Supplementary information

Ultra-dense plasmonic nanogap arrays for reorientable molecular fluorescence enhancement and spectrum reshaping

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Figure S1. (a) Schematic showing the template-assisted deposition and angles θ and ϕ as control knobs for manipulating intra- and inter-cell nanogaps. (b) Schematic showing the processes flow of large-scale nanogap arrays featuring symmetric and asymmetric pair designs for tailored intra-cell and inter-cell couplings in a cross-sectional view.

Text S1. Estimating enhancement of excitation rates

3D numerical simulations of the nanogap arrays were implemented at the excitation wavelength of 405 nm following the experimental configuration. A linearly-polarized plane wave source at the far-field was adopted to mimic the excitation light. The incidence angle is perpendicular to the substrate. In order to extract the enhancement of excitation rates, two rounds of simulations with and without the existence of plasmonic nanoarrays are performed.

We adopt a pump laser wavelength of 405 nm during the experiment, which is spectrally detuned away from the LSPR band. The maximum enhancement ratio of excitation rate $\gamma_{ex}/\gamma_{ex}^{0}$ can be estimated based on numerically simulated local electric field distributions in Figure S2. Compared with the maximum electric field amplitude at the hotspot for two wavelengths of 405 nm and 550 nm (close to the absorption peak of the cyanine dye). One can see a ~3-fold difference of $|E_{max}|^2/|E_0|^2$ between the two cases for the transverse mode at the intra-cell gaps (Figure S2a) and also the longitudinal mode at the inter-cell gaps (Figure S2b). Therefore, spectrally aligning the pump laser wavelength with the resonance band will be an efficient way to further improve the enhancement factor.



Figure S2. Simulated electric field amplitude distributions with linear-polarized excitation. (a) Symmetric pair design (dy = 0 nm) with transversely-polarized excitation at 405 nm (left) and 550 nm (right). (b) asymmetric pair design (dy = 18 nm) with longitudinally-polarized excitation at 405 nm (left) and 550 nm (right).



Figure S3. (a) Measured mode wavelengths λ_L and λ_T as a function of temperature using a chip a nanogap array with asymmetric pair design. (b) Measured PL intensity as a function of

temperature.