Supporting Information

A Plasmonic Fano Switch

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This documents contains details regarding the description of the single particle measurements, reconstructed polarized spectra of the octamer using the orthogonal polarized Fano-like and non-Fano-like spectra, experimental and simulated spectra of an octamer in air, dark-field image of a liquid crystal cell composed of octamers and IDA electrodes, and Jones matrix description for the simulation of the liquid crystal transmittance spectra.

Single particle spectroscopy:

All single particle scattering measurements were performed using a homebuilt dark-field spectroscopy setup based on a commercial microscope (Zeiss, Observer.D1m) in the transmission dark-field geometry. The light source was a halogen lamp. The unpolarized light from the lamp was focused by a dark-field condenser (Zeiss, numerical aperture (NA) = 1.40), and the transmitted scattered light was collected by a 63X oil immersion objective (Zeiss, NA = 0.7). The image was then focused onto a 50 μm pinhole and guided to a liquid nitrogen cooled CCD camera attached to a spectrograph (Horiba Jobin Yvon). The pinhole replicated a confocal scheme and only transmitted the scattered light from a specific region of interest. A polarizer was inserted in front of the detector. All measured single particle spectra were corrected for the background scattering of the liquid crystal solvent. All data analysis was performed using Matlab.

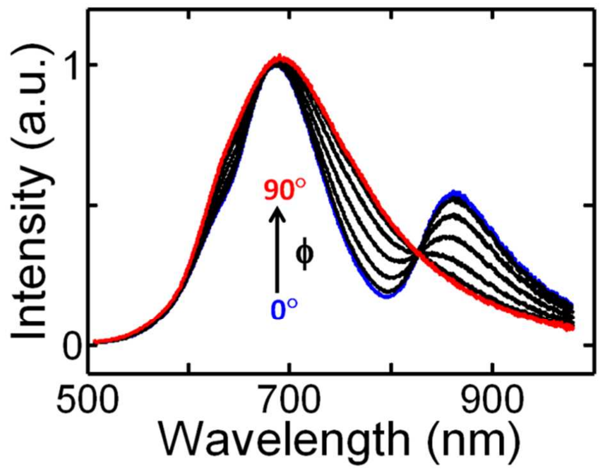


Figure S1: Reconstructed polarized spectra of the octamer using the orthogonal polarized Fano-like and non-Fano-like spectra (0° and 90°). The spectra were calculated according to $I_{cal}(\phi) = cos^2\phi \times I(0°) + sin^2\phi \times I(90°)$, where $I_{cal}(\phi)$ is the calculated spectrum with arbitrary polarization angle ϕ, *I(0°)* is the spectrum of the octamer when the detection polarizer was oriented at 0° (blue line from Figure 1A), *I(90°)* is the spectrum of the octamer when the detection polarizer was oriented at 90° (red line from Figure 1A). These weighted averages (black lines) of the 0° and 90° cases reproduce the experimental spectra obtained when the polarizer was varied between 0° and 90° (Figure 1A), illustrating that the 0° and 90° cases are the fundamental modes of the asymmetric octamer.

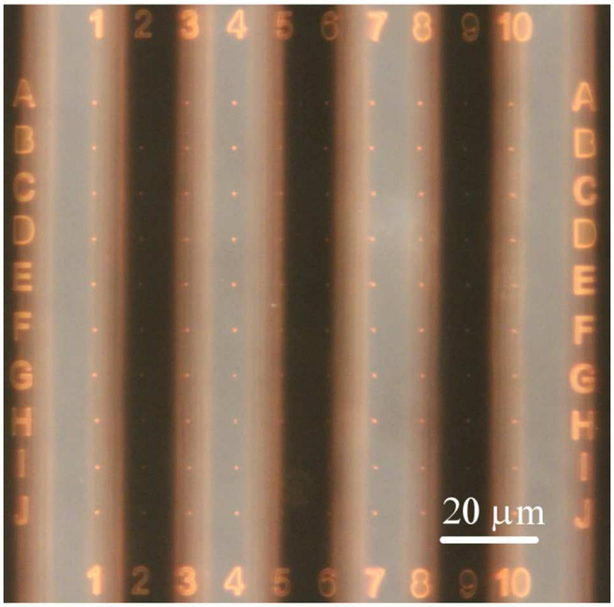


Figure S2: Dark-field image of a liquid crystal cell composed of octamers and IDA electrodes. The bright dots are the octamers while the black bars are the IDA electrodes.

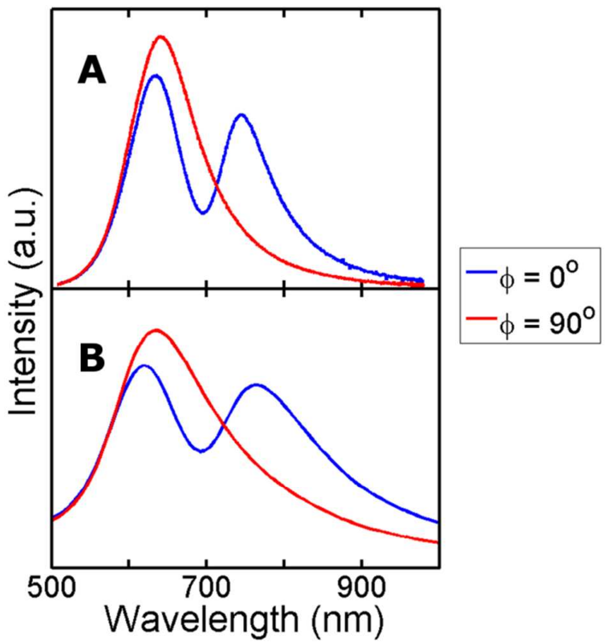
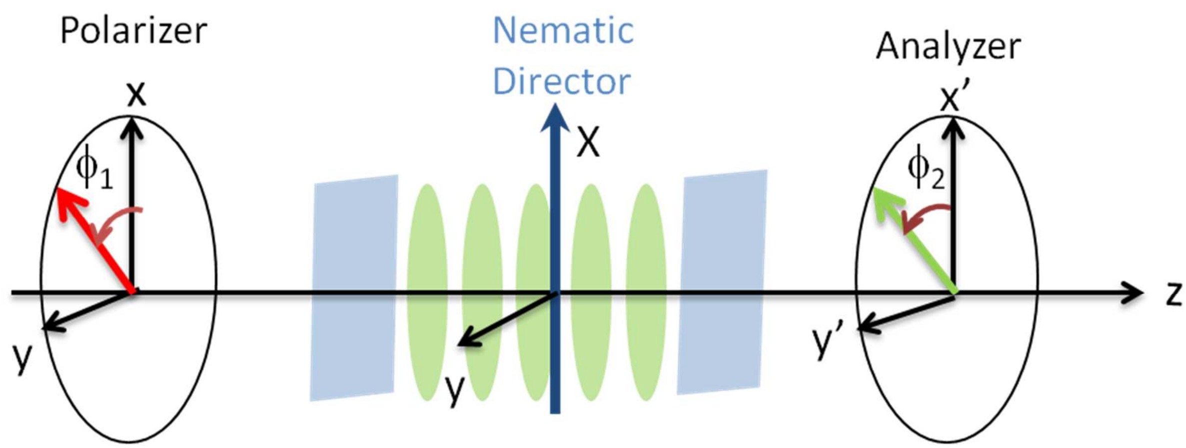


Figure S3: Experimental (A) and simulated (B) spectra of an octamer in air with polarization at 0° (blue) and 90° (red).

Simulation of the liquid crystal transmittance spectra:

A. Homogeneous nematic liquid crystal phase



The director of the liquid crystal was assumed to be along the x-axis. The input and output polarization directions were ϕ_1 and ϕ_2, respectively, defined with respect to the x-axis. For a homogenous nematic liquid crystal, the Jones matrix describing the change in polarization of an incident electromagnetic wave is given by:[1]

$$M_{HNLCD} = \begin{pmatrix} 1 & 0 \\ 0 & e^{-i\chi} \end{pmatrix} \qquad \text{(S1)}$$

where $\chi = \frac{2\pi d \Delta n}{\lambda}$. λ is the wavelength, *d* is the cell thickness and measured to be 7.5 μm, and $\Delta n = 0.2$ is the difference in extraordinary and ordinary refractive indices for 5CB.

The Jones vector describing the incident light wave through the polarizer is:

$$\begin{bmatrix} \boldsymbol{E}_x \\ \boldsymbol{E}_y \end{bmatrix} = \begin{bmatrix} \cos\phi_1 \\ \sin\phi_1 \end{bmatrix} \qquad \text{(S2)}$$

After passing through the homogeneous liquid crystal cell and an analyzer one obtains:

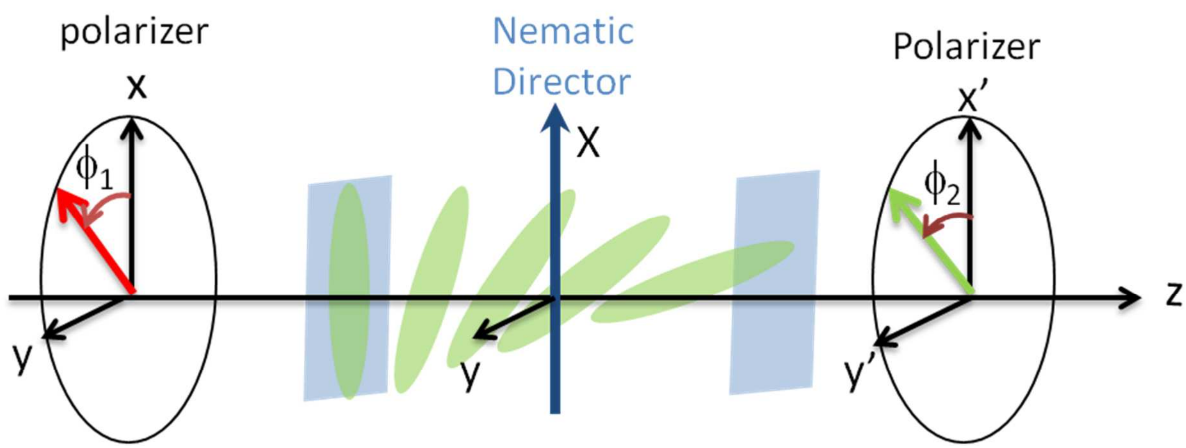
$$\begin{bmatrix} \boldsymbol{E}_{x'} \\ \boldsymbol{E}_{y'} \end{bmatrix} = \begin{bmatrix} \cos\phi_2 & -\sin\phi_2 \\ \sin\phi_2 & \cos\phi_2 \end{bmatrix} \begin{bmatrix} 1 & 0 \\ 0 & 0 \end{bmatrix} \begin{bmatrix} \cos\phi_2 & \sin\phi_2 \\ -\sin\phi_2 & \cos\phi_2 \end{bmatrix} \begin{bmatrix} 1 & 0 \\ 0 & e^{-i\chi} \end{bmatrix} \begin{bmatrix} \cos\phi_1 \\ \sin\phi_1 \end{bmatrix} \qquad \text{(S3)}$$

The transmittance is given by:

$$\boldsymbol{T} = \left|\boldsymbol{E}_{x'}\right|^2 + \left|\boldsymbol{E}_{y'}\right|^2 = \cos^2(\phi_1 + \phi_2) + 4\sin\phi_1\cos\phi_1\sin\phi_2\cos\phi_2\cos^2\frac{\chi}{2} \quad \text{(S4)}$$

Therefore, $T = 1$ when $\phi_1 = 0°$, $\phi_2 = 0°$ or $\phi_1 = 90°$, $\phi_2 = 90°$ independent of λ. $T = 0$ when $\phi_1 = 0°$, $\phi_2 = 90°$ or $\phi_1 = 90°$, $\phi_2 = 0°$ independent of λ.

B. Twisted nematic liquid crystal phase



The director of the twisted nematic liquid crystal cell rotates from the x- to the y-axis across the sample thickness (z-axis). The input and output polarization directions are ϕ_1 and ϕ_2, respectively, defined with respect to the x-axis. For a twisted nematic liquid crystal, the Jones matrix describing the change in polarization of an incident electromagnetic wave is given by:[1]

$$\boldsymbol{M}_{TNLCD} = \exp(-i\beta)\begin{pmatrix} \cos\alpha & -\sin\alpha \\ \sin\alpha & \cos\alpha \end{pmatrix}\begin{pmatrix} \cos\gamma - \frac{i\beta\sin\gamma}{\gamma} & \frac{\alpha\sin\gamma}{\gamma} \\ -\frac{\alpha\sin\gamma}{\gamma} & \cos\gamma + \frac{i\beta\sin\gamma}{\gamma} \end{pmatrix} \quad \text{(S5)}$$

where $\alpha = \frac{\pi}{2}$, $\beta = \pi d\Delta n/\lambda$, and $\gamma = \sqrt{\alpha^2 + \beta^2}$.

The Jones vector describing the incident light wave through the polarizer is:

$$\begin{bmatrix} \boldsymbol{E}_x \\ \boldsymbol{E}_y \end{bmatrix} = \begin{bmatrix} \cos\phi_1 \\ \sin\phi_1 \end{bmatrix} \tag{S6}$$

The Jones vector describing the light polarization after the twisted liquid crystal cell and an analyzer is given by:

$$\begin{bmatrix} \boldsymbol{E}_{x'} \\ \boldsymbol{E}_{y'} \end{bmatrix} = \exp(-\boldsymbol{i\beta}) \begin{bmatrix} \cos\phi_2 & -\sin\phi_2 \\ \sin\phi_2 & \cos\phi_2 \end{bmatrix} \begin{bmatrix} 1 & 0 \\ 0 & 0 \end{bmatrix} \begin{bmatrix} \cos\phi_2 & \sin\phi_2 \\ -\sin\phi_2 & \cos\phi_2 \end{bmatrix} \begin{pmatrix} 0 & -1 \\ 1 & 0 \end{pmatrix} \begin{pmatrix} \cos\gamma - \frac{\boldsymbol{i\beta}\sin\gamma}{\gamma} & \frac{\boldsymbol{\alpha}\sin\gamma}{\gamma} \\ -\frac{\boldsymbol{\alpha}\sin\gamma}{\gamma} & \cos\gamma + \frac{\boldsymbol{i\beta}\sin\gamma}{\gamma} \end{pmatrix} \begin{bmatrix} \cos\phi_1 \\ \sin\phi_1 \end{bmatrix} \tag{S7}$$

The transmittance is given by:

$$\boldsymbol{T} = \left|\boldsymbol{E}_{x'}\right|^2 + \left|\boldsymbol{E}_{y'}\right|^2$$

$$\boldsymbol{T} = \left[-\cos\gamma \sin(\phi_1 - \phi_2) + \frac{\boldsymbol{\alpha}\sin\gamma}{\gamma}\cos(\phi_1 - \phi_2) \right]^2 + \left[\frac{\boldsymbol{\beta}\sin\gamma}{\gamma}\sin(\phi_1 + \phi_2) \right]^2 \tag{S8}$$

For $\phi_1 = 0°$, $\phi_2 = 0°$ or $\phi_1 = 90°$, $\phi_2 = 90°$, $T = \left(\frac{\alpha}{\gamma}\right)^2 sin^2\gamma \approx 0$.

For $\phi_1 = 0°$, $\phi_2 = 90°$ or $\phi_1 = 90°$, $\phi_2 = 0°$, $T = 1 - \left(\frac{\alpha}{\gamma}\right)^2 sin^2\gamma \approx 1$.

Considering the experimental parameters of d = 7.5 μm, Δn = 0.2, and λ = 500 - 1000 nm, the quantity $\left(\frac{\alpha}{\gamma}\right)^2 sin^2\gamma$ is always smaller than 0.03 for the entire wavelength range.

These equations were applied to model the transmission spectra (500 - 1000 nm) for light passing through our liquid crystal devices with different polarizer and analyzer settings in the voltage off and on states. Essentially, in these simulations unpolarized white light was incident on a polarizer, then passed through a liquid crystal device, followed by an analyzer, and then observed by a detector. The polarizer/analyzer angles are defined relative to the nematic director, which was modeled as a homogenous nematic phase (V_{off} state) and a twisted nematic phase (V_{on} state). The transmittance was calculated by normalizing the detected optical intensity with respect to the intensity after the initial polarizer and before the liquid crystal device. Figures S3A and S3B illustrate the simulated transmittance spectra of the liquid crystal device with the analyzer angle set to A = 0° (S3A) and A = 90° (S3B), respectively. The green (magenta) lines represent the transmittance with the polarizer angle set to P = 0° (P = 90°). The solid (dashed) lines indicate the V_{off} (V_{on}) state in the liquid crystal device. Without applying a voltage to the device, the transmittance is 100% when the polarizer and analyzer angles are aligned parallel to each other (solid green line in Figure S3A and solid magenta line in Figure S3B). The transmittance is 0% when the polarizer and analyzer are oriented orthogonal to each other (solid magenta line in Figure S3A and solid green line in Figure S3B). These results show that the polarization state of the incident light remains unaltered after passing through the homogenous nematic liquid crystal (V_{off} state), independent of wavelength over the wavelength range used to characterize the device. Indeed, at P = 0° and 90°, the incident polarization is parallel to the extraordinary and ordinary optical axes of the liquid crystal, respectively and therefore the incident light does not experience any net birefringence which would result in phase retardation. However, when a voltage is applied to the liquid crystal device (V_{on}), the polarization is rotated by 90° by the twisted nematic phase of the liquid crystal. The twisted nematic phase is formed because the liquid crystal molecules in the gap between the IDA electrodes reorient with the direction of the electric field, but the liquid crystal molecules near the polyimide layer on the ITO substrate remain aligned along the rubbing direction. Thus the nematic director changes continuously from 0° to 90° throughout the cell in the direction of light propagation. This twisted nematic phase causes the polarization of light passing through the liquid crystal to be rotated with the director, inducing an orthogonal polarization rotation. In the V_{on} state, the transmittance is almost 100% if the polarizer and analyzer angles are orthogonal (magenta dashed line in S3A and green dashed line in S3B) and near 0% when the polarizer and analyzer angles are aligned parallel (green dashed line in S3A and magenta dashed line in S3B).

Using the results from Figure S3, we can confirm that the far-field scattering spectra of the octamer are rotated by 90° after passing through the liquid crystal device in the V_{on} state (Figures 4A and 4B). The resulting spectra in Figure 4 were calculated according to

$$\boldsymbol{I}(\lambda,\phi_2) = \boldsymbol{T}(\lambda,\phi_1,\phi_2)\boldsymbol{I}_0(\lambda,\phi_1) \tag{1}$$

where $I_0(\lambda,\phi_1)$ is a scattering spectrum of the octamer (Figure 1B); $T(\lambda,\phi_1,\phi_2)$ is the transmittance spectrum of the liquid crystal device shown in Figures S3A and S3B; $I(\lambda,\phi_1)$ is the detected scattering spectrum after passing through the liquid crystal device and the analyzer; λ is the wavelength; ϕ_1 and ϕ_2 are the polarizer and analyzer angles, respectively.

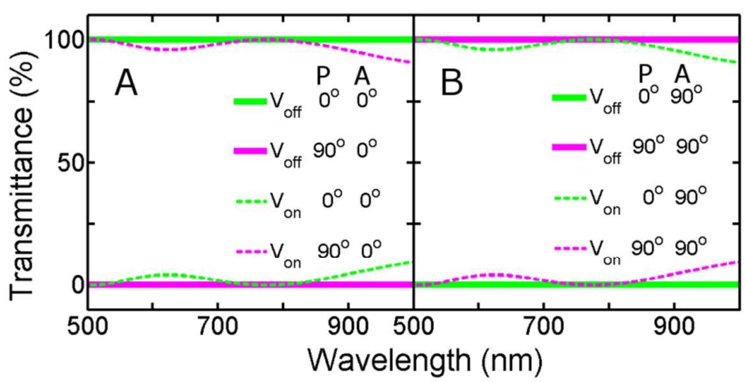


Figure S4: Simulated transmittance spectra of the liquid crystal device with the analyzer angle set to A = 0° (A) and A = 90° (B). Green and magenta lines (solid, dashed) represent incident light with polarizer angle set to P = 0° and 90°, respectively. Solid and dashed lines identify the V_{off} and V_{on} states, respectively. With V_{off}, the incident polarization was invariant after passing through the liquid crystal device independent of wavelength. With V_{on}, the incident polarization was rotated almost perfectly by 90° over the entire wavelength range.

References:

1. Yamauchi, M.; Márquez, A.; Davis, J. A.; Franich, D. J. *Opt. Comm.* **2000,** 181, 1-6.