Supplementary Information

Supporting Note 1

For calculating the lasing wavelength at different A/D ratio on the interfacial laser, the laser rate equations based on the donor and acceptor molecules are carried out in Eqn. (S1):

$$\frac{dq}{dt} = \frac{n_a cq}{m}\sigma_{ea}(\lambda) - \frac{(N_a - n_a)cq}{m}\sigma_{aa}(\lambda) + \frac{n_d cq}{m}\sigma_{ed}(\lambda) - \frac{(N_a - n_a)cq}{m}\sigma_{ad}(\lambda) - \frac{q}{\tau_{cavity}}, \quad \text{(S1)}$$

Herein, we consider the total concentration of molecules and total excited state molecules for simply:

$$\frac{dq}{dt} = \frac{ncq}{m}\sigma_e(\lambda) - \frac{(N - n)cq}{m}\sigma_a(\lambda) - \frac{q}{\tau_{cavity}}, \quad \text{(S2)}$$

where $N = N_a + N_d$ is the total concentration of molecules and $n = n_a + n_d$ is the concentration of excited state acceptor molecules. $\sigma_e(\lambda)$ and $\sigma_a(\lambda)$ are the emission and absorption cross-section of the acceptor/donor system. q is the emitted lasing photon number density in the cavity. τ_{cavity} is the lifetime of the photons in the cavity which equals to $Q\lambda/2\pi c$. m and c represent the refractive index of the surrounding medium and light speed in the vacuum. Under steady-state conditions, we can obtain Eqn. (S3):

$$n\sigma_e(\lambda) - (N - n)\sigma_a(\lambda) - \frac{2\pi m}{\lambda\eta Q} = 0, \quad \text{(S3)}$$

For the acceptor/donor system, the emission and absorption cross-section can be describes as Eqn. (S4) and (S5):

$$\sigma_e(\lambda) = \frac{N_a\sigma_{ea}(\lambda) + N_d\sigma_{ed}(\lambda)}{N_a + N_d}, \quad \text{(S4)}$$

$$\sigma_a(\lambda) = \frac{N_a\sigma_{aa}(\lambda) + N_d\sigma_{ad}(\lambda)}{N_a + N_d}, \quad \text{(S5)}$$

From Eqn. (S3), we can derive the fraction of molecules at the excited state in the lasing threshold condition:

$$\gamma_{th} = \frac{n}{N} = \frac{1}{\sigma_e(\lambda) + \sigma_a(\lambda)}\left[\sigma_a(\lambda) + \frac{2\pi m}{N_a\lambda Q}\right], \quad \text{(S6)}$$

According to Eqn. (S6), different C6 donor concentrations (A/D ratios) will give rise to γ_{th} values at respective wavelengths. Fig. S2b plots the γ_{th} values for various A/D ratios based on Eqn. (S6). According to the simulation results, the wavelength corresponding to the minimum fractional acceptor molecules blue shifts as the donor concentration increases (A/D ratio decreases).

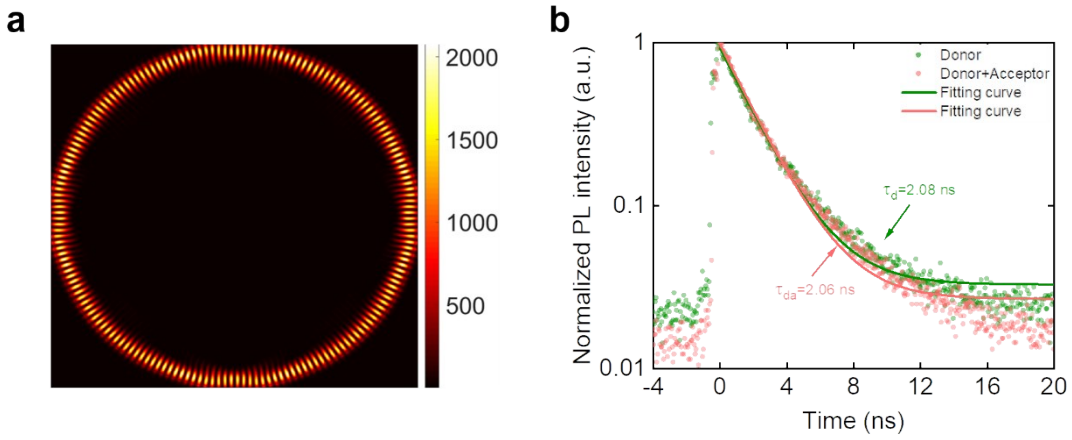

Figure S1. (a) Simulation of 10 μm diameter WGM resonators displaying the electric field distribution. **(b)** Time-resolved fluorescence lifetime measurement of donor microdroplet before and after adding acceptor solution (A/D ratio = 0.01). The FRET efficiency is around 1.5% ($E_f = 1-\tau_{da}/\tau_d$). The solid curves were fitted by the scattered dots according to the exponential decay functions.

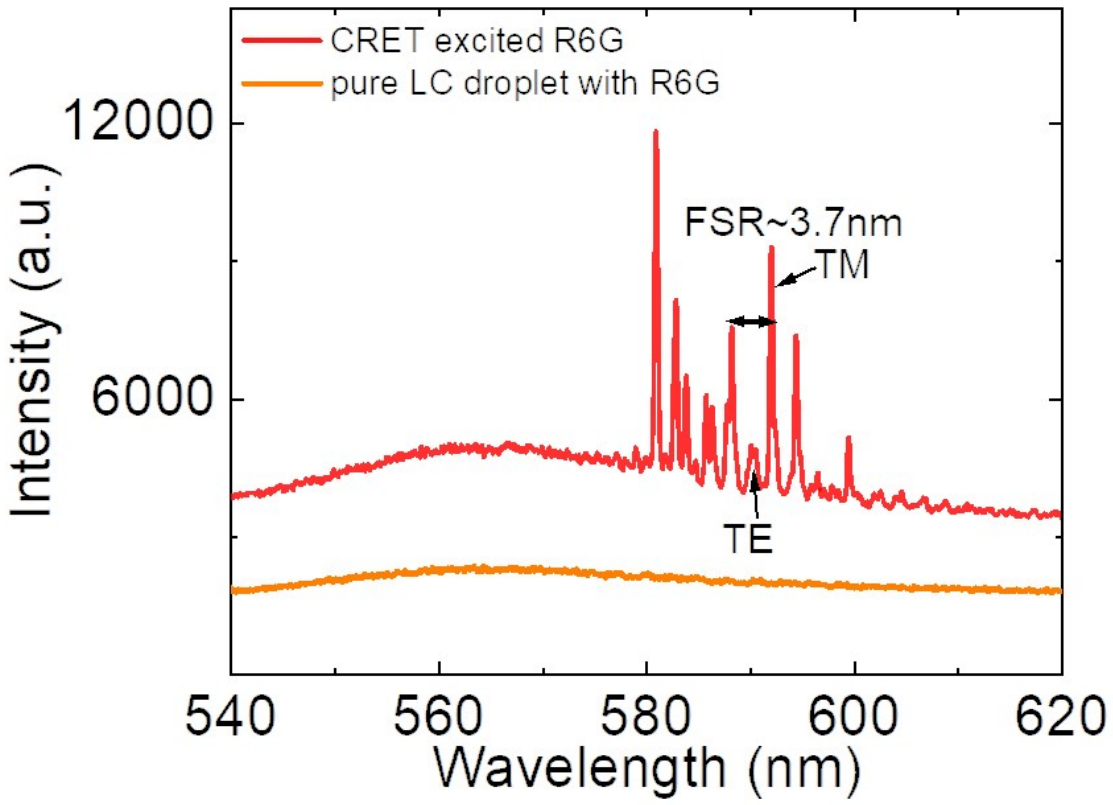

Figure S2. Laser and fluorescence emission spectra of a pure microdroplet (i.e., no dyes inside) and C6-doped microdroplet immersed in 0.1 mM R6G solution under the condition of the same pump energy and same excitation wavelength. (excitation wavelength: 450 nm). TE and TM modes are clearly labeled within the spectrum. Free spectral range (FSR) is around 3.7nm. Diameter of droplet= 24 μm.

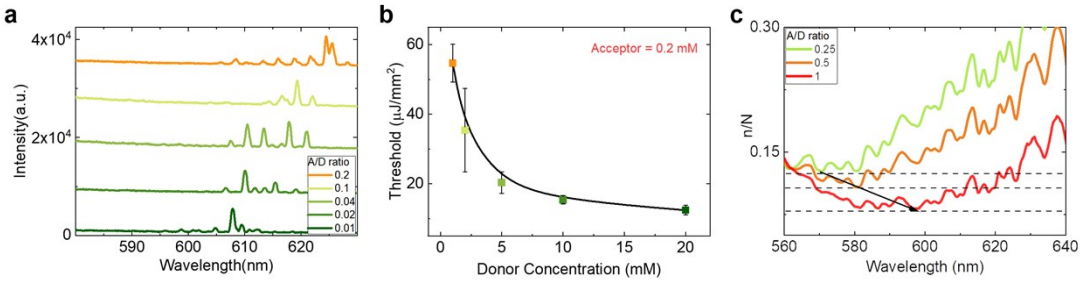

Figure S3. (a) Lasing spectra of various concentration C6 (donor) microdroplet immersed in 0.2 mM R6G solution (acceptor), excitation wavelength: 450 nm. **(b)** Lasing threshold values as a function of concentration of the donor concentration inside the microdroplet. **(c)** The fraction of molecules in the first excited states needed at the laser threshold for various representative C6 concentrations based on Eqn (S6). As the A/D ratio increases (donor concentration decreases), the wavelength which requires minimum excited molecules fraction appears red shifts. Acceptor concentration = 0.5 mM.

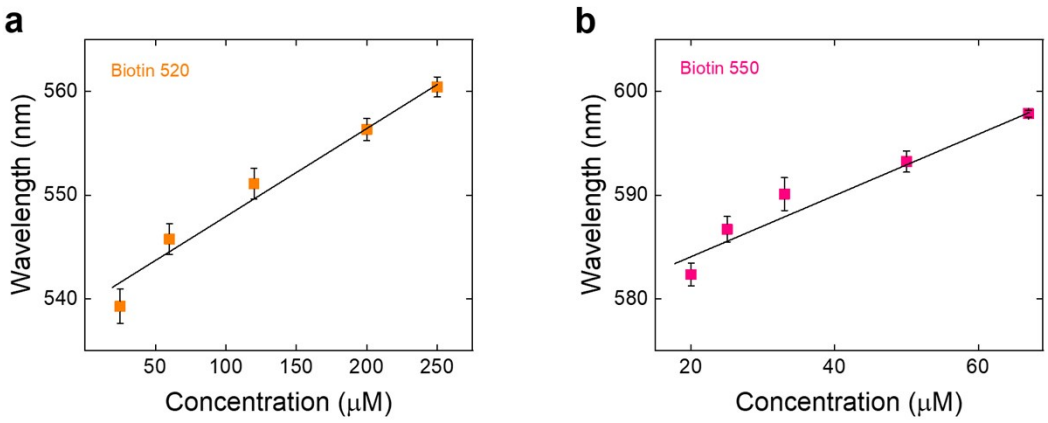

Figure S4. (a) Lasing output wavelength as a function of various concentrations of Biotin 520 (A/D ratio) which binding to C6 microdroplet surface streptavidin (SA) extracted from the spectra in **Fig. 3c**, the solid line is the linear fit. **(b)** Lasing output wavelength as a function of various concentrations of Biotin 550 (A/D ratio) which binding to C6 microdroplet surface Streptavidin-AlexaFluor 514 (SA-AF514) extracted from the spectra in **Fig. 3e**, the solid line is the linear fit.

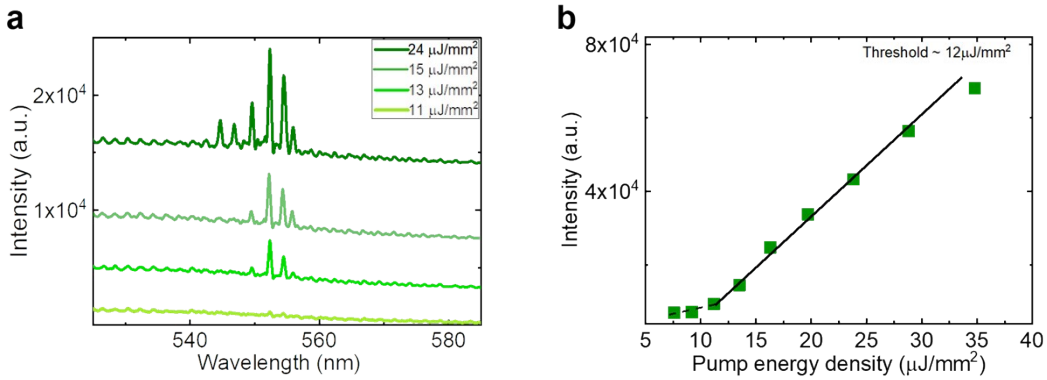

Figure S5. (a) Lasing spectra of 120-μM Biotin520 binding to 20-mM C6 microdroplet (A/D ratio = 0.006) with relatively low pump energy densities; the curves have been vertically shifted for clarity. **(b)** Spectrally integrated Biotin520 laser output as a function of pump energy density extracted from the spectra shown in **(a)**, where the wavelength range if 545-560 nm, and the solid line denotes a linear fit; the lasing approximately 12 μJ/mm^2.

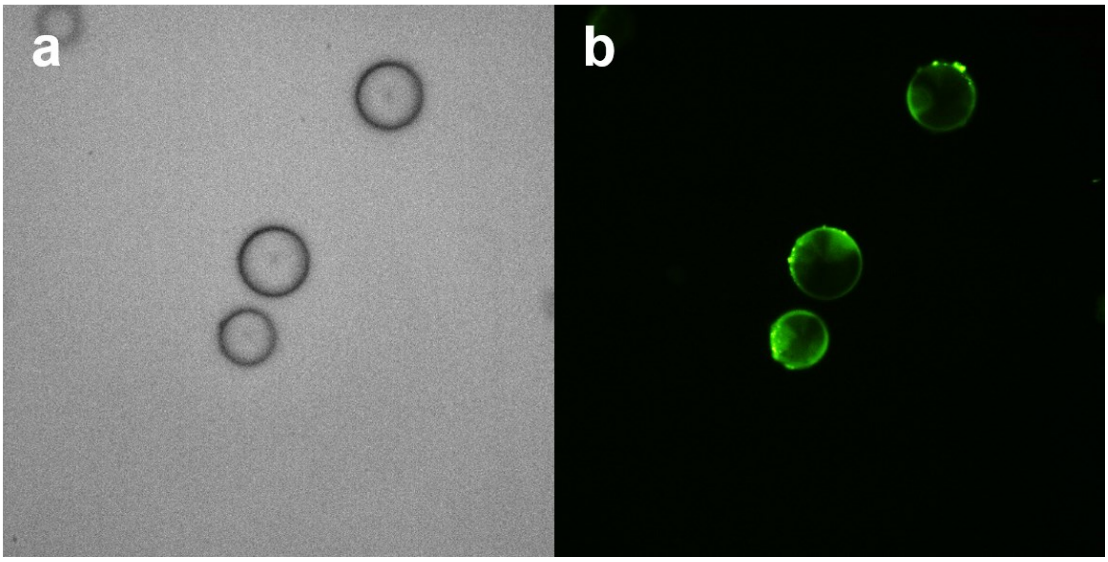

Figure S6. (a) Bright-field image of plain LC microdroplets. **(b)** Fluorescence image showing binding of SA-AF514 on the plain LC microdroplets surface.

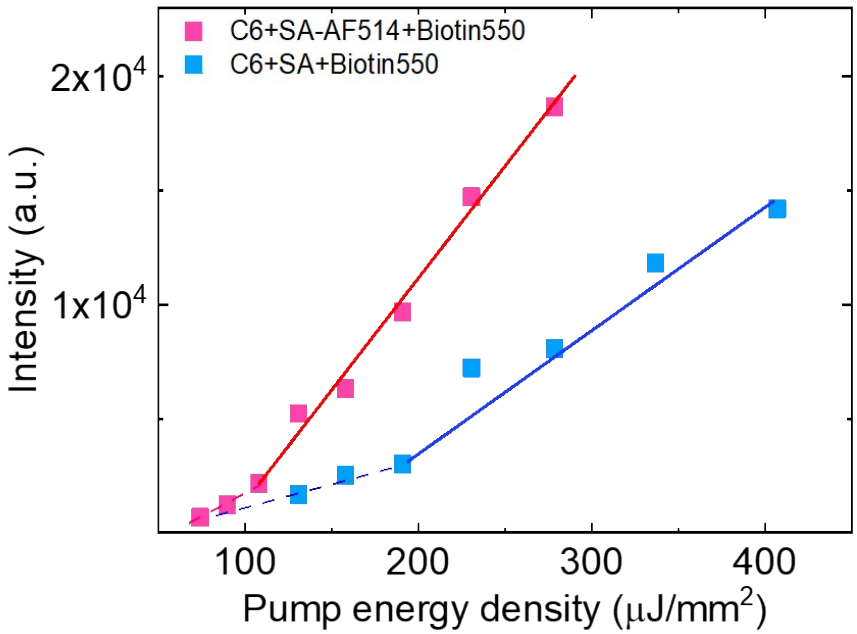

Figure S7. Spectrally integrated FRET laser output as a function of pump energy density. Pink line: 450 nm excitation for SA-AF514 coated microdroplet binding with Biotin 550. Blue line: 450 nm excitation for pure SA coated microdroplet binding with Biotin 550. The solid lines show linear fits.

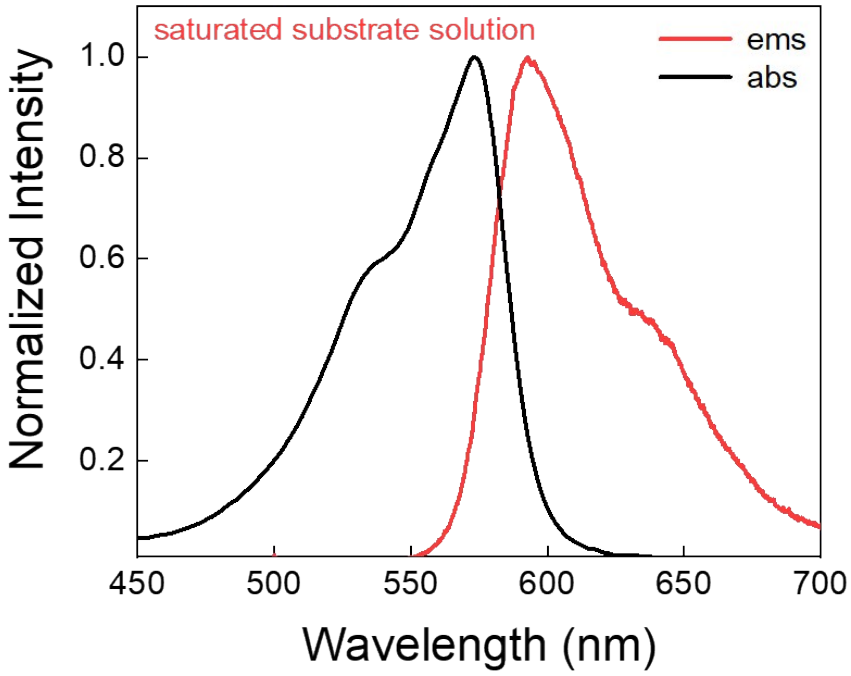

Figure S8. Normalized absorption and emission spectra of saturated substrate solution.

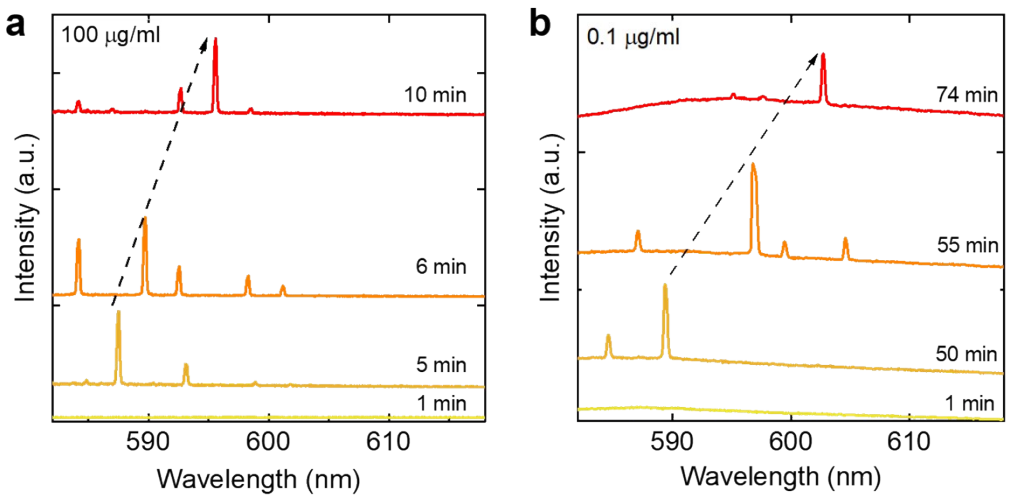

Figure S9. Lasing spectra of Bodipy microdroplet with various enzyme-substrate reaction time based on the identical droplet. **(a)** 100 μg/ml. **(b)** 0.1 μg/ml.

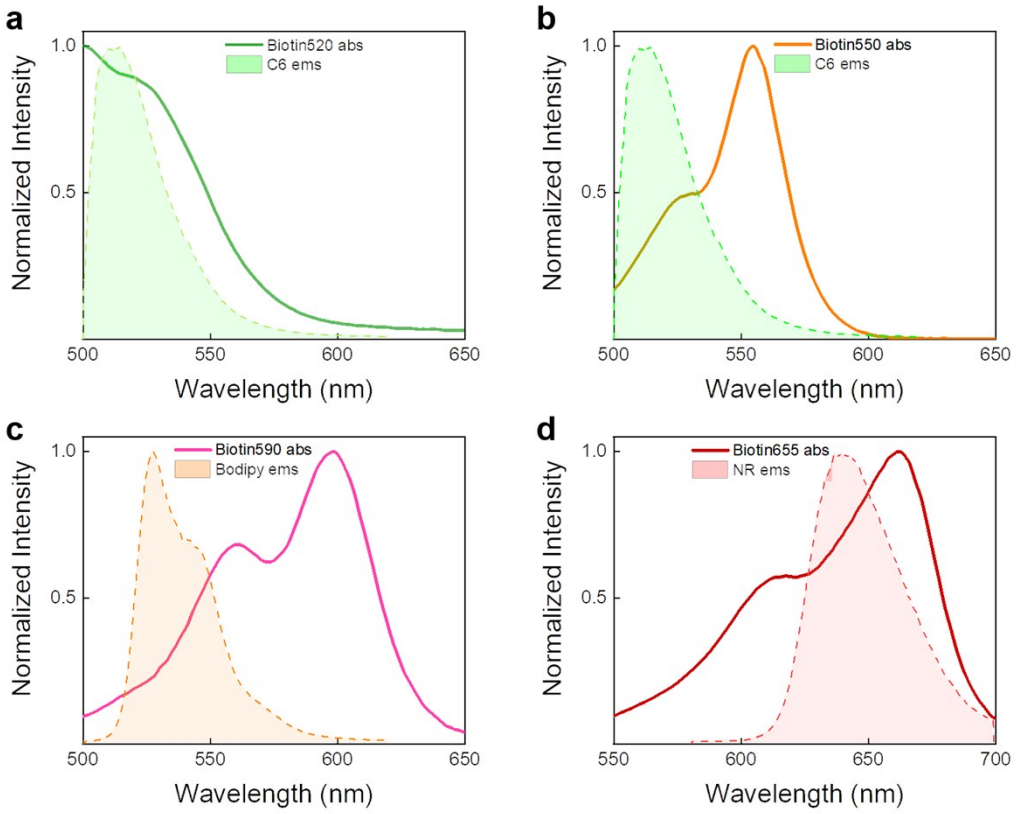

Figure S10. Normalized emission spectra of the respective donor in liquid crystal and absorption spectra of corresponding acceptor molecules in water. **(a)** Coumarin 6 (C6) droplet with Biotin 520 molecule; **(b)** C6 droplet with Biotin 550 molecule; **(c)** Bodipy droplet with Biotin 590 molecule; **(d)** Nile Red (NR) droplet with Biotin 655 molecule.

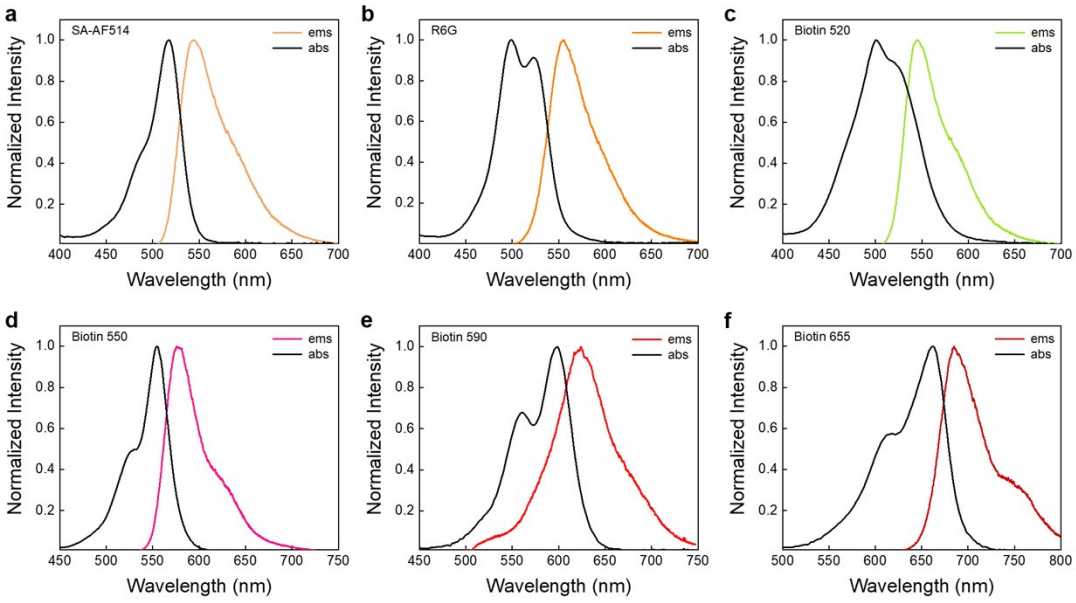

Figure S11. Normalized absorption and emission spectra of **(a)** SA-AF514; **(b)** R6G; **(c)** Biotin 520; **(d)** Biotin 550; **(e)** Biotin 590; **(f)** Biotin 655.

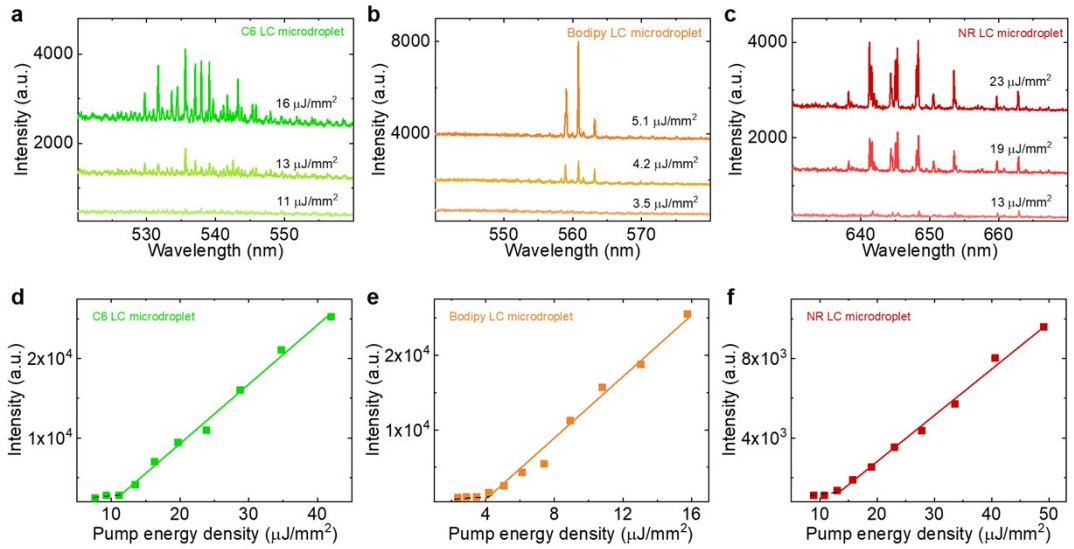

Figure S12. (a-c) Lasing spectra of C6, Bodipy and NR LC microdroplet, with relatively low pump energy densities (excitation wavelength: 450 nm, 488 nm and 532 nm). **(d-f)** Spectrally integrated R6G laser output as a function of pump energy density extracted from the spectra shown in **(a-c)**.