Q-plate enabled spectrally diverse orbital-angular-momentum conversion for **STED** microscopy: **supplementary material**

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1. Mode conversion of $q=1/2$ plate

Equation (2) in the main text shows the output of a left/right circularly polarized Gaussian beam passing through a q -plate, allowing us to use the complete basis of circular polarizations $\hat{\sigma}^+$ and $\hat{\sigma}^-$ to linearly decompose any state of polarization (SOP) to calculate the corresponding output. Particularly, for a $q=1/2$ -plate in which the birefringence is maximal at a certain wavelength, i.e.

$\Delta\delta = (2m + 1)\pi$, for $m \in \text{integer}$ (and ignoring the constant phase offset α_0), we obtain for different input polarizations, all possible vortex modes, whose analytical forms are well known [1]:

$$\mathbf{TM}_{01} \propto \mathbf{M} \cdot \hat{\mathbf{x}} = \frac{1}{\sqrt{2}} (\hat{\sigma}^+ e^{-i\varphi} + \hat{\sigma}^- e^{i\varphi}) \quad (\text{S1})$$

$$\mathbf{TE}_{01} \propto \mathbf{M} \cdot \hat{\mathbf{y}} = \frac{1}{i\sqrt{2}} (\hat{\sigma}^+ e^{-i\varphi} - \hat{\sigma}^- e^{i\varphi}) \quad (\text{S2})$$

$$\mathbf{OAM}^{++} \propto \mathbf{M} \cdot \hat{\sigma}^+ = \hat{\sigma}^- e^{i\varphi} \quad (\text{S3})$$

$$\mathbf{OAM}^{*-} \propto \mathbf{M} \cdot \hat{\sigma}^- = \hat{\sigma}^+ e^{-i\varphi} \quad (\text{S4})$$

Figure S1 Camera images of converted TE_{01} , TM_{01} , OAM^+ and OAM^- modes through a $q=1/2$ plate (and a half-wave plate for $\text{OAM}^{(\pm)}$), with and without passing through a linear polarizer placed in front of the camera. The circles with arrows indicate the different transmission axis orientation of the polarizer.

where TM_{01} and TE_{01} are the radially and azimuthally polarized cylindrical vector modes, and $\text{OAM}^{*(\pm)}$ are the so-called spin-orbit anti-aligned orbital angular momentum (OAM) modes since the helical phase front ($e^{i\varphi}$) and the polarization ($\hat{\sigma}^-$) have the opposite handedness. These are, however, not the desired beams for STED microscopy, because they result in non-zero intensity at the center when focused with a high numerical aperture (NA) lens [2]. Using a half-wave plate, $\text{OAM}^{*(\pm)}$ can be readily converted into $\text{OAM}^{(\pm)}$ that are spin-orbit aligned, and which preserve the central null as required for STED – we perform this operation in all the microscopy experiments described in this report. Fig. S1 shows camera images of the polarization projections of each of the modes we excited, confirming that all polarization patterns predicted by Eqs. (2, S1-S4) may be experimentally realized with our q -plate

setup. Note that, because OAM modes are circularly polarized, the projected intensity patterns do not change as the polarizer is rotated. An interesting feature of the q -plate is that switching between the radially, azimuthally, and circularly polarized vortex modes is achieved only by altering the input SOP [3].

2. Phase retardation and group index difference

Light propagating in a medium accumulates phase that is given by $\delta = k \cdot d$, where $k = n \cdot 2\pi/\lambda$ is the wavenumber in that medium, and n is the refractive index of the medium and usually a dispersive quantity, and d is the thickness of the q -plate. For birefringent and dispersive medium like q -plates, we model the two polarizations as two modes in analogy to optical fibers [4]. The phase retardation $\Delta\delta$ in Eq. (1-4) acting as a detuning parameter, after Taylor expansion at some central frequency ω_0 (the central frequency is chosen such that $\Delta\delta = (2m + 1)\pi$, for $m \in \text{integer}$, is given by

$$\Delta\delta = d\Delta k = d \left[\Delta k(\omega_0) + \Delta k'(\omega_0) \cdot \Delta\omega + \frac{1}{2!} \Delta k''(\omega_0) \cdot \Delta\omega^2 + \dots + O(\Delta\omega^n) \right] \quad (\text{S5})$$

where the primes represent derivatives with respect to ω . These derivatives are related to physical quantities such as differential group index Δn_g , differential group velocity (v_g) dispersion (GVD), etc. as the following:

$$\begin{aligned} \Delta n_g &= c\Delta k' = \Delta n(\omega) + \omega\Delta n' \\ \Delta \text{GVD} &= \Delta v_g' = \Delta k'' \end{aligned} \quad (\text{S6})$$

We point out that the birefringence induced phase retardation, and therefore the mode (periodic) conversion property of the q -plate described by Eq. (2), is dominantly controlled by the group index difference Δn_g rather than the index difference Δn of the two polarizations, much like in optical fibers. Higher order terms, like GVD may matter either over very large wavelength ranges or in waveguides (where these quantities can be engineered [4]).

Ignoring GVD effects, and utilizing Eq. (3) from the main manuscripts along with Eq. S5, S6, it becomes clear that the phase retardation is oscillatory with frequency, with the oscillation period $\Delta\omega_p$ being directly related to dispersive characteristics of the liquid crystal by [5]:

$$\Delta\omega_p \propto \frac{1}{d \cdot \Delta n_g} \quad (\text{S7})$$

This provides considerable flexibility in designing the q -plate's material as well as geometric parameters to control $\Delta\omega_p$, which then controls the color combinations possible with STED.

3. Methods

Definition of Extinction Ratio (ER)

Equation (2) in the main text indicates that the local intensity at the center of the output beam comes only from the Gaussian mode, thus allowing us to quantify the modal content of the output beam.

We first set the q -plate bias to generate a Gaussian output (equivalent to when no q -plate is placed in the beam path), and note the peak intensity of such Gaussian beam as 0 dB. Then line-cut profiles through the beam-center are measured at other bias values, and the ratio of the output beam center intensity to the aforementioned Gaussian peak intensity is defined as extinction ratio (ER) as shown in Fig. S2. Clearly, the ER approaches to unity

(0 dB) for a Gaussian beam, and zero ($-\infty$ dB; but practically limited

Figure S2 Measuring the extinction ratio (ER).

by the dynamic range of the imaging camera) for an OAM beam.

Confocal microscopy setup with a q -plate

A home-built confocal laser scanning microscopy setup is used to characterize the performance of the q -plate. Laser beams of multiple colors (indicated by λ_1 and λ_2 in Fig. 4) are combined into a single mode fiber (SMF: RGB 400, Corning) with polarization controllers (F-POL-IL, Newport) to obtain the desired vortex modes (which, as mentioned in S1, may be achieved by controlling input polarization). The combined beam is collimated by a reflective collimator (RC08FC-P01, Thorlabs) to minimize chromatic aberrations, and then passed through a q -plate ($q=1/2$) and a half-wave plate (to form a spin-orbit aligned OAM beam that is normally used in STED nanoscopy), and then focused by an objective lens (UPlanSApo 60X/1.35 oil, Olympus) onto the sample plane. We measure the PSF of the excitation and depletion beams by scanning (NanoMax-TS, Thorlabs) a sample of gold beads (150 nm Au nanoparticle, Cytodiagnostics) mounted using Glycerol on a standard microscope cover slip. The scattered/reflected light is collected by a multimode fiber (M50L02S-A, Thorlabs) that also acts as a confocal pinhole (although confocal is in principle not required in STED), and is detected by a single photon detector (SPCM-AQR-15, Perkin Elmer). A custom-made Labview program together with a data acquisition (DAQ) card (PCIe 6353, National Instruments) are used to control the system. A supercontinuum laser with built-in bandpass filters (Solea, PicoQuant) and a CW Ti: sapphire laser (3900S, Spectral Physics) pumped by a coherent Verdi 10D laser are used to characterize the multicolor PSFs.

The q -plate is biased by a function generator (model 20, Wavetek) that outputs 2 kHz sinusoidal AC voltage with zero DC offset. The q -plate can also be biased through the DAQ to provide further system integration and more accurate and dynamic tuning. All the experiments described in this letter are performed under 2 kHz AC bias.

Determination of the bandwidth of the (Gaussian) excitation beam

We take free-space mode images and measure the corresponding PSFs at different bias settings. Fig. S3 shows the line-cut profiles, mode images and PSFs are obtained, and Fig. S2 discussed how to obtain the ER of a beam. The Gaussian-ness of our beams is quantified by noting that a perfect Gaussian beam, under high-NA focus, will result in a PSF that is a *Jinc* function. Thus, we may define an overlap integral (OL) between line-cuts of the experimentally measured PSFs, with appropriately resized *Jinc* functions (Fig. S3).

We note that when the ER is better than -3 dB (i.e., more negative value), the OL varies only <1% from that of a pure Gaussian beam. No central intensity drop is observed, and the size of the PSF only changes by <30 nm. In contrast, as the ER drops below -7 dB, a central dip of about 22 % intensity drop becomes evident, and the OL drops by about 5%. Thus, we set -3 dB as the

Figure S3 Dashed blue curves: the ideal *Jinc* function. Scatter lines: line-cut profiles of measured PSFs (bottom insets). ER: extinction ratio obtained from free-space mode image (top insets); OL: overlap integral between the scatter line and the *Jinc* function.

ER at which to describe a beam as being Gaussian enough for the purposes of STED microscopy, and we find such a Gaussian like beam may be obtained over as large a bandwidth as 60 nm (Fig. 2 of main text).

4. PSFs results at other wavelength combinations

Besides the results we show in Fig. 5 of main text, PSFs at other wavelength pairs are also measured. Specific wavelengths are chosen based on commonly deployed fluorescence dyes in order to test the feasibility of our system to be applicable for a range of STED microscopy applications. Fig. S4 shows the measured overlapped PSFs (with false colors corresponding to the respective wavelengths of illumination) in the lateral and the axial planes.

We note the following: 1) for the STED beams, all pairs have extinction ratios better than -13 dB, as required for STED nanoscopy, indicating the high quality of donut PSFs generated in our system across a wide spectral (RGB) range; 2) for each wavelength pair, PSFs of the excitation and STED beams are well aligned both in the lateral and the axial plane, which addresses the co-alignment challenge of currently deployed STED systems. It is also expected that since two colors originate from the same single mode fiber aperture, they will be much less susceptible to environmental changes compared to systems using two distinct sources; 3) other than changing the *q*-plate bias settings, no optical re-alignment is needed for switching between wavelength pairs.

The transmission loss of the *q*-plate is measured at different wavelengths. Fig. S5 shows the results. The overall loss in the visible wavelength range is less than 0.5 dB, corresponding to >89% transmission. Note that, this already low loss value can be further reduced by applying appropriate antireflection coatings to the substrate of the *q*-plate device.

Figure S4 Overlapped PSFs of excitation and STED beams at multiple wavelengths (at corresponding bias settings for the *q*-plate). Blue traces are line-cut profiles along the center of donut PSFs, and inset numbers describe the STED wavelength and the PSF extinction ratio in dB. Scale bars: 500 nm.

Figure S5 Measured transmission loss of the q -plate as a function of wavelength. The power meter reading has a fluctuation of about ± 0.06 dB, manifested as the error bars shown in the figure.

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