Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Information

Measuring the Order Parameter of Vertically Aligned Nanorods Assembly

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In-plane alignment of nanorods by electro-optical switching

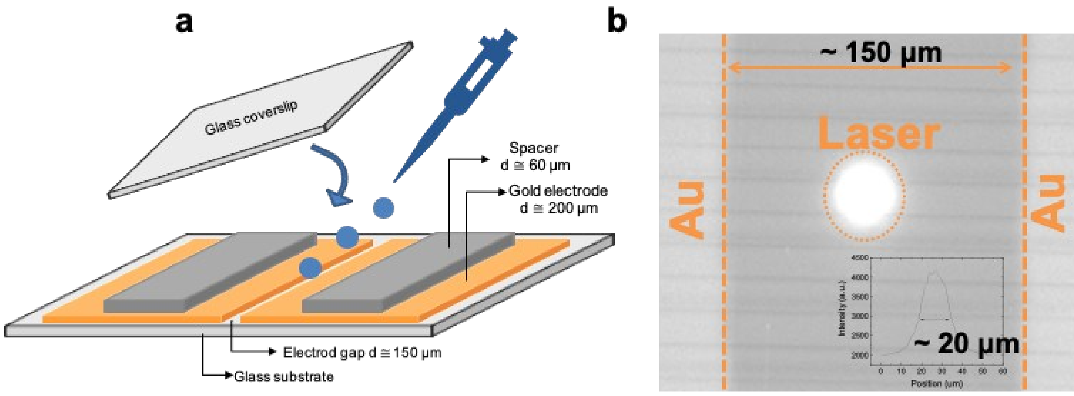

Figure S1. **(a)** Illustration of preparation of lab-made in-plane switching (IPS) electro-optical cell and switching experiment **(b)** CCD captured image of lab-made IPS electro-optical cell. Inset: intensity profile of laser excitation area with a full-width-half-max (FWHM) of ~20 µm.

Figure S1a shows the lab-made in-plane switching (IPS) cell. The cell has a gap of ~ 150 µm between Au electrodes on the glass substrate. Around 20 µL of $NaYF_4$:Eu colloidal nanorods dispersed in ethylene glycol (EG) is deposited between electrodes and covered by a glass cover slip with the spacer of thickness of around 60 µm. **Figure S1b** shows the CCD-captured image of lab-made IPS cell. The bright circle in the middle of electrodes is the position where the $NaYF_4$:Eu nanorods are excited by UV-laser (λ~394 nm). As the E-field $\vec{n}$ of IPS cell is transverse only at the middle of electrodes, it is important to collect PL signals at the middle of two electrodes from the smallest region**. Figure S1b-inset** shows the measured intensity profile of the excited area, showing around 20 µm of full-width-half-max (FWHM).

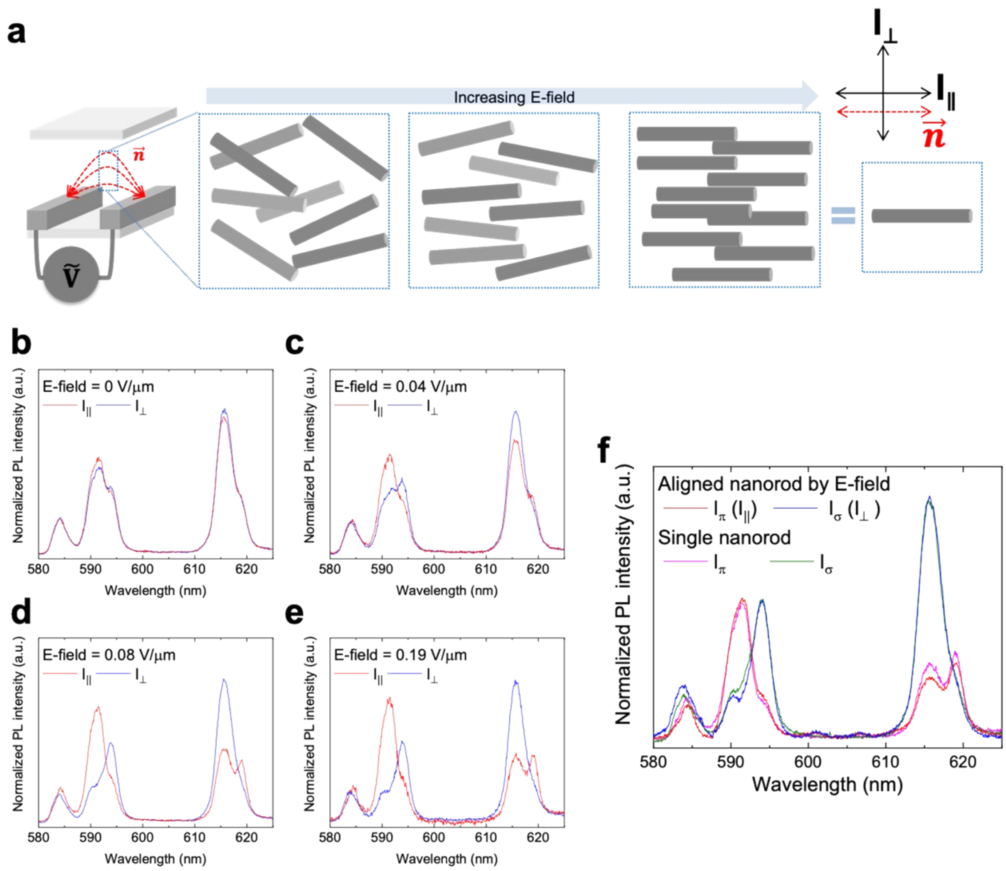

Figure S2. **(a)** Schematic illustration of IPS electro-optical (E/O) cell with aligning nanorods as increasing E-field. Red dashed line stands for the direction of E-field $\vec{n}$. At the middle of two electrodes, E-field direction $\vec{n}$ is transverse where polarized photoluminescence (PL) spectra of aligned $NaYF_4$:Eu nanorods are measured. Polarized photoluminescence spectra of orientation-controlled $NaYF_4$:Eu nanorods measured at different E-field intensity of **(b)** 0 **(c)** 0.04 **(d)** 0.08 and **(e)** 0.19 V/μm (f = 1 kHz) with the polarizer set to parallel ($I_{\parallel}$, red line) and perpendicular ($I_{\perp}$, blue line) to the direction of E-field direction $\vec{n}$ **(f)** Reference spectra (I_{π}, I_{σ}) obtained from aligned nanorods under the E-field (at saturated voltage, E ~ 0.47 V/μm) and from the single nanorod.

Nanorods submitted in such in-plane E-field begin to align parallel to field direction $\vec{n}$ and in-plane order parameter **S** increases as schematized in **Figure S2a**. When there is no applied E-field, measured spectra at two orthogonal polarization shows negligible changes as nanorods are randomly oriented (**Figure S2b**). Upon increasing E-field strength, observed line shape of PL signal is different when measured with a polarizer set to parallel ($I_{\parallel}$) and perpendicular ($I_{\perp}$) to $\vec{n}$ (**Figure S2c-e**). When E-field intensity is higher than ~ 0.47 V/μm, line shape showed no longer changes. At this saturating E-field intensity, the line shape of π signal obtained from $I_{\parallel}$ (red line) and the σ signal obtained from $I_{\perp}$ (blue line) is identical to those obtained from the single nanorod (magenta and olive line), indicating alignment of nanorods by E-field is close to perfect in-plane alignment (**S**~1) (**Figure S2f**).

Line shape comparison of $I_\pi + I_\sigma$ and $I_{\pi+\sigma}$

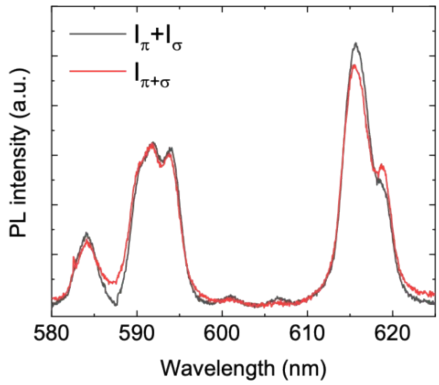

Figure S3. Sum of separately measured emission spectra of π and σ configuration (I_π+I_σ) from the electrically aligned $NaYF_4$:Eu nanorod assembly and the emission spectrum of randomly deposited $NaYF_4$:Eu nanorods lying on the substrate ($I_{\pi+\sigma}$).

Photoluminescence signal of $NaYF_4$:Eu nanorods as a function of relaxation time after the E-field turned off from saturating E-field

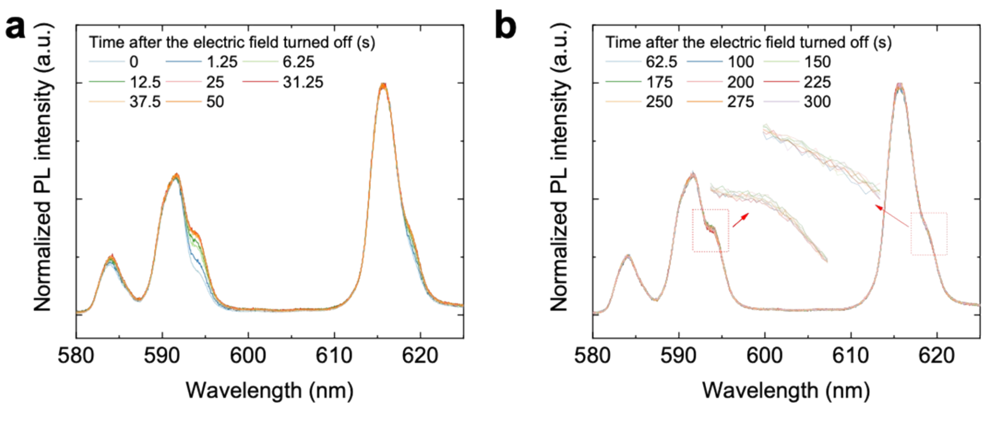

Figure S4. Photoluminescence spectra of $NaYF_4$:Eu nanorods collected as a function of relaxation time after the E-field field turned off from the saturating E-field. All spectra are normalized to the intensity at peak λ~615 nm.

Simulated profile of homeotropic order parameter analysis

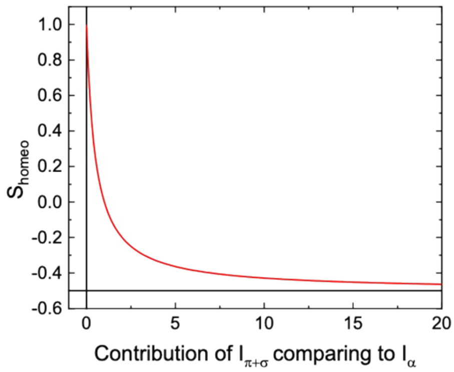

Figure S5. Simulated homeotropic order parameter $\mathbf{S_{homeo}}$ as a function of $I_{\pi+\sigma}$ contribution comparing to I_{α} . Simulation is made according to **Equation 5**.

$NaYF_4$:Eu nanorods characterization

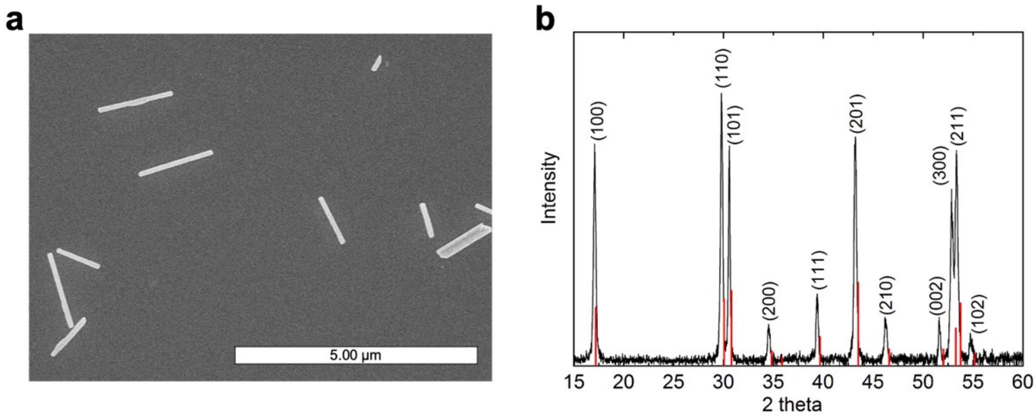

Figure S6. **(a)** Scanning electron microscopy (SEM) image of $NaYF_4$:Eu nanorods. **(b)** X-ray diffraction (XRD) pattern of $NaYF_4$:Eu nanorods. Reference pattern of hexagonal β-$NaYF_4$ (JCPDS 16-0334) is plotted in red line.