60 \text{ } \mu\text{m}$	$R = 80 \text{ } \mu\text{m}$
Width of Coupling waveguide		$w_{\text{wg}} = 1.9 \text{ } \mu\text{m}$	$w_{\text{wg}} = 0.8 \text{ } \mu\text{m}$	$w_{\text{wg}} = 0.8 \text{ } \mu\text{m}$
Coupling gap		$0.3 \text{ } \mu\text{m}$	$0.6 \text{ } \mu\text{m}$	$0.6 \text{ } \mu\text{m}$
Quality factor (TE)		1.4×10^5	5.5×10^5	7.2×10^5
Quality factor (TM)		1.4×10^5	3.2×10^5	5.8×10^5
Repetition rate at C/L band		175 GHz (1.4 nm)	325 GHz (2.6 nm)	250 GHz (2 nm)