

Supplementary Information: White-light driven photoluminescence from a few nanoplatelets in a plasmonic nanogap

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S1. Materials and Methods

Cadmium nitrate tetrahydrate ($\text{Cd}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$, 99.997%, trace metals basis), sodium myristate (99%), cadmium acetate dihydrate ($\text{Cd}(\text{OAc})_2 \cdot 2\text{H}_2\text{O}$, 98%), selenium (Se, 99.99%, trace metals basis), zinc acetate ($\text{Zn}(\text{OAc})_2$, 99.99%, trace metals basis), 1-octanethiol (98.5%), 1-octadecene (ODE, technical-grade), oleic acid (OA, 90%), and oleylamine (OAm, 70%), were purchased from Sigma-Aldrich. Methanol, hexane, toluene, and ethanol were obtained from Merck Millipore and used without any further purification.

Synthesis of Cadmium Myristate

Cadmium myristate ($\text{Cd}(\text{Myr})_2$) was synthesized according to the previous methods in the literature.¹ In a typical synthesis, cadmium nitrate tetrahydrate of 1.23 g and sodium myristate of 3.13 g were dissolved in 40 and 250 mL of methanol, respectively. After both the chemicals had completely dissolved, the solutions were mixed and stirred for 1 hour, which resulted in the $\text{Cd}(\text{Myr})_2$ white precipitate. The white precipitate was centrifuged, redispersed in methanol, and washed to remove the unreacted precursors or impurities, and the same steps were repeated thrice. Finally, the residue was kept under a vacuum overnight at 50 °C and the completely dried white powder was ground and used further.

Synthesis of 4 Monolayer (ML) CdSe Core Nanoplatelets (NPLs)

Synthesis of the CdSe 4ML core NPLs procedure was adopted from our previously reported protocol with slight modifications.² First, in a 50 mL round-bottomed flask 170 mg of Cd(Myrr)₂ and 12 mg of Selenium in 15 mL of ODE were taken and degassed for 1 hour at 95 °C. Then under N₂ gas temperature was set to 240 °C. At approximately 198 °C where the color of the solution changes to an orangish color, 80 mg of Cd(OAc)₂ · 2H₂O was swiftly added. The reaction mixture was kept at 240 °C for 10 minutes the reaction was quenched with a water bath and cooled to room temperature. The growth was terminated by adding 1 mL of OA at 150 °C and 10 mL of hexane at 60 °C. The resulting 4 ML core CdSe NPLs were precipitated by adding 5 mL of ethanol as antisolvent and centrifuged at 4000 rpm for 10 minutes. Using the size-selective precipitation, the monodispersed 4 ML core CdSe NPLs were achieved and redispersed in hexane and stored for further use.

Synthesis of CdSe/CdZnS Core/shell NPLs

Synthesis of CdSe/CdZnS core/shell NPLs was adopted from the previous report from our group with few modifications.² In a 50 mL three-neck round-bottomed flask, 0.1 mmol (26.5 mg) of Cd(OAc)₂ · 2H₂O, 0.3 mmol (55 mg) of Zn(OAc)₂, 1 mL of OA, and 10 mL of ODE were degassed for 30 minutes at 100 °C. Later the temperature was increased to 200 °C by switching the atmosphere to N₂ gas and maintained for 30 minutes and the solution was clear. The temperature was decreased to 60 °C and 1 mL of 4ML CdSe Core NPLs (having optical density: 2 at 350 nm for 100 μL core NPLs in 3 mL of hexane) were introduced. The solution mixture was degassed for 45 minutes to remove the hexane residuals. After that, N₂ gas was purged in the flask, and the

temperature was set at 300°C and 1 mL of OAm was added at 90°C. 4 mL of 0.1 M octanethiol (in ODE) was used as an S-source which starts injecting at 165°C with a syringe pump at 10 mL/h and after 240°C the injection rate was decreased to 5 mL/h. After completion of the injection, the reaction mixture was kept for another 45 minutes for annealing to complete the shell growth. The reaction was quenched using a water bath and at 60°C 10 mL hexane was added. The NPLs were washed twice using ethanol as an antisolvent, centrifuged at 6000 rpm for 5 minutes, and redispersed in 5 mL hexane for further use.

Self-assembly of CdSe/CdZnS core/shell NPLs monolayer

We improved the liquid-air interface self-assembly method previously developed by our group to create monolayer films.^{3,4} In this process, a substrate was placed into an 8 mm diameter Teflon container filled with acetonitrile (ACN) subphase. Subsequently, a 10 μ L hexane solution of NPLs was dropped onto the ACN from the edge of the container. Upon contact with the ACN, the NPLs dispersed across the surface of the subphase. After the hexane evaporated, a uniform, face-down NPLs film was obtained on top of the subphase. To transfer this film onto the substrate, a peristaltic pump was used to remove the ACN from the bottom of the container, allowing the film to settle onto the substrate. Note that to achieve a uniform film, the height of the ACN above the substrate should be as low as possible to minimize movement of the assembled film during drainage. Additionally, we used a silicone oil surfactant to compress the monolayer film further, preventing cracks and voids due to capillary forces from the container walls during deposition.

Characterization methods for colloidal NPLs

Optical Characterization

Absorbance measurements of the NPLs were recorded using the Shimadzu UV-1800 spectrophotometer and PL measurements were recorded by a Shimadzu RF- 5301 PC Spectrofluorophotometer. Our PL-QY measurement setup was equipped with an Ocean Optics S4000 spectrometer, and an integrating sphere was used. For the PL-QY measurements, a clean glass substrate was used for reference, and NPLs solutions were excited at a wavelength of 405 nm (Cobolt Laser).

Transmission electron microscope (TEM)

A single drop of diluted NPLs solution is drop casted on top of a copper grid covered with an amorphous carbon film. The TEM images of the NPLs were obtained using a JEOL 2100F TEM at 200 KV.

Atomic Force Microscopy (AFM)

To confirm the self-assembled NPLs monolayer thin film AFM measurement was performed using the scan-assist mode with 512 resolutions by a Bruker Dimension Icon scanning probe microscope (Bruker 18 Co., Germany).

Scanning Electron Microscope (SEM)

SEM images were obtained using a JSM-7600F (JEOL) instrument at an accelerating voltage of 2 kV in secondary electron detection mode.

Thermogravimetric Analysis (TGA)

TGA analysis was performed using an SDT Q600 instrument under ambient conditions with a powder sample.

S2. Simulation Methods

In this section, we detail the derivation of how the three QNMs (α , β_1 , and β_2) are identified and how their coupling rates are extracted from the scattering cross-section (SCS). We begin with full-wave simulations and establish the connection between classical and quantum models through QNM analysis and a non-Hermitian scattering framework.

Full-wave simulation

Full-wave simulations solving the three-dimensional Maxwell's equations are performed using the finite element method (FEM) within COMSOL Multiphysics (RF module).

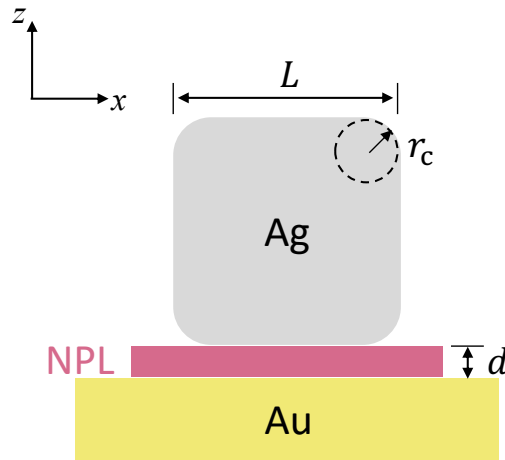


Figure S1: Geometry of the NPoM used in full-wave simulations.

The geometry of the NPoM used in the full-wave simulations is shown in Fig. S1. The cube has a side length of $L = 75$ nm and a corner radius of $r_c = 5$ nm. The gap thickness is set to $d = 7$ nm and is filled by the NPL, which is modeled as a lossless dielectric with a constant refractive index of $n = 1.952$. The permittivity of silver is modeled using the Drude model:

$$\epsilon(\omega) = \epsilon_{\infty} \left(1 - \frac{\omega_p^2}{\omega^2 + i\omega\Gamma} \right), \quad (\text{S1})$$

where $\epsilon_{\infty} = 1$, $\omega_p = 1.37 \times 10^{16}$ rad/s, and $\Gamma = 0.0023\omega_p$ ⁵, and the permittivity of gold is described by a Drude-Lorentz model:

$$\epsilon(\omega) = \epsilon_{\infty} \left(1 - \frac{\omega_{p,1}^2}{\omega^2 - \omega_{0,1}^2 + i\omega\Gamma_1} - \frac{\omega_{p,2}^2}{\omega^2 - \omega_{0,2}^2 + i\omega\Gamma_2} \right), \quad (\text{S2})$$

with parameters: $\epsilon_{\infty} = 6$, $\omega_{p,1} = 5.37 \times 10^{15}$ rad/s, $\omega_{0,1} = 0$ and $\Gamma_1 = 6.22 \times 10^{13}$ rad/s; $\omega_{p,2} = 2.26 \times 10^{15}$ rad/s, $\omega_{0,2} = 4.57 \times 10^{15}$ rad/s and $\Gamma_2 = 1.33 \times 10^{15}$ rad/s. The surrounding vacuum is treated as a lossless medium with a constant refractive index of $n = 1$.

To model the scattering, the NPoM structure is excited by a plane wave representing white light, with electric field components in the ratio $E_x : E_y : E_z = 1 : 1 : \sqrt{2}$ and an amplitude of $|E_0| = 1$ V/m. This configuration effectively excites bright QNMs with radiative decay channels—such as α modes—within the energy range of 1.50–2.25 eV. A Perfectly Matched Layer (PML) boundary condition is applied to suppress back reflections. The scattering power P_{sc} is obtained by integrating the power flux around the scatterer, and the scattering cross-section (SCS) is calculated using $P_{\text{sc}}/I_{\text{inc}} = P_{\text{sc}} \cdot (2Z)/|E_0|^2$, where I_{inc} represents the intensity of the incident wave and $Z = \sqrt{\mu\mu_0/(\epsilon\epsilon_0)}$ is the impedance of the medium. Note that tunneling effects⁶ and nonlocal effects⁷ are neglected throughout the simulations.

To model the PL, we use the same geometric configuration as in the scattering simulations. Instead of a plane wave, the PL is simulated as the emission from an excited quantum emitter (QE) positioned at the corner of the gap region (as shown in the inset of Fig. 3(a)). An electric dipole source is introduced, oscillating in the in-plane direction with component ratios $|\vec{\mu}_x| : |\vec{\mu}_y| = 1 : 1$. The dipole moment is set to $\mu = 4.8 \times 10^{-29}$ C·m, based on experimental measurements. The PL signal is obtained by integrating the radiated power flux in the far-field surrounding the NPoM.

Quasinormal modes analysis

Quasinormal modes (QNMs) are the intrinsic resonant modes of a system, defined without the need for external excitation ⁵. In nonmagnetic systems, QNMs are obtained by solving the eigenvalue problem of the time-harmonic, source-free Maxwell's equations:

$$\begin{bmatrix} 0 & -i\mu_0^{-1}\nabla \times \\ i\varepsilon(\tilde{\omega}_j) & 0 \end{bmatrix} \begin{bmatrix} \tilde{\mathbf{H}}_j \\ \tilde{\mathbf{E}}_j \end{bmatrix} = \tilde{\omega}_j \begin{bmatrix} \tilde{\mathbf{H}}_j \\ \tilde{\mathbf{E}}_j \end{bmatrix}, \quad (\text{S3})$$

where $\varepsilon(\tilde{\omega}_j)$ is the permittivity, μ_0 is the vacuum permeability, and $\tilde{\mathbf{H}}_j, \tilde{\mathbf{E}}_j$ are the QNMs' electric and magnetic fields, satisfying the Sommerfeld outgoing-wave conditions. For dispersive materials like silver and gold, this eigenvalue problem becomes nonlinear due to the frequency dependence of permittivity. To simplify it, we use the Drude model for silver and the Drude–Lorentz model for gold—consistent with our full-wave simulations.

The Drude-Lorentz poles introduce two hidden auxiliary fields to account for the intraband

transitions of free electrons:

$$\tilde{\mathbf{P}} = (\epsilon_{(Au)}(\omega) - \epsilon_{(Au),\infty}) \tilde{\mathbf{E}} \quad (\text{S4})$$

$$\tilde{\mathbf{J}} = -i\omega \tilde{\mathbf{P}}. \quad (\text{S5})$$

Here, $\tilde{\mathbf{P}}$ and $\tilde{\mathbf{J}}$ represent the auxiliary polarization and electric current, respectively. The eigenvectors are now expanded to $\tilde{\psi}_j = [\tilde{\mathbf{H}}_j, \tilde{\mathbf{E}}_j, \tilde{\mathbf{P}}_j, \tilde{\mathbf{J}}_j]^T$, and the system becomes a linear eigenproblem:

$$\tilde{H} \tilde{\psi}_j = \begin{bmatrix} 0 & -i\mu_0^{-1} \nabla \times & 0 & 0 \\ i\epsilon_\infty^{-1} \nabla \times & 0 & 0 & -i\epsilon_\infty^{-1} \\ 0 & 0 & 0 & i \\ 0 & i\omega_p^2 \epsilon_\infty & 0 & -i\Gamma \end{bmatrix} \tilde{\psi}_j = \tilde{\omega}_j \tilde{\psi}_j. \quad (\text{S6})$$

To compute the QNMs of the NPoM, we use COMSOL to define the structure and material parameters, consistent with our full-wave simulation settings. Then, we employ the open-source modal analysis toolbox “MAN”⁸, integrated with MATLAB and COMSOL Multiphysics (RF module), to determine the characteristics of the QNMs⁹.

Non-Hermitian scattering model

In a scattering problem involving n quasi-normal modes (QNMs), where $n \geq 2$, the total Hamiltonian is described as $\hat{H} = \hat{H}_0 + \hat{V}$. Here, $\hat{H}_0$ corresponds to the free Hamiltonian, while $\hat{V}$ represents the interaction potential. Both $\hat{H}_0$ and $\hat{V}$ are formulated using Fock states, and are expressed as

follows:

$$\hat{H}_0 = \sum_j^n \hbar \omega_j a_j^\dagger a_j + \sum_l \hbar \omega_l B_l^{\dagger r} B_l^r + \sum_m \hbar \omega_m D_m^{\dagger \text{nr}} D_m^{\text{nr}}, \quad (\text{S7})$$

$$\hat{V} = \sum_j^n \sum_k^n \hbar g_{jk} (a_j^\dagger a_k + a_k^\dagger a_j) \quad (\text{S8})$$

$$+ \sum_j^n \sum_l \hbar V_{jl} (B_l^{\dagger r} a_j + a_j^\dagger B_l^r) + \sum_j^n \sum_m \hbar V_{jm} (D_m^{\dagger \text{nr}} a_j + a_j^\dagger D_m^{\text{nr}}). \quad (\text{S9})$$

Here, a_j (or $a_j^\dagger$) represents the annihilation (or creation) operator for the j^{th} mode with resonance energy $\hbar \omega_j$. Similarly, the vacuum and thermal reservoirs are quantized in terms of Fock states, denoted by integers l and m , which correspond to the system's radiative and non-radiative decay channels, respectively. The operators B_l^r (or $B_l^{\dagger r}$) and D_m^{nr} (or $D_m^{\dagger \text{nr}}$) represent the annihilation (or creation) operators for the l^{th} and m^{th} states.

The first term in $\hat{V}$ describes the near-field coupling between a_j and a_k , where $j \neq k$, with the interaction characterized by the coupling rate g_{jk} ; note that $g_{jk} = 0$ when $j = k$. The second and third terms account for energy leakage through coupling to the vacuum and thermal reservoirs, governed by the channel coefficients V_{jl}^r and V_{jm}^{nr} ¹⁰.

According to Fermi's golden rule, the scattering cross section (SCS) is determined by calculating the transition rate between the system's initial (i) and final (f) states. This process involves evaluating the matrix element of the interaction responsible for the transition and integrating over the density of final states, which accounts for the number and distribution of possible final states.

The SCS is expressed as:

$$\sigma_{\text{sc}} = \frac{\sum_{i,f} W_{i \rightarrow f}}{\phi} = \frac{2\pi}{\hbar \phi} \sum_{i,f} |\mathcal{T}_{i \rightarrow f}|^2 \delta(E_f - E_i), \quad (\text{S10})$$

where $\mathcal{T}_{i \rightarrow f}$ represents the transition matrix element, which is crucial in determining the SCS. This matrix element is formulated as the product of three components, each enclosed in square brackets:

$$\mathcal{T}_{i \rightarrow f} = \langle f | \hat{V}^\dagger \hat{P}^\dagger \hat{G}(\tilde{\omega}) \hat{P} \hat{V} | i \rangle = \sum_{j,k} [\langle f | \hat{V}^\dagger | \mathbf{1}_j^a \rangle] [\langle \mathbf{1}_j^a | \hat{G}(\tilde{\omega}) | \mathbf{1}_k^a \rangle] [\langle \mathbf{1}_k^a | \hat{V} | i \rangle]. \quad (\text{S11})$$

Here, the dyadic $\hat{P} = \sum_j^n |\mathbf{1}_j^a\rangle \langle \mathbf{1}_j^a|$ is established within the system's Hilbert space to facilitate the connection between the input vector $\hat{V}|i\rangle$ and the conjugate of the output vector $[\hat{V}|f\rangle]^\dagger$ via the system's Green's function $\hat{G}(\tilde{\omega})$.

Following Ref.¹⁰, the transition matrix $\mathcal{T}_{i \rightarrow f}$ is given by:

$$\begin{aligned} \mathcal{T}_{i \rightarrow f} &= \sum_{j,k} [\langle f | \hat{V} | \mathbf{1}_j^a \rangle] \cdot \left[\frac{1}{\langle \mathbf{1}_j^a | \tilde{\omega} | \mathbf{1}_k^a \rangle - \langle \mathbf{1}_j^a | \hat{H}_0 | \mathbf{1}_k^a \rangle - \langle \mathbf{1}_j^a | \hat{V} | \mathbf{1}_k^a \rangle} \right] \cdot [\langle \mathbf{1}_k^a | \hat{V} | i \rangle] \\ &= \begin{bmatrix} V_1^r & \cdots & V_n^r \end{bmatrix} \cdot \begin{bmatrix} \tilde{\omega} - \omega_1 + \frac{i}{2}(\gamma_1^r + \gamma_1^{\text{nr}}) & \cdots & -g_{1n} + \frac{i}{2}\sqrt{\gamma_1^r \gamma_n^r} \\ \vdots & \ddots & \vdots \\ -g_{n1} + \frac{i}{2}\sqrt{\gamma_n^r \gamma_1^r} & \cdots & \tilde{\omega} - \omega_n + \frac{i}{2}(\gamma_n^r + \gamma_n^{\text{nr}}) \end{bmatrix}^{-1} \cdot \begin{bmatrix} V_1^r \\ \vdots \\ V_n^r \end{bmatrix}. \quad (\text{S12}) \end{aligned}$$

We now examine a straightforward single-mode scattering process to extract V_j^r , focusing on a QNM characterized by the complex frequency $\omega_0 - i\gamma_0^t$. The SCS for this single-mode is calculated given by:

$$\sigma_0(\omega) = \frac{1}{(\omega - \omega_0)^2 + (\gamma_0^t/2)^2} \cdot \frac{2\pi}{\phi} \sum_{i,f} |V_0^r \cdot V_0^r|^2 \delta(E_f - E_i) \quad (\text{S13})$$

$$= \frac{1}{(\omega - \omega_0)^2 + (\gamma_0^t/2)^2} \cdot \frac{2\pi}{\phi} \int d\Omega dk_f \left(\frac{L_0}{2\pi} \right)^3 k_f^2 \left[\frac{\mu_0^r \mu_0^r \omega_0^2}{2\epsilon_0 \sqrt{\omega_f \omega_i} \nu_0} \right]^2 [(\vec{e}_0 \cdot \vec{e}_f)(\vec{e}_0 \cdot \vec{e}_i)]^2 \delta(\omega_f - \omega_i). \quad (\text{S14})$$

For a single photon, the radiative flux density can be expressed as $\phi = c/L_0^3$. Assuming ideal alignment where the modal field vector $\vec{e}_0$ perfectly aligns with the excitation field vector $\vec{e}_i$ such

that $\vec{e}_0 \cdot \vec{e}_i = 1$, the SCS simplifies accordingly:

$$\sigma_0(\omega) = \frac{1}{(\omega - \omega_0)^2 + (\gamma_0^t/2)^2} \cdot \frac{1}{4\pi^2 c} \frac{(\mu_0^r \mu_0^r \omega_0^2)^2}{4\varepsilon_0^2} \int d\Omega (\vec{e}_0 \cdot \vec{e}_f)^2 dk_f \frac{k_f^2}{\omega_f^2} \quad (\text{S15})$$

$$= \frac{1}{(\omega - \omega_0)^2 + (\gamma_0^t/2)^2} \cdot \frac{(\mu_0^r \mu_0^r \omega_0^2)^2}{16\pi^2 \varepsilon_0^2 c^3} \frac{8\pi}{3} \quad (\text{S16})$$

Since the spontaneous radiative decay rate in a vacuum is given by $\gamma_0^r = \omega_0^3 (\mu_0^r)^2 / (3\pi c^3 \varepsilon_0)$, we can substitute $(\omega_0 \mu_0^r)^2 = 3\pi c^3 \varepsilon_0 \gamma_0^r / \omega_0$ into Eq. (S16):

$$\sigma_0(\omega) = \frac{3}{2\pi} \frac{1}{(\omega - \omega_0)^2 + (\gamma_0^t/2)^2} \cdot \left(\frac{\lambda_0 \gamma_0^r}{2} \right)^2 \quad (\text{S17})$$

$$= \frac{3}{2\pi} \left| \sqrt{\frac{\lambda_0 \gamma_0^r}{2}} \frac{1}{\omega - \omega_0 + i\gamma_0^t/2} \sqrt{\frac{\lambda_0 \gamma_0^r}{2}} \right|^2, \quad (\text{S18})$$

where λ_0 represents the wavelength associated with ω_0 , and $\sqrt{\lambda_0 \gamma_0^r/2}$ is identified as the dressed channel coefficient, which is derived from V_0^r .

Expanding this approach to encompass scattering processes involving any number of QNMs, the SCS is generalized and expressed as follows:

$$\sigma_{\text{sc}}(\omega) = \frac{3}{2\pi} \left| \begin{bmatrix} \sqrt{\frac{\lambda_1 \gamma_1^r}{2}} \\ \vdots \\ \sqrt{\frac{\lambda_n \gamma_n^r}{2}} \end{bmatrix}^T \cdot \begin{bmatrix} \tilde{\omega} - \omega_1 + \frac{i}{2}(\gamma_1^r + \gamma_1^{\text{nr}}) & \cdots & -g_{1n} + \frac{i}{2}\sqrt{\gamma_1^r \gamma_n^r} \\ \vdots & \ddots & \vdots \\ -g_{n1} + \frac{i}{2}\sqrt{\gamma_n^r \gamma_1^r} & \cdots & \tilde{\omega} - \omega_n + \frac{i}{2}(\gamma_n^r + \gamma_n^{\text{nr}}) \end{bmatrix}^{-1} \cdot \begin{bmatrix} \sqrt{\frac{\lambda_1 \gamma_1^r}{2}} \\ \vdots \\ \sqrt{\frac{\lambda_n \gamma_n^r}{2}} \end{bmatrix} \right|^2. \quad (\text{S19})$$

Hence, we fit the SCS with the black dashed line in **Fig. 3(c)** by:

$$\sigma_{\text{sc}}(\tilde{\omega}) = \frac{3}{2\pi} \left| \begin{bmatrix} \sqrt{\frac{\lambda_{\alpha}\gamma_{\alpha}^{\text{r}}}{2}} \\ \sqrt{\frac{\lambda_{\beta_1}\gamma_{\beta_1}^{\text{r}}}{2}} \\ \sqrt{\frac{\lambda_{\beta_2}\gamma_{\beta_2}^{\text{r}}}{2}} \end{bmatrix}^{\text{T}} \cdot \begin{bmatrix} \tilde{\omega} - \omega_{\alpha} + \frac{i}{2}(\gamma_{\alpha}^{\text{r}} + \gamma_{\alpha}^{\text{nr}}) & -g_{\alpha\beta_1} + \frac{i}{2}\sqrt{\gamma_{\alpha}^{\text{r}}\gamma_{\beta_1}^{\text{r}}} & -g_{\alpha\beta_2} + \frac{i}{2}\sqrt{\gamma_{\alpha}^{\text{r}}\gamma_{\beta_2}^{\text{r}}} \\ -g_{\alpha\beta_1} + \frac{i}{2}\sqrt{\gamma_{\beta_1}^{\text{r}}\gamma_{\alpha}^{\text{r}}} & \tilde{\omega} - \omega_{\beta_1} + \frac{i}{2}(\gamma_{\beta_1}^{\text{r}} + \gamma_{\beta_1}^{\text{nr}}) & -g_{\beta_1\beta_2} + \frac{i}{2}\sqrt{\gamma_{\beta_1}^{\text{r}}\gamma_{\beta_2}^{\text{r}}} \\ -g_{\alpha\beta_2} + \frac{i}{2}\sqrt{\gamma_{\beta_2}^{\text{r}}\gamma_{\alpha}^{\text{r}}} & -g_{\beta_1\beta_2} + \frac{i}{2}\sqrt{\gamma_{\beta_2}^{\text{r}}\gamma_{\beta_1}^{\text{r}}} & \tilde{\omega} - \omega_{\beta_2} + \frac{i}{2}(\gamma_{\beta_2}^{\text{r}} + \gamma_{\beta_2}^{\text{nr}}) \end{bmatrix}^{-1} \cdot \begin{bmatrix} \sqrt{\frac{\lambda_{\alpha}\gamma_{\alpha}^{\text{r}}}{2}} \\ \sqrt{\frac{\lambda_{\beta_1}\gamma_{\beta_1}^{\text{r}}}{2}} \\ \sqrt{\frac{\lambda_{\beta_2}\gamma_{\beta_2}^{\text{r}}}{2}} \end{bmatrix} \right|^2 \quad (\text{S20})$$

S3. Calibration of white-light irradiance and comparison to 405 nm laser excitation

We estimate the average irradiance at the sample plane under epi-dark-field white-light illumination for the settings used in this work to be $I_{\text{DF}} = 0.6\text{--}14\text{ mW cm}^{-2}$, using the calibration procedure in Eqs. (S21)-(S23). For comparison, using the same $100\times$, $\text{NA} = 0.90$ objective, a focused 405-nm CW laser with $P_{\text{laser}} = 0.9\text{ mW}$ corresponds to a peak irradiance of $I_0 \approx 3.8 \times 10^5\text{ W cm}^{-2}$ (Eq. (S25)). The following steps define how I_{mea} , the entrance-pupil geometry, and the beam radius are used to convert the measured beam power density to I_{DF} in the sample.

Estimation of excitation power densities The measured irradiance of the white-lamp excitation is $0.6\text{--}14\text{ mW cm}^{-2}$, which is much lower than that of the focused laser excitation ($\sim\text{ kW cm}^{-2}$). For comparison, we used a continuous-wave (CW) 405 nm diode laser (Thorlabs, PL205 model). Below we detail the procedures used to estimate the power densities of both light sources.

Focused white light under epi–dark-field (DF) illumination. We first measured the power density I_{mea} (W cm^{-2}) of the excitation beam using a power meter (Nova portable power meter). Under epi–bright-field (BF) illumination, the total power entering the objective can be estimated from the objective entrance-pupil area:

$$P_{\text{BF}} = I_{\text{mea}} A_{\text{pup}}, \quad A_{\text{pup}} = \pi r_{\text{pup}}^2, \quad (\text{S21})$$

where we used a $100\times$ objective with $\text{NA} = 0.90$ (Nikon CFI TU Plan Fluor BD) and an entrance-pupil radius $r_{\text{pup}} = 1.8$ mm.

In the epi–DF configuration, illumination is restricted to an annular region defined by an outer numerical aperture $\text{NA}_{\text{outer}} \approx 0.95$ and an inner numerical aperture $\text{NA}_{\text{inner}} \approx 0.91$. Because the excitation beam is uniform and larger than the objective entrance pupil, the optical power coupled into the objective scales with the corresponding pupil area. The power delivered to the sample under DF excitation can therefore be estimated as

$$P_{\text{DF}} = \frac{\text{NA}_{\text{outer}}^2 - \text{NA}_{\text{inner}}^2}{\text{NA}^2} P_{\text{BF}}. \quad (\text{S22})$$

The average incident power density at the sample plane under DF excitation was finally estimated as

$$I_{\text{DF}} = \frac{P_{\text{DF}}}{A_{\text{beam}}}, \quad A_{\text{beam}} = \pi r_{\text{beam}}^2, \quad (\text{S23})$$

where the measured beam radius is $r_{\text{beam}} \approx 75$ μm . With the measured power density of the excitation beam ranging from $I_{\text{mea}} = 11\text{--}267$ $\mu\text{W cm}^{-2}$, the corresponding focused white-light power densities are estimated to be $I_{\text{DF}} = 0.6\text{--}14$ mW cm^{-2} .

Focused 405 nm CW laser. Using the same $100\times$ NA = 0.90 objective, the light is concentrated into a diffraction-limited spot with radius

$$r_0 \approx 0.61 \frac{\lambda}{\text{NA}} \approx 0.275 \mu\text{m} \quad (\lambda = 405 \text{ nm}). \quad (\text{S24})$$

Neglecting optical losses, the full laser power $P_{\text{laser}} = 0.9 \text{ mW}$ reaches the sample plane. This results in a peak power density at the center of the focus of approximately

$$I_0 \approx \frac{P_{\text{laser}}}{\pi r_0^2} \approx 3.8 \times 10^5 \text{ W cm}^{-2}. \quad (\text{S25})$$

S4. Additional Figures

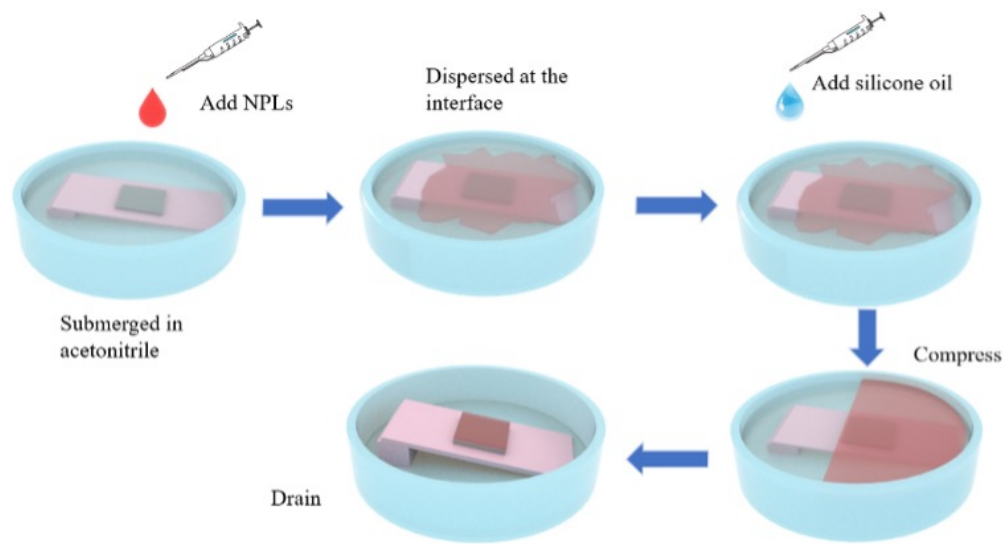


Figure S2: Schematic illustration of the liquid-air interface self-assembly process used for NPL monolayer deposition.

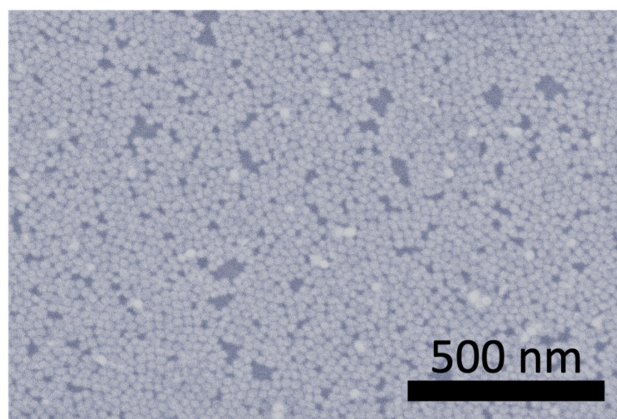


Figure S3: Scanning Electron Microscope (SEM) image of deposited NPL monolayer showing NPLs lying flat on the substrate. The few brighter regions indicate regions where NPLs are overlapping with one another.

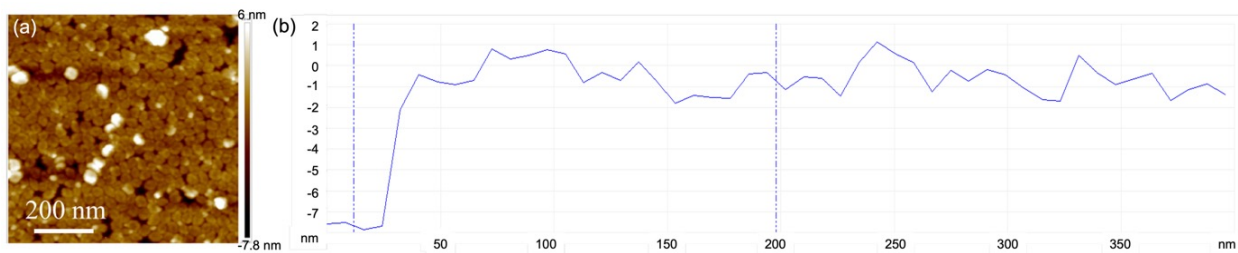


Figure S4: Topography of the surface of the monolayer NPL film taken via atomic force microscopy (AFM). (b) Film thickness and roughness measurement for a monolayer of NPL.

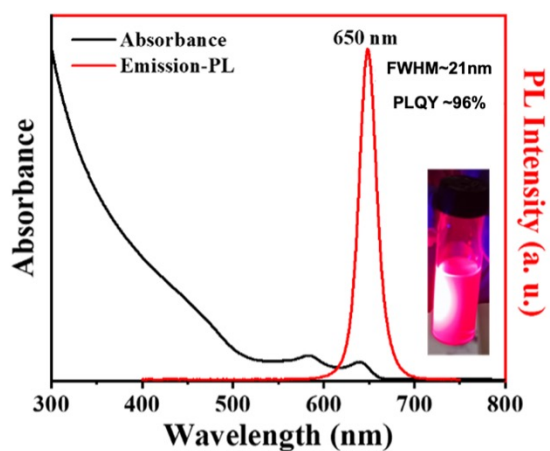


Figure S5: Absorbance (black line) and PL emission spectra (red line) of the CdSe/CdZnS NPLs in solution; heavy-hole and light-hole absorption at 643 and 587 nm, emission at 650 nm, quantum yield 96 ± 2 (method in S1). Inset: NPL colloidal solution under UV excitation.

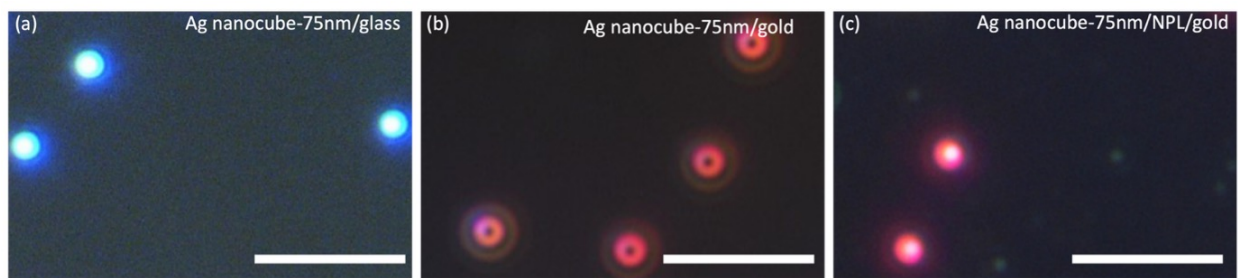


Figure S6: White-light dark-field scattering images of 75 nm Ag NCs on (a) a glass substrate, where they appear blue; (b) a template-stripped gold substrate, where they appear as donut/ring-like structures due to anisotropic scattering from plasmon coupling; and (c) a template-stripped gold substrate with the NPL monolayer as the spacer, where the scattered light appears as solid red dots. Scale bar: 10 μm .

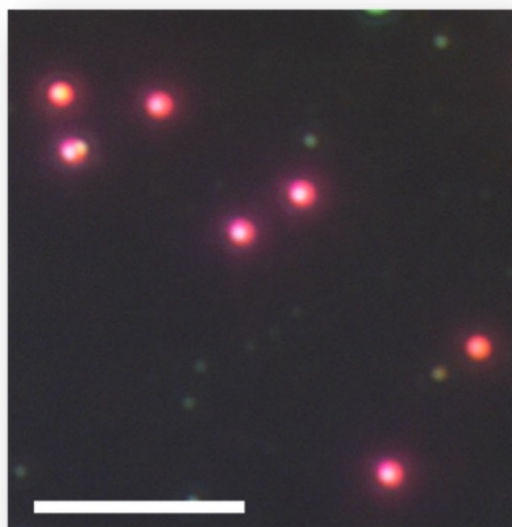


Figure S7: Fluorescence image under laser excitation at 405 nm of 75 nm Ag NC on gold substrate with core-shell NPL monolayer in between. Scale bar: 10 μm .

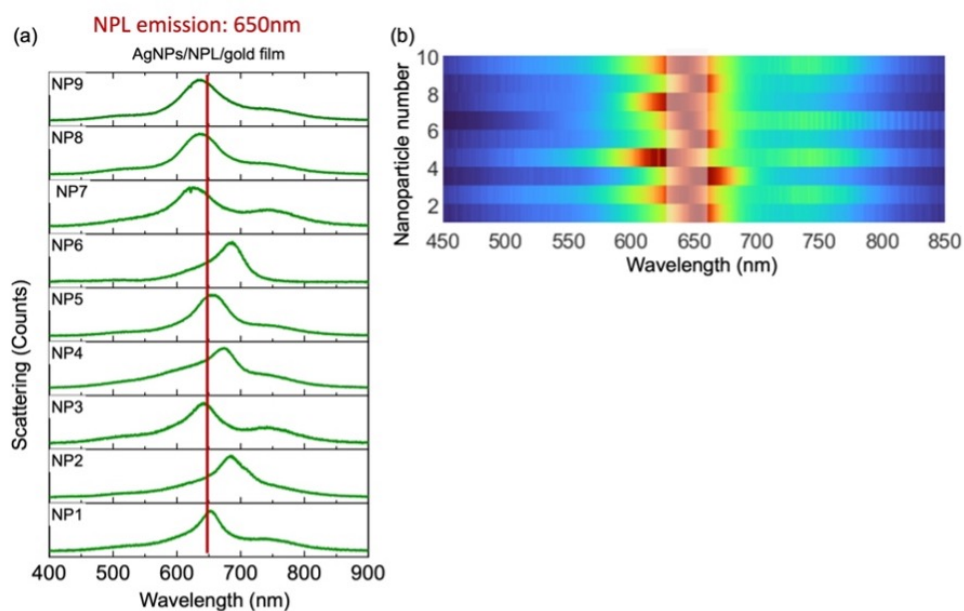


Figure S8: Dark-field scattering data for different single-particle NPoMs. (a) Spectra of nine NPoMs (NP1–NP9); the red line marks the NPL emission wavelength (650 nm). (b) Normalized spectra of ten NPoMs as a heat map.

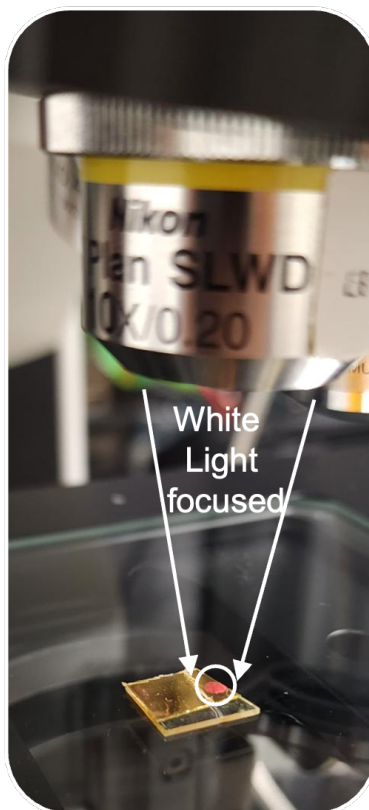


Figure S9: Setup for observing the emission by eye under focused white-light illumination.

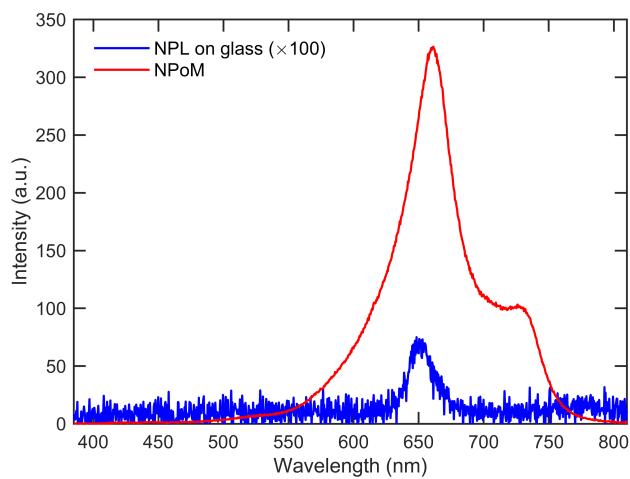


Figure S10: Quantitative comparison of peak intensity of NPL monolayer on glass and NPoM sample. The monolayer-on-glass signal is 435 $\times$ lower than the NPoM hotspot under identical acquisition parameters

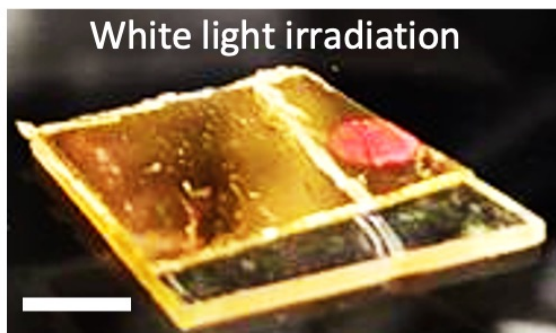


Figure S11: Photograph of the red emission spot, visible by eye, from densely packed Ag NCs on an NPL monolayer under white-light illumination. Scale bar: 1 cm.

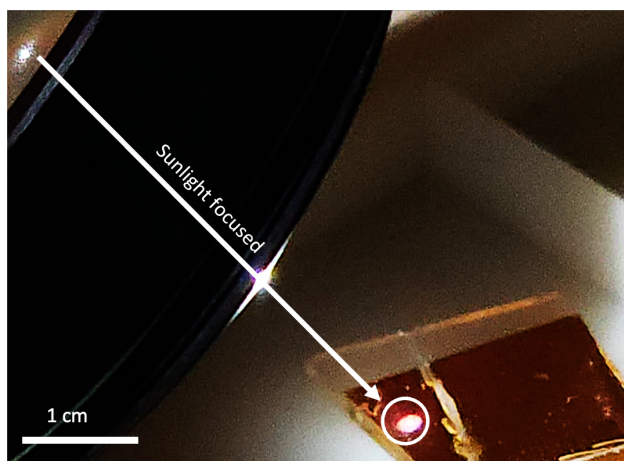


Figure S12: Setup for observing the emission by eye under focused sunlight.

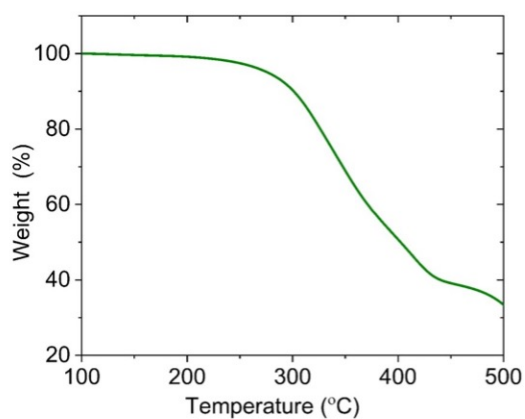


Figure S13: Thermogravimetric analysis of NPLs.

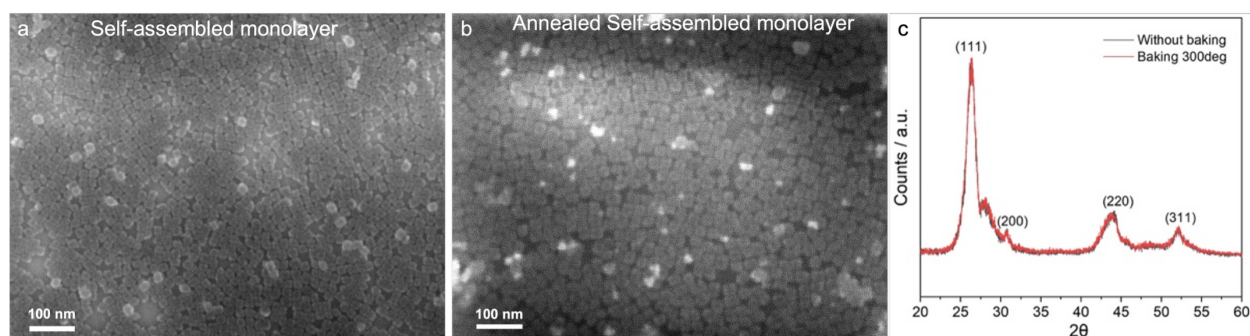


Figure S14: Structural and crystallographic integrity of the NPL monolayer spacer before and after thermal degradation. (a) SEM image of a self-assembled nanoplatelet (NPL) monolayer on glass prior to thermal treatment, showing a uniform, close-packed morphology with no evidence of aggregation or deformation. (b) SEM image of the same NPL monolayer after annealing at 300 °C, used as the degraded spacer in control NPoM experiments. No discernible changes in NPL shape, lateral dimensions, or surface morphology are observed, indicating that the thermal treatment does not induce large-scale morphological reconstruction. (c) X-ray diffraction (XRD) patterns of the NPL monolayer before (black) and after (red) annealing at 300 °C. The peak positions corresponding to the dominant crystallographic planes ((111), (200), (220), (311)) and their relative intensities remain unchanged within experimental resolution, confirming preservation of crystal structure and facet orientation and the absence of phase transformation.

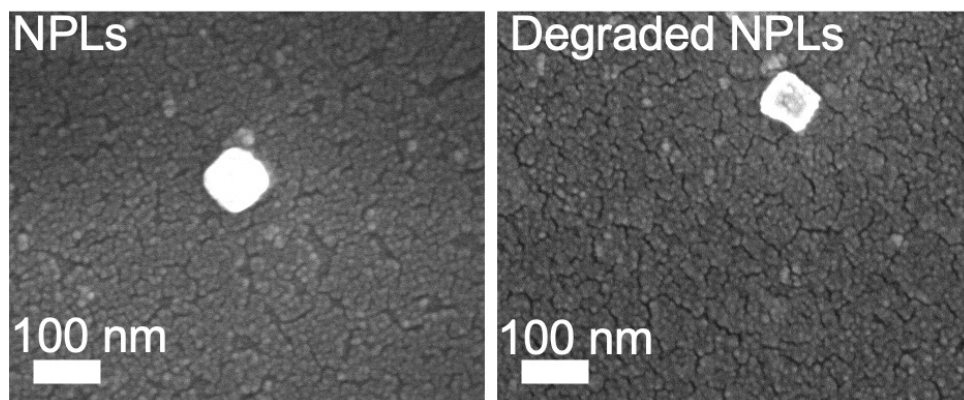


Figure S15: SEM images of the NPL monolayer with a Ag NC on top, before (left) and after (right) thermal degradation. Scale bars: 100 nm.

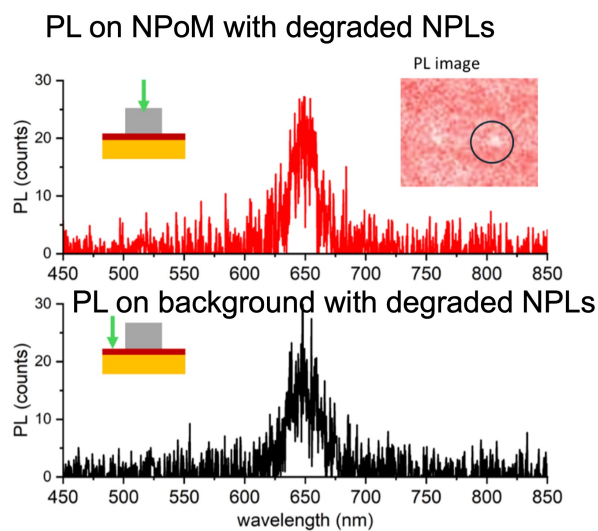


Figure S16: Photoluminescence (PL) spectra measured under 405 nm laser excitation from (top) the NPoM location on a sample containing a degraded (quenched) NPL monolayer spacer and (bottom) of an adjacent background region. The near-identical spectra show that the weak residual PL signal is background rather than NPL emission, confirming complete quenching of the NPLs after degradation.

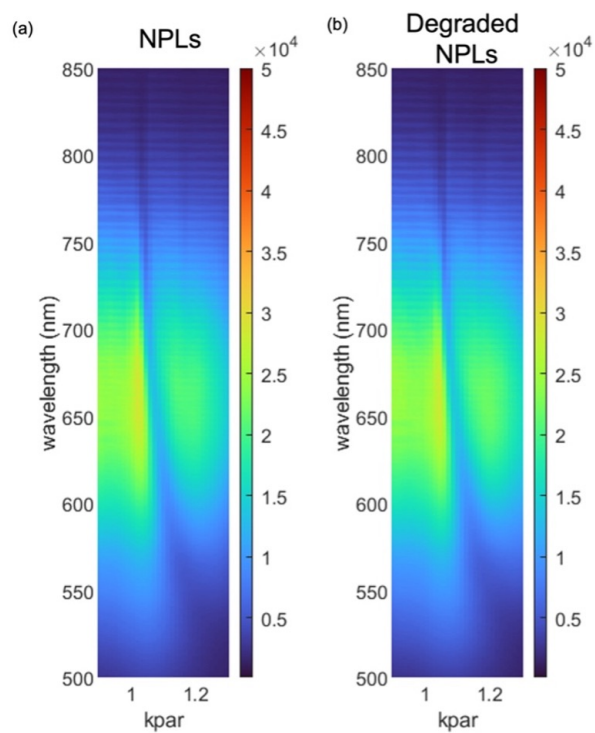


Figure S17: Comparison of the surface plasmon dispersion for (a) the emissive and (b) the degraded NPL monolayer, both deposited on a 45 nm-thick silver film. Both plots appear identical indicating that degradation leaves the linear optical properties of NPLs unchanged.

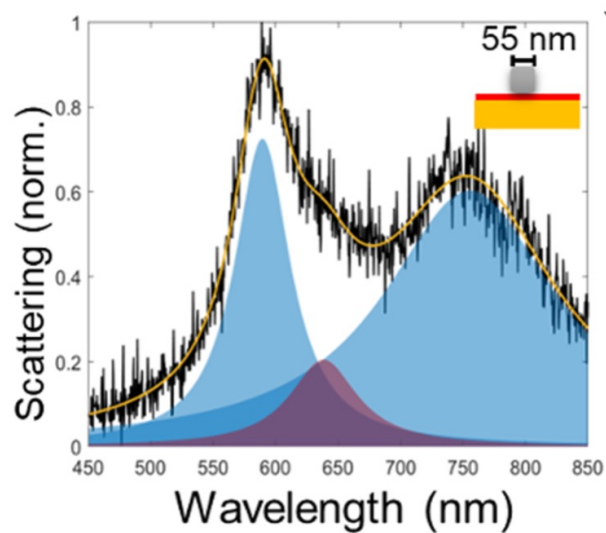


Figure S18: Dark-field scattering spectra for core shell red emitting CdSe/ CdZnS NPLs in NPoM cavity with smaller 55 nm Ag NC. Multiple peaks of the scattering spectrum are resolved. Lorentzian fitting resolves three peaks, at 575, 650, and 750 nm: the detuned plasmonic modes (blue) and the NPL emission at 650 nm (red).

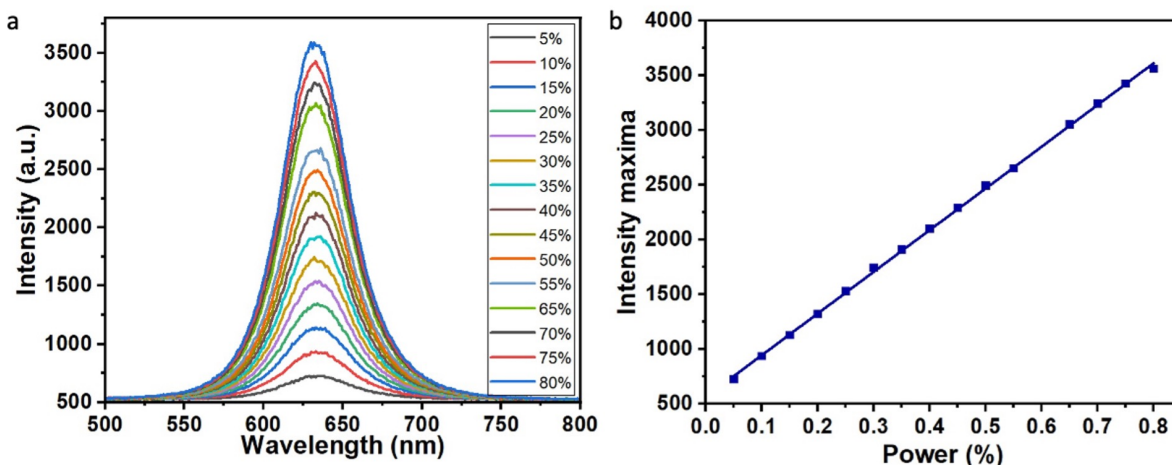


Figure S19: (a) PL spectra of CdSe/CdZnS core/shell NPLs spin-coated on a glass substrate under increasing 405 nm laser excitation power; percentages denote the fraction of the maximum laser power (percentages indicated). (b) Intensity maxima versus laser excitation power for NPLs on glass, showing linear dependence.

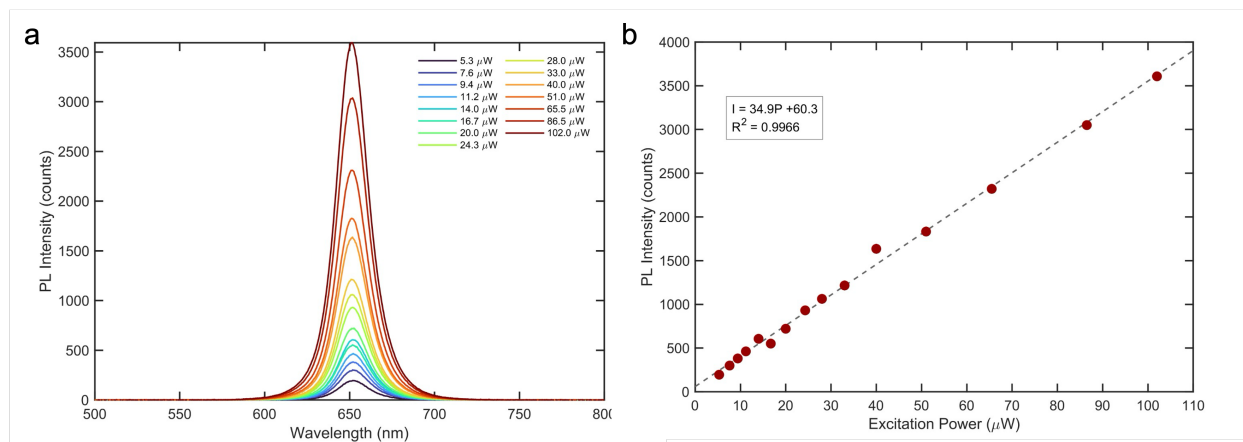


Figure S20: Power-dependent photoluminescence from a single NPoM under 405 nm excitation. (a) PL spectra under 405-nm excitation for increasing excitation power (measured at the sample plane). (b) Lorentzian-fit extracted peak intensity (650 nm) versus excitation power, showing linear scaling ($I = 34.9P + 60.3$, $R^2 = 0.9966$).

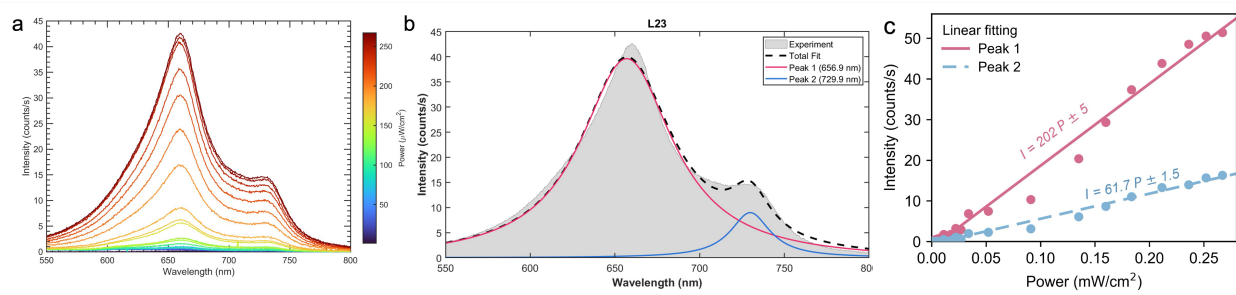


Figure S21: Figure S21: Power dependence of NP2 under white-light dark-field excitation. (a) Exposure-normalized spectra acquired at increasing incident power density (colour scale: 1.2–267 $\mu\text{W cm}^2$). (b) Highest-power spectrum (grey markers) fitted with two Lorentzian components: Peak 1 (660 nm) and Peak 2 (729 nm); dashed curve, total fit. (c) Fitted peak intensities versus incident power density with linear fits. Slopes are 202 ± 5 and 61.7 ± 1.5 counts s^{-1} per mW cm^2 for Peak 1 and Peak 2, respectively ($R^2 = 0.986$ and 0.987).

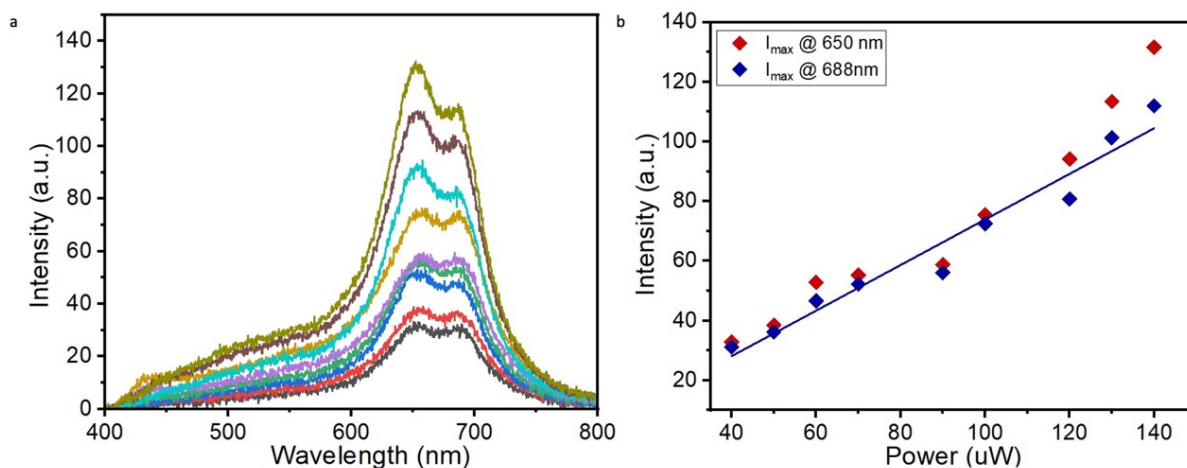


Figure S22: (a) Dark-field scattering spectra of a single NPoM with increasing excitation power. (b) Intensity maxima versus pump power for the two peaks at 650 nm and 688 nm, with linear fits.

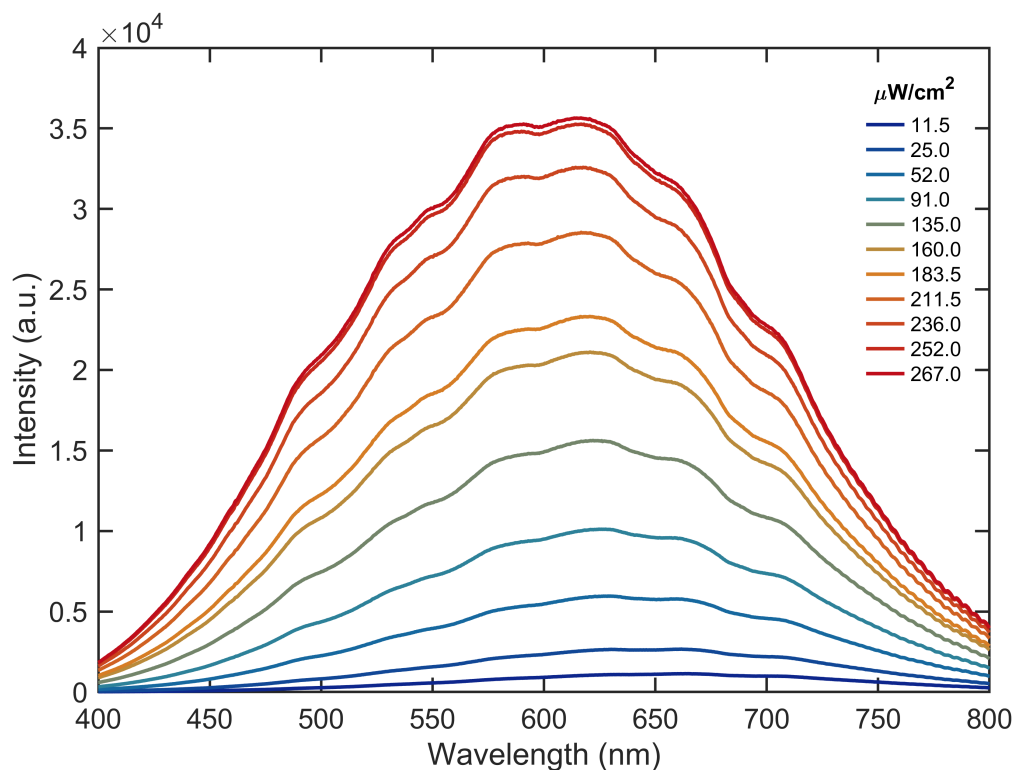


Figure S23: Excitation spectra of the white-light source measured at the sample plane for the irradiance settings used in the power-dependence measurement (legend). All spectra were acquired using the same illumination optics/attenuation configuration and identical spectrometer settings. The spectrum remains broadband and smooth across the full power range, with only minor gradual reshaping and no emergent feature near 650 nm.

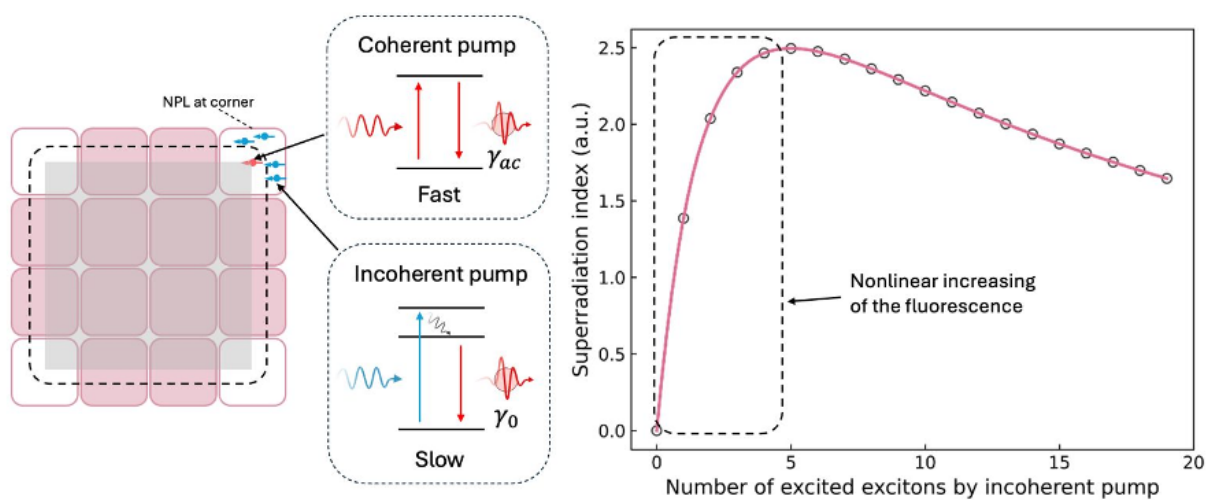


Figure S24: In this scenario, increasing the white-light fluence raises the number of incoherently excited excitons, which would enhance the cooperative emission and produce superlinear growth of the 650 nm peak before eventual saturation (right panel). Within the accessible power range the response is linear and unsaturated, so the emission is described by linear photoluminescence (see main text).

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