Supporting Information

DNA Self-Switchable Microlaser

Yifan Zhang [1], Xuerui Gong [1], Zhiyi Yuan [1], Wenjie Wang [2★], Yu-Cheng Chen [1, 3★]

[1] School of Electrical and Electronic Engineering, Nanyang Technological University, 50 Nanyang Avenue, 639798, Singapore

[2] Key Lab of Advanced Transducers and Intelligent Control System of Ministry of Education, Taiyuan University of Technology, 030024, P. R. China

[3] School of Chemical and Biomedical Engineering, Nanyang Technological University, 62 Nanyang Drive, 637459, Singapore

★*Correspondence Email:* yucchen@ntu.edu.sg

★*Correspondence Email:* wangwenjie@tyut.edu.cn

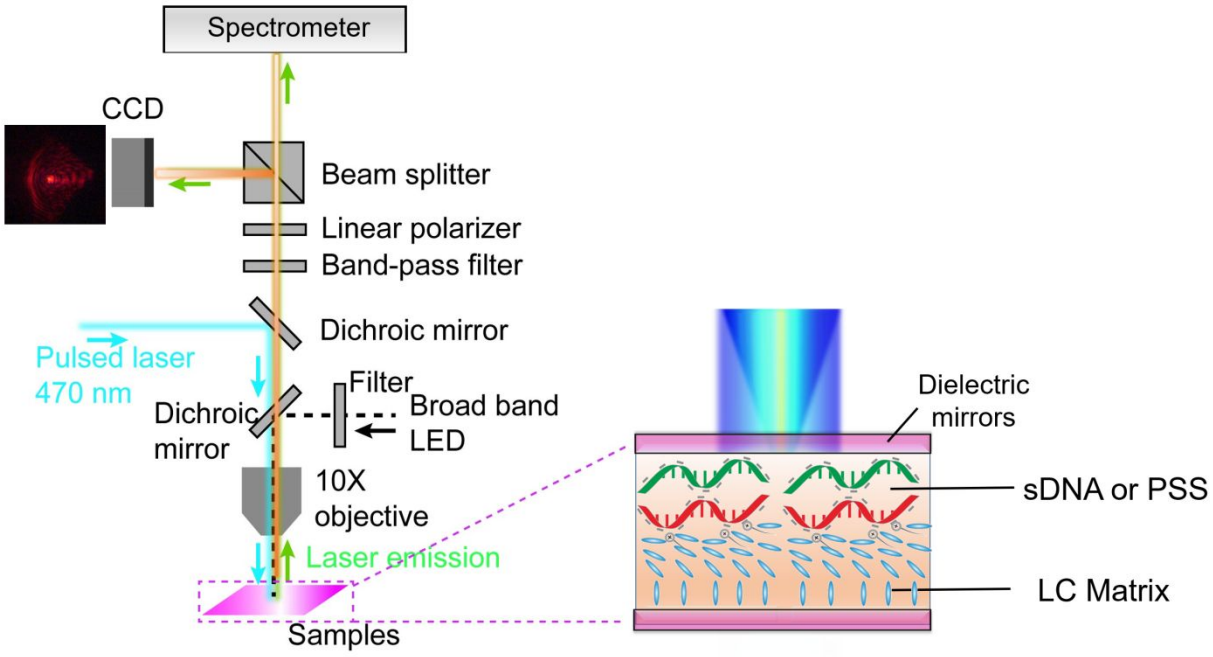

Figure S1. Schematic diagram of the optical experiment setup. Excitation=470 nm. The F-P cavity is formed by two highly reflective dielectric mirrors with LC matrix and PSS/DNA filled inside.

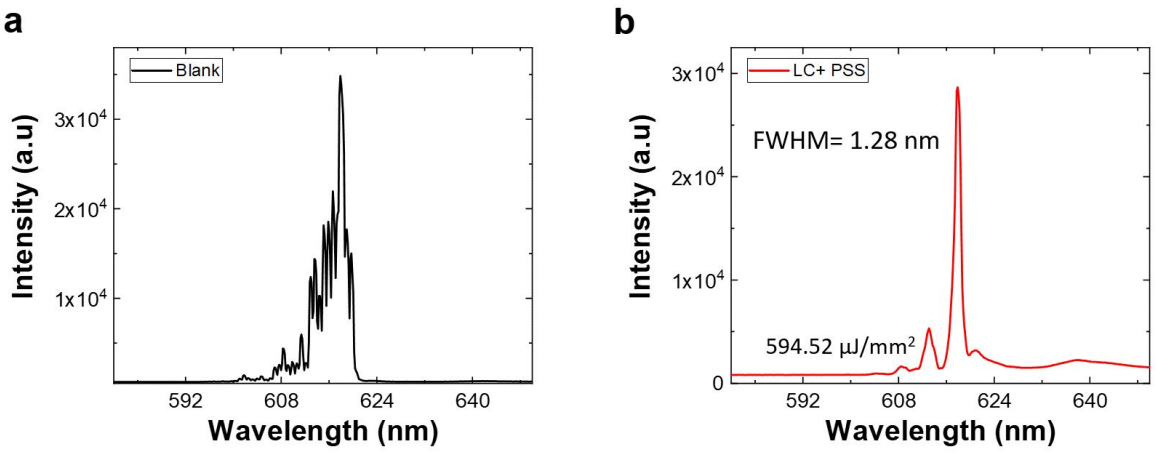

Figure S2. (a) Lasing spectra of DCM-LC matrix filled with DI water in F-P cavity. (b) Lasing spectra of DCM-LC matrix filled with PSS solution in F-P cavity. Pump energy density~595 μJ/mm^2.

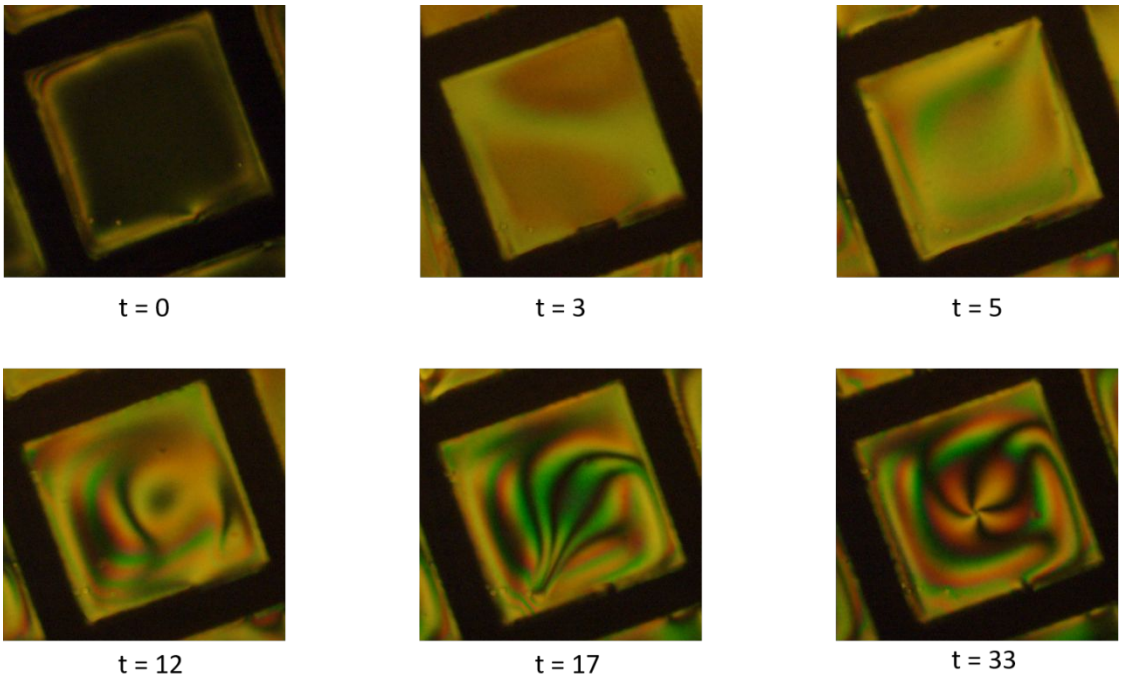

Figure S3. The polarization of LC grid starts to change after injecting sDNA solution. The birefringent regions spread out the whole grid. The CCD images were collected by CCD at a different time (s). The alignment changes upon polyanionic polymer absorption on the OTAB monolayer. The main contribution of the surfactant is to attract sufficient anionic molecules to the LC surface through electrostatic interaction. Poly(sodium 4-styrenesulfonate) (PSS), with polyanionic chain and the hydrophobic moieties, was selected as the polymer to stimulate the birefringence changes of liquid crystals matrix. The birefringence can be obtained by the equation:

$$\Delta n = \frac{1}{\theta_{bulk} - \theta_{OTAB}} \int_{\theta_{OTAB}}^{\theta a} \frac{n_e n_o d\theta}{\sqrt{n_e^2 \cos^2\theta + n_o^2 \sin^2\theta}} - n_o$$

where θ_{bulk} is the LC director zenithal tilted angle below the OTAB layer, θ_{OTAB} is the induced LC tilted angle by OTAB, n_e is the extraordinary refractive index, n_o is the ordinary refractive index. The intermediate LC orientation alters the polarization angles between the LC molecular-axis and the pump polarization, thus changing the effective refractive index of the LC matrixes. During the reorientation, the rotational viscosity of the LCs and the lateral interactions between molecules determines the response time from homeotropic to planar.

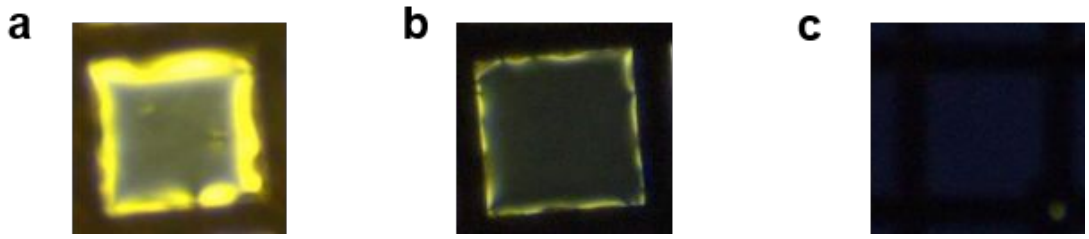

Figure S4. The appearance of the LC grid with (a) an insufficient surfactant; (b) minimum and sufficient surfactant coverage; (c) too much surfactant. The whole grid turns black after too much surfactant applied. Grid length = 200 μm.

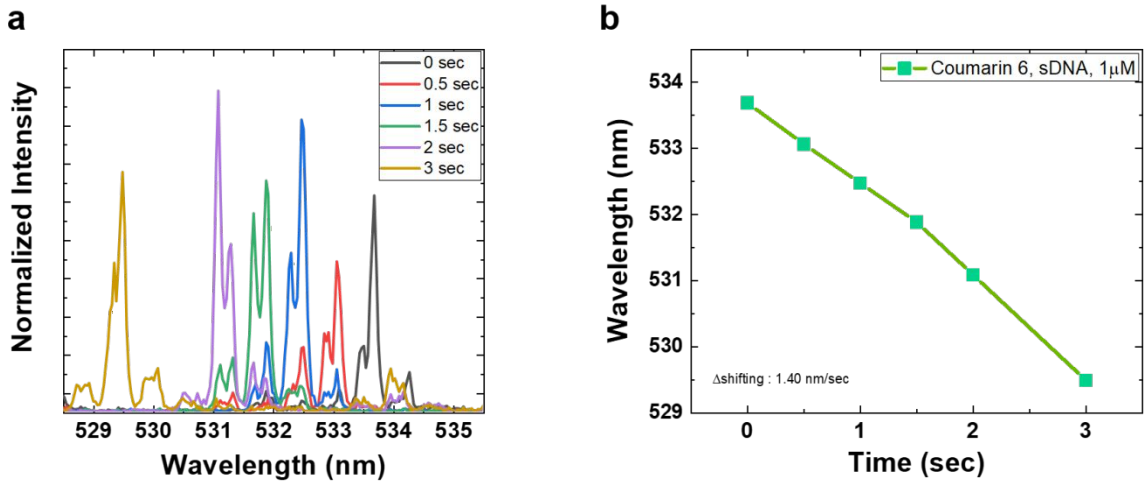

Figure S5. (a) Similar shifting behavior is observed in the Coumarin 6-doped LC matrix after injecting sDNA solution. (b) Time trajectories of laser peak shifting when LC interacts with 1 μM sDNA solution. The calculated average shifting rate is 1.40 nm/sec.

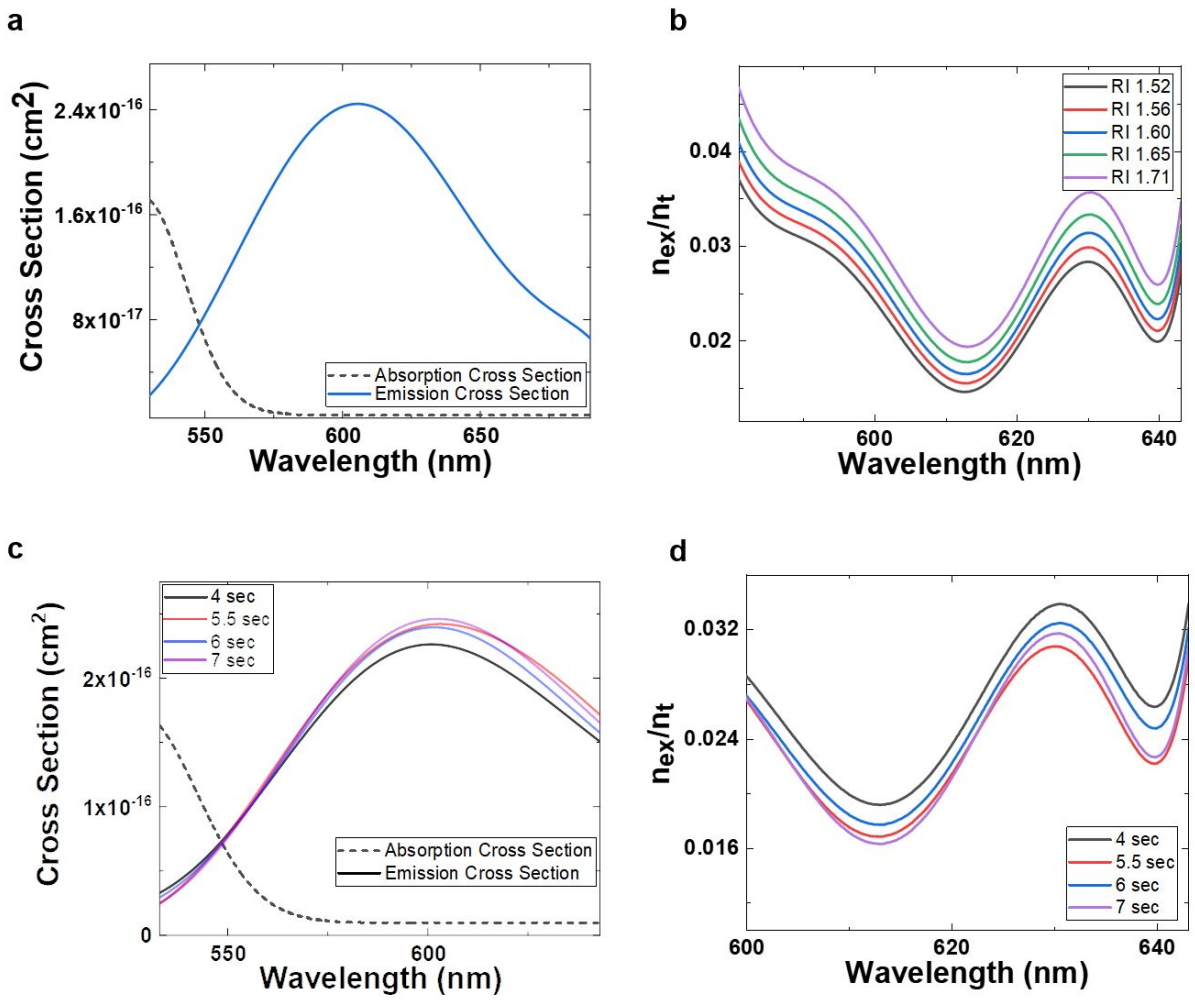

Figure S6. (a) Absorption and emission cross section of DCM-LC solution measured under static condition. (b) Fraction of DCM molecules in the excited states required at lasing threshold based on various effective refractive indices (theoretical calculation). The absorption and emission cross sections were based on the data in (a). (c) Emission cross sections measured at different time after applying sDNA molecules in the DCM-LC matrix. (d) Fraction of DCM molecules in the excited states required at lasing threshold based on different emission cross sections in (c) after applying sDNA. Note that the absorption cross section (dashed curves) and effective refractive index (n=1.6) was fixed for calculating the results in (d).

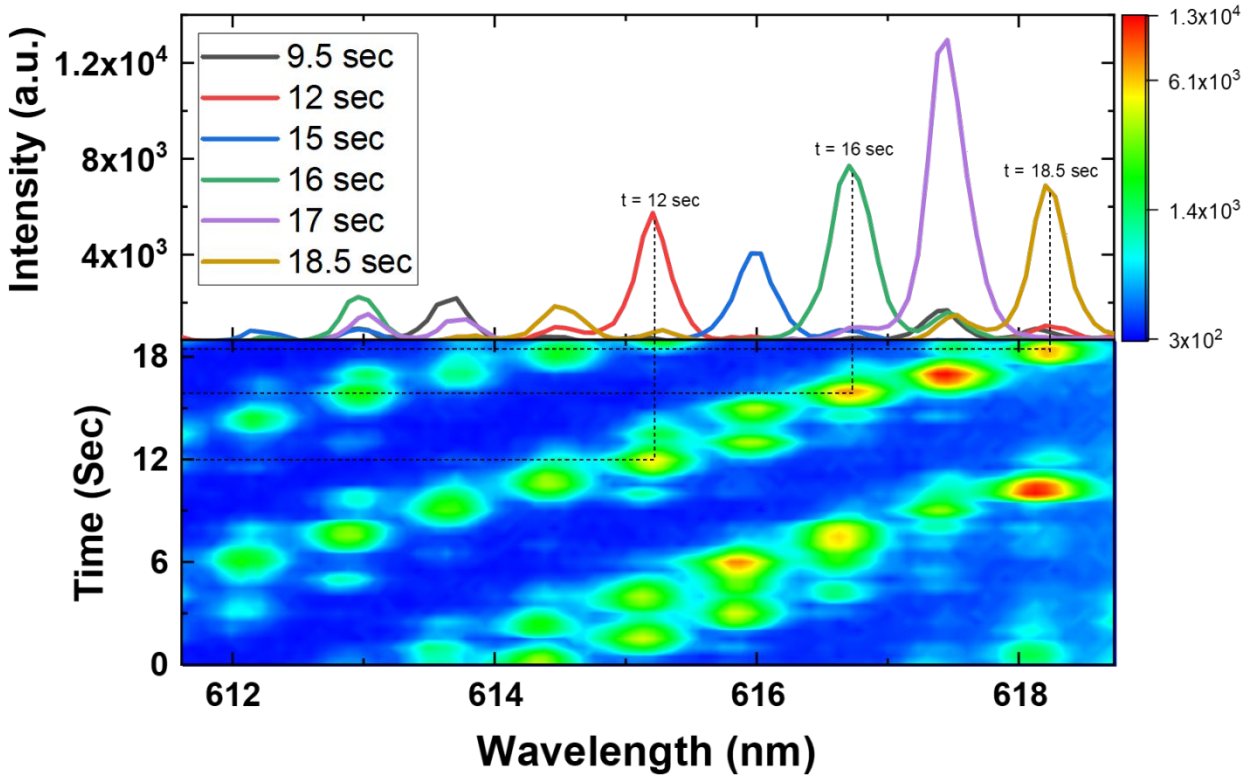

Figure S7. The top panel shows the redshift behaviour of lasing wavelength after infiltration of cDNA in sDNA chip (DNA hybridization). The wavelength shifts periodically for a certain time and vanishes after liquid crystals re-orientation stops. Several cycles can be clearly observed from the bottom panel. The colour bar represents the intensity of lasing emission.

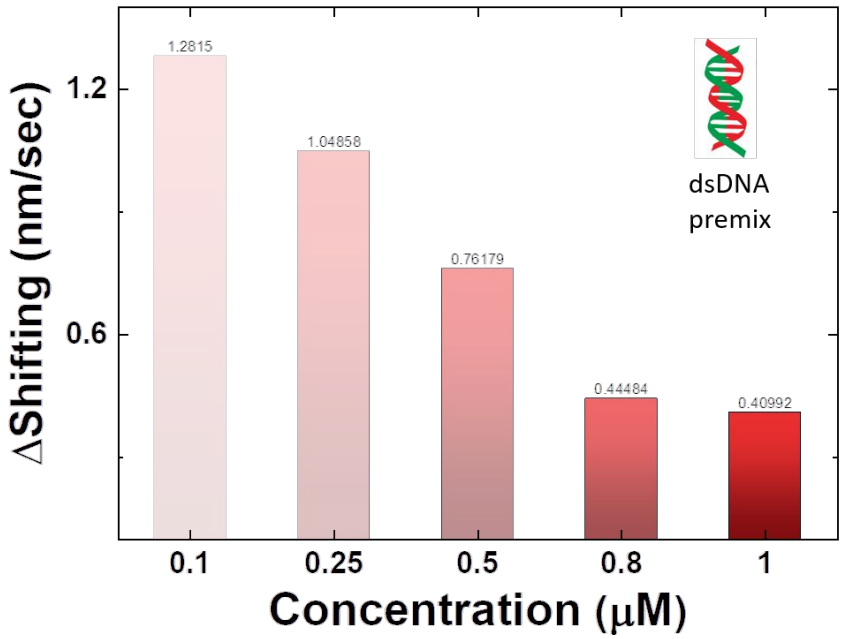

Figure S8. Summary of the average lasing wavelength shifting for pre-annealed dsDNA. (sDNA and cDNA were pre-mixed before applied to the LC matrix). The higher the dsDNA concentration, the slower the shifting rate it induces.

Calculation of required population inversion (threshold)

Based on the absorption and emission spectra of DCM, the derived absorption cross-section and emission cross-section can be derived. In particular, the absorption cross-section ($\sigma_a(\lambda)$) of DCM can be calculated by

$$\sigma_a(\lambda) = 3.8\times10^{-21}\varepsilon(\lambda)$$

where ε is the extinction coefficient and is derived from the absorbance (D). $D = [DCM]\varepsilon L$ where $[DCM]$ is the concentration of DCM and L is the optical path of F-P cavity.

The emission cross-section of a laser transition ($\sigma_e(\lambda)$) can be derived from the fluorescence spectrum and the quantum yield:

$$\sigma_e(\lambda) = \frac{\lambda^4 E(\lambda)}{8\pi c n^2 \tau_F}$$

where $E(\lambda)$ is the fluorescence quantum distribution of DCM. It is proportional to the fluorescence intensity but normalized to the quantum yield, q , that is $\int E(\lambda)\cdot d\lambda = q$. τ_F is the fluorescence lifetime. DCM is a charge transfer dye with a cylindrical molecular structure. Its fluorescence lifetime and quantum yield highly depend on the polarity of the solvent. In highly polar solvent like liquid crystal, the values of fluorescence lifetime and quantum yield are approximately 2.2 ns and 0.81. c is the speed of light in vacuum and n is the medium effective refractive index ($n \approx 1.60$ for 5CB). As a result, both the absorption cross section and emission cross section of DCM are both plotted in the above-mentioned **Supplementary Figure S6a**.

To illustrate the laser switching behavior, we perform a theoretical analysis of the DCM population inversion. we carried out the laser rate equations based on DCM molecules. Under steady state conditions, which corresponds to the lasing threshold,

$$n_{ex}\sigma_e(\lambda_L) = (n_t - n_{ex})\sigma_a(\lambda_L) + \frac{2\pi n}{\lambda_L \eta Q_o}$$

By re-arranging the equation, the fractional DCM at the excited state can be expressed by,

$$\frac{n_{ex}}{n_t} = \frac{1}{\sigma_e(\lambda) + \sigma_a(\lambda)}[\sigma_a(\lambda) + \frac{2\pi n}{n_{ex}\lambda\eta Q_o}]$$

Here n_t is the total concentration of DCM and n_{ex} is the concentration of DCM in the excited state. η is the fraction of the light (~10%) and Q_o is the average Q-factor in F-P cavity (~$5*10^4$).