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

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Abstract

Quantum emitters (QE) weakly coupled to plasmonic cavities are known to exhibit scattering spectra that are largely indistinguishable from those in the absence of the emitters themselves. Here, we report a strong fluorescence peak visible in dark-field spectra of gap plasmons. This emission arises under incoherent white-light excitation from a few emitters in a monolayer of nanoplatelets (NPLs) within nanoparticle-on-mirror (NPoM) cavities. Single-NPoM spectra reveal an intense 650 nm peak from a few NPLs, matching their fluorescence and confirmed as emission by quenching and detuning controls. Simulations resolve the dipole mismatch of face-down NPLs: the bright cavity mode feeds dark modes whose in-plane corner fields drive the excitons. The 650 nm peak grows linearly with excitation power, as photoluminescence does, and at nearly twice the rate of scattering. The NPoM chip appears red to the naked eye under focused sunlight, at irradiances about seven orders of magnitude lower than conventional focused-laser excitation. Dark-field spectra of emitter-loaded nanocavities carry both scattering and emission, enabling laser-free few-emitter spectroscopy and sunlight-driven nanophotonics.

Keywords: Nanoparticle on mirror, Nanoplatelets, Monolayer, White-light illumination, Gap Plasmons

Introduction

A central paradox in modern optoelectronics is that while active light emitters have been miniaturized to the nanoscale, they often require cumbersome lasers as the pumping source.¹⁻⁴ Driving such emitters with an incoherent source such as a lamp, an LED or the sun, removes the laser from the system. Harnessing ubiquitous, incoherent light offers a transformative pathway to enabling fully integrated on-chip devices. Incoherent solar radiation offers an abundant, environmentally friendly, and freely available energy source, making solar-driven devices such as solar cells⁵ and solar-pumped light sources^{6,7} increasingly compelling. Despite this potential, while fluorescence from large emitter ensembles under incoherent illumination is well established, efficiently harnessing broadband radiation with only a few nanoscale quantum emitters (QEs) is far more demanding. The obstacles are (i) the low spectral

61 irradiance of a broadband source within the absorption band of the emitter, (ii) the small total absorption
62 cross-section of a few nanoscale emitters, and (iii) the weak outcoupling of their emission against the
63 scattering background of any cavity used to enhance it.^{8,9}

64 A well-established strategy to bridge these gaps involves embedding QEs within plasmonic nanocavities,
65 which exploit tightly confined gap plasmons to enhance excitation through field confinement and emission
66 through the Purcell effect¹⁰. However, direct *experimental* observation of a distinct emitter fluorescence
67 signature emerging within white-light dark-field spectra from deep-subwavelength plasmonic gaps
68 remains scarce. Unlike dielectric microcavities that offer high quality factors but suffer from poor spatial
69 confinement and complex emission patterns¹¹, plasmonic cavities, such as bow-tie antennas¹², dimers¹³,
70 and film-coupled nanoparticles¹⁰, enable sub-diffraction field localization¹⁴ and efficient, directional
71 emission.¹⁵ Among these, the nanoparticle-on-mirror (NPoM) configuration is particularly advantageous
72 due to its lithography-free fabrication and extreme field confinement capabilities.^{14,16,17} It has even
73 resolved the confined phonon modes of a single CdSe nanoplatelet by surface-enhanced Raman
74 scattering¹⁸. Indeed, for emitters in these sub-10 nm metal-metal gaps, radiative rates and quantum yields
75 are enhanced by orders of magnitude. For instance, Hoang *et al.* demonstrated gap fields in NPoMs with
76 ~200-fold enhancement, boosting embedded-emitter fluorescence by up to ~2,300-fold under laser
77 excitation while maintaining high radiative efficiency and directional output;¹⁶ in the same nanopatch-
78 antenna geometry, fluorescence enhancements exceeding 30,000-fold have been reported.¹⁹ Suspending
79 WSe₂ flakes over sub-20 nm trenches in gold gives a ~20,000-fold fluorescence enhancement.²⁰ Most
80 reports of strongly enhanced emission from plasmonic nanogap cavities employ laser excitation,¹⁹⁻²¹
81 which provides high spectral irradiance within the emitter absorption band. Under white light illumination,
82 the spectral irradiance available for excitation is reduced, and fluorescence is typically obscured in
83 conventional dark-field scattering measurements. Shang *et al.* observed resonant emission from a

monolayer semiconductor in a Fabry–Pérot microcavity under white-light illumination²², but the signal was weak: without nanoscale field confinement, the spectral irradiance that a lamp delivers within the emitter absorption band is orders of magnitude too low for strong emission.

Here, we report the observation of an intense, narrow emission from few NPLs within an NPoM cavity driven by incoherent broadband white light, together with controls that separate this emission from the coexisting scattering of the same cavity. This emission is conventional cavity-enhanced photoluminescence, not resonance fluorescence, and it is directly visible within the white-light dark-field spectrum of an individual nanogap cavity. This effect is achieved by integrating a monolayer of CdSe/CdZnS core/shell NPLs, precisely assembled via an improved self-assembly technique²³⁻²⁵, between a gold mirror and silver nanocubes (Ag NCs). This method ensures a uniform spacer layer with sub-nanometre gap control, offering a consistent dielectric environment superior to previous methods. NPLs, with their atomically precise thickness, large oscillator strengths, and exceptionally narrow emission linewidths^{1,23,24}, are ideally suited for strong light-matter interactions in NPoMs. Although the NPLs are oriented face-down and host in-plane dipoles-conditions that typically couple poorly to out-of-plane plasmon fields-we overcome this inefficiency by exciting dark cavity modes that generate strong in-plane fields.²⁶ Strikingly, under white light excitation, we observe an intense, narrow fluorescence peak at 650 nm from few NPLs in an NPoM cavity, spectrally congruent with their laser-excited fluorescence.

We chose CdSe/CdZnS nanoplatelets because they have broad absorption range and large absorption cross-section²⁷ at 500-640 nm that is six to seven orders of magnitude above that of the lanthanide ions used in earlier low-irradiance nanocavity work²⁸⁻³⁰, and because they emit with a quantum yield of 96 %. The emitter alone does not produce the effect: under identical white light the same monolayer gives 435 times less signal on glass, no visible emission on flat gold, and no resolvable peak under a detuned cube. The peak appears only when a resonant nanogap enhances excitation and

107 outcoupling together across the lamp spectrum. This combination of a broadband-absorbing two-
108 dimensional emitter with a nanogap cavity is what turns lamp white light into few-emitter
109 photoluminescence.

110 These findings reveal a previously unidentified coupling mode between QEs and NPoM cavities,
111 wherein a small number of NPLs produce bright, spectrally narrowband emission under incoherent
112 broadband excitation. These results show that, for a few-emitter nanogap cavity, the emission can become
113 spectrally resolvable directly in a white-light dark-field spectrum, an experimental regime where the
114 response is usually interpreted as purely scattering. This achievement suggests a new route towards
115 practical, solar-powered nanophotonic technologies, paving the way for integrated chip-scale light
116 sources, single-photon emitters, and sensors potentially driven by ambient light, circumventing the
117 complexity and costs associated with coherent laser sources while also advancing our understanding of
118 the well-studied NPoM configuration. Nanogap cavities loaded with such emitters can therefore be driven
119 by ambient light rather than a laser. The implication for nanocavity spectroscopy is immediate: once high-
120 quantum-yield emitters experience strong cavity enhancement, the dark-field spectrum is no longer a
121 purely scattering response - the emission peak is comparable to the plasmonic scattering – and the
122 quenching, detuning and filtered-excitation controls established here disentangle the two.

123 **Results**

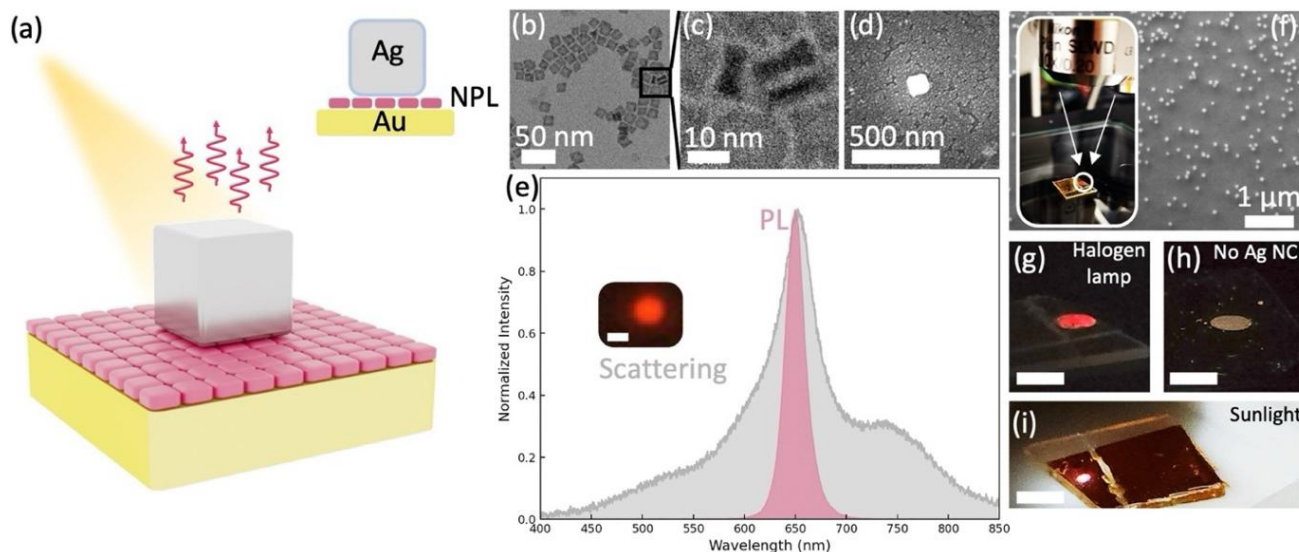


Figure 1. (a) 3D schematic illustrating photoluminescence emission due to broadband white light excitation of the nanoparticle on mirror (NPoM) cavity consisting of 200 nm thick Au on glass substrate, 7 nm nanoplatelet (NPL) spacer and 75 nm Ag nanocube (NC); cross-sectional schematic of the geometry inset. (b) Transmission electron microscopy (TEM) image of NPLs. (c) TEM image of the edge-aligned NPLs indicating the 4 nm thickness of NPLs. (d) Scanning electron microscope (SEM) image of the top-view of NPoM cavity. (e) Measured dark-field spectrum (grey) and 405 nm-excited photoluminescence (PL) spectrum (red) of a single NPoM with an NPL monolayer spacer. Inset: dark-field scattering image of the same NPoM. The scale bar is 500 nm. (f) SEM image of highly concentrated Ag NC sample on NPL monolayer on template stripped 200 nm Au substrate. Inset: Setup illustrating emission detection using white light excitation observable by the naked eye. (g) Photograph showing the distinct red emission spot observable by naked eye from densely arranged Ag NCs on top of a monolayer of NPLs, under illumination of a halogen lamp. (h) Control under identical illumination and camera settings: Au film coated with the same NPL monolayer but without Ag nanocubes, showing no distinct red emission. (i) Photograph showing the distinct red emission spot observable by naked eye, under real sunlight focused with a lens at 4pm in Sg sun. The scale bar in (g)-(i) is 1 cm.

Nanoplatelet monolayers in NPoM cavities

Figure 1a illustrates the plasmonic cavity-emitter architecture: 75 ± 10 nm silver nanocubes (Ag NCs, nanoComposix) are spin-coated onto a self-assembled monolayer of CdSe/CdZnS core/shell NPLs that form a uniform 7 nm layer on a 200 nm thick template-stripped gold attached to a glass substrate. Transmission electron microscopy (TEM) images (**Figure 1b**) show that the NPLs are square-shaped, averaging 21 ± 2 nm in width and ~ 4 nm in thickness (**Figure 1c**). Comprehensive synthesis procedures

for these NPLs are detailed in the **Supplementary Information**. A silicone-oil-assisted, refined liquid-air self-assembly process (**Figure S2**) produces a densely packed NPL monolayer by minimizing inter-particle spacing.^{23,24} **Figure S3** shows a densely packed mosaic-like pattern and a highly uniform assembled monolayer where individual NPLs are visibly oriented flat on the surface. This precise arrangement places each Ag NC atop about <10 NPLs. A top-view SEM image (**Figure 1d**) confirms the successful and reproducible positioning of individual Ag NCs on the NPL monolayer. Atomic force microscopy (AFM) corroborates these results (**Figure S4**), revealing a consistent monolayer thickness of ~7 nm with low surface roughness (RMS ~1 nm), minimized by the face-down orientation of the NPLs. Given the NPL core/shell thickness of 4.0 nm (**Figure 1(c)**), the additional ~3 nm is attributed to the organic ligand layer (oleic acid and oleylamine) coating the NPLs. The gap is designed to be large enough to prevent quenching caused by the nonlocal effects that emerge at sub-nanometer separations and small enough to leverage upon the strong field enhancement.^{31,32}

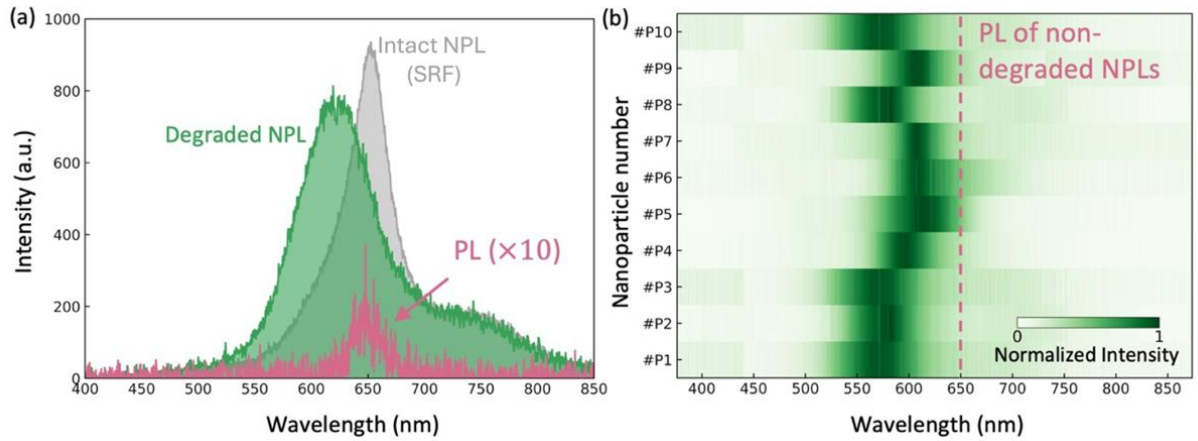
Emission resolved in the dark-field spectrum of a single cavity

Optical characterization in a custom-built microscopy setup enabled individual NPoM structures to be identified through dark-field and fluorescence imaging as well as spectroscopy (**see Methods**). The heavy-hole and light-hole excitonic absorption peaks of the NPLs are observed at 643 nm and 587 nm, respectively, while the fluorescence emission is centred at 650 nm with a quantum yield of 96 ± 2 % in solution (**Figure S5**; method in Methods). NPoM systems prepared by sparse or random emitter deposition often suffer from inconsistent placement of emitters relative to the bottom facet of the nanoparticles; close-packed colloidal-emitter monolayers assembled at liquid interfaces have recently been shown to overcome this, yielding highly consistent single-particle responses.³³ Our self-assembled NPL monolayer extends this uniformity to oriented two-dimensional emitters, producing uniform brightness and a consistent red appearance across both dark-field (**Figure S6**) and fluorescence images (**Figure S7**). **Supplementary**

171 **Figure 6** illustrates that the white-light dark-field scattering images of 75 nm Ag NCs on glass and gold
172 substrates, respectively, highlighting significant differences compared to the dark-field scattering images
173 obtained when NPLs are embedded in the cavity. Remarkably, the dark-field spectra of individual NPoMs
174 obtained under white-light illumination (**Figure 1(e)**), which comprises both scattering and emission
175 reveals an intense narrow resonance peak at 650 nm attributed to NPL emission, and broader plasmonic
176 resonances at 575 nm and 750 nm. The fluorescence peak remains within the nanoplatelet emission band
177 but shows particle-to-particle variation in peak position and linewidth (**Figure S8**). The variation follows
178 from the cavity: the emission that reaches the far field is the intrinsic NPL PL filtered by each cavity's
179 emission-enhancement and outcoupling response, which depends on facet size, corner rounding and gap
180 thickness.¹⁹ Thus, we hypothesize that this new spectral feature originates from the fluorescence emission
181 of the embedded NPLs, a departure from the conventional expectation of purely plasmonic responses
182 under white-light illumination and test the assignment below with quenching, detuning and spectrally
183 filtered excitation. Prior studies typically report spectral broadening due to emitter-induced dielectric
184 variations^{14,21,26}, and even reports of substantial fluorescence enhancement have not shown explicit
185 fluorescence signatures in white-light scattering experiments^{14,17,28}. This finding represents a significant
186 advancement in understanding emitter-plasmon interactions within NPoM architectures. The dark-field
187 spectrum of a single emitter-loaded NPoM therefore carries a resolvable emission component alongside
188 its plasmonic resonances, a co-existence we disentangle below through quenching, detuning and spectrally
189 filtered excitation.

190 The emission is bright enough to see by eye. For this we prepared a sample with Ag NCs packed
191 more densely on the NPL monolayer than for the single-particle spectroscopy above (SEM in **Figure 1f**).
192 This sample exhibited a distinct red emission under illumination of focused white light from a halogen
193 lamp (setup shown in inset **Figure 1f**, **Figure S9** and zoomed-in sample image shown in **Figure 1g**). To

disentangle nanogap-cavity emission from trivial NPL film brightness and metal-related effects, we ran two controls under identical white-light focusing and camera settings. The NPoM arrays exhibit a bright red spot (**Figure 1g, zoomed in image shown in Figure S11**). A gold film coated with the same monolayer but without Ag nanocubes shows only the yellow appearance of gold (**Figure 1h**), as expected: the planar Au mirror reflects the incident light and the optical field within the thin NPL layer remains weak, so little pump light is absorbed, in contrast to the strong excitation-field enhancement within the nanogap cavities. The quantitative benchmark is therefore a NPL monolayer on glass, free of both metal-induced quenching and reflection effects, measured under identical white-light focusing and acquisition settings. The monolayer-on-glass signal is $\sim 435\times$ weaker (**Figure S10**). Together, these comparisons demonstrate that the visible red emission requires the excitation-field enhancement and antenna-mediated outcoupling provided by the nanogap cavities, rather than the intrinsic brightness of the NPL film alone. The sample was also observed under sunlight, focussed via a lens and the sample appeared red, clearly visible to the naked eye as can be seen in **Figure 1i** (setup in **Figure S12**).



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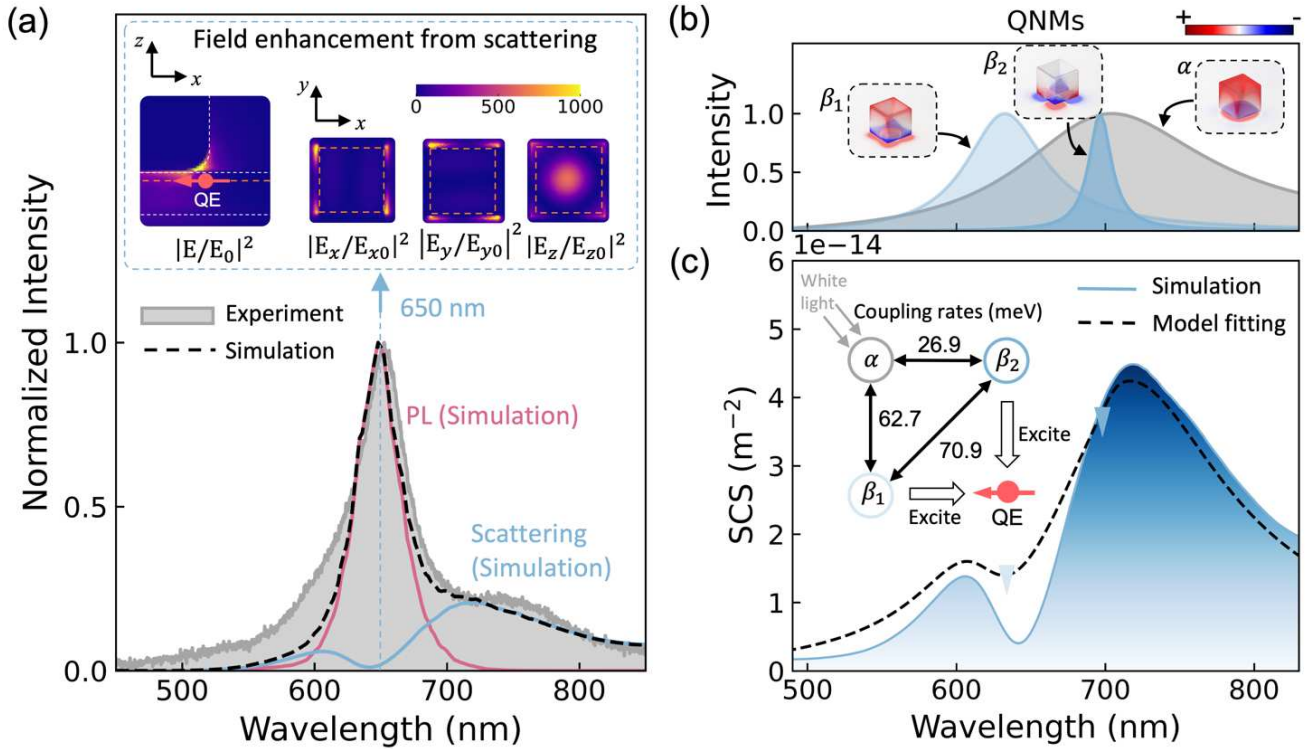
Figure 2. (a) Grey: White-light dark-field spectrum of a single NPoM with an intact (fluorescent) NPL monolayer spacer. Green: white-light dark-field scattering spectrum of a single NPoM prepared on a degraded (quenched) NPL spacer (300 °C anneal). Red: photoluminescence spectrum of the degraded NPL monolayer ($\times 10$ for visibility). (b) Statistics of single particle dark-field scattering spectra from 10 NPoMs with degraded NPL spacer (dashed line indicates the emission maximum wavelength of intact NPLs).

214

215 Quenching and detuning controls

216 To confirm that the 650 nm peak arises from NPL fluorescence rather than a plasmonic resonance,
217 we performed control experiments designed to quench the NPL emission in white-light dark-field
218 measurements. In the first experiment, we degraded the fluorescent NPL monolayer by removing its
219 organic ligands partially via methanol followed by heat treatment at 300 °C under ambient conditions
220 before depositing the Ag NCs. Partial removal of the passivating organic ligands from semiconductor
221 nanocrystals exposes surface trap states that open efficient non-radiative recombination channels,
222 resulting in pronounced photoluminescence quenching. The decomposition temperature was found using
223 thermogravimetric analysis (TGA) (**Figure S13**) which shows that organic ligands start decomposing at
224 300 °C. The SEM image of the degraded sample shows that the monolayer of NPLs remains intact on the
225 substrate (**Figure S14,15**). The degraded NPL monolayer now behaves solely as a non-emissive spacer.
226 **Figure S16** shows the PL spectrum collected at the NPoM location on the degraded-NPL sample together
227 with a spectrum from an adjacent background region. The two spectra are near-identical: the residual
228 signal is background, not NPL emission, and the degraded monolayer is non-emissive. As expected, the
229 dark-field scattering of the degraded (quenched monolayer) NPoM sample (green curve) in **Figure 2(a)**
230 now shows only two broad plasmonic resonances at 575 nm and 700 nm, with the absence of any peak at
231 650 nm while the red curve is the PL (x10 for visibility) from the quenched NPL monolayer. NPoMs on
232 the degraded monolayer show no 650 nm peak (**Figure 2(b)**), against NPoMs on the intact monolayer that
233 all do (Supplementary Fig. 8). The degradation process only quenches the fluorescence (red curve in
234 **Figure 2a**) while leaving the dispersive optical constants (n, k) essentially unchanged. The optical
235 constants n and k mainly reflect the underlying band structure and absorption cross-section, whereas
236 photoluminescence is highly sensitive to surface defect states that open non-radiative recombination
237 channels when ligands are removed. The preservation of its optical constants is confirmed by comparing

the propagating surface plasmon dispersion of the NPL monolayer on the silver film before and after degradation, as shown in **Figure S17**. In the second control experiment, we used smaller Ag NCs of 55 nm size to detune the cavity resonance from the NPL emission at 650 nm. As shown in **Figure S18**, only two peaks (blue curves) are well observed, showing a valley as predicted by the simulation, while the peak at 650 nm is nearly invisible in the dark-field spectrum.



243

Figure 3. (a) Comparison of experimental scattering (grey shaded) with the summed response (black dashed line) of simulated scattering (blue line) and simulated PL (red line) for a single NPoM. The vertical blue dashed line marks NPL emission wavelength (650 nm). Inset: Simulated field enhancement distribution in the xz -cross section (top left) and decomposed field enhancement in the xy -cross section (0.5 nm from the bottom of the cube) showing the x -, y -, and z -components at 650 nm. (b) Schematic illustration and spectral line shapes of the identified quasinormal modes (QNMs): α , β_1 , and β_2 modes. (c) Simulated scattering cross-section (SCS, blue shaded area) and analytical model fitting (black dashed line). Shallow and deep blue inverted triangles indicate the peak positions of the β_1 and β_2 modes, respectively. Inset: Diagram showing the extracted modal coupling rates (in meV) between the three modes from the analytical model. In this scheme, α is driven by white light and subsequently excites β_1 and β_2 through near-field coupling, then β_1 and β_2 excite the NPL QE via in-plane field resonances.

255

256 Simulations show that the emission is actively excited and clearly emerges above the scattering
257 background. As shown in **Figure 3a**, the simulated scattering spectrum (blue line), obtained by modelling
258 the NPL layer as a passive dielectric (see SI for details, **Figure S1**), exhibits a “hole-burning” feature
259 aligned to the position of the emission peak at ~ 650 nm. This feature indicates that the 650 nm emission
260 peak does not originate from structural scattering, which only contributes to the broadband background.
261 In contrast, the simulated PL spectrum (red line), where the NPL layer is modelled as a light-emitting
262 source, shows a clear emission peak at 650 nm. This peak closely matches the experimental result (grey
263 shaded), confirming its fluorescent origin. We then proceed to explain how this emission is excited by
264 white light. NPL excitons are known to exhibit in-plane resonance, allowing us to model the NPL layer
265 as a collection of quantum emitters (QEs) with in-plane dipole moments, referred to as in-plane QEs.³⁴
266 However, previous studies^{23,26} have highlighted the difficulty of aligning localized field polarization with
267 such in-plane dipoles. To clarify this mechanism, we analyse scattering simulations that reveal the field
268 response of the NPoM under white-light illumination. Specifically, we demonstrate the field enhancement
269 at the fluorescence wavelength of 650 nm. The upper inset of **Figure 3a** displays both in-plane and out-
270 of-plane field enhancement distributions. In the xz -cross section, intense hotspots are observed at the cube
271 corners near the gap, indicating highly confined mode volumes. The decomposed field enhancement in
272 the xy -cross section (0.5 nm from the bottom of the cube, marked by the orange dashed lines in the xz -
273 cross section) reveals dominant in-plane components along the x - and y -directions, with enhancements up
274 to ~ 1000 . In contrast, the z -component (out-of-plane) shows stronger enhancement at the facet center but
275 weaker at the corners. These hybrid hotspots, dominated by in-plane fields, effectively excite in-plane
276 QEs at the corners, enabling resonant NPL excitation and enhancing spontaneous emission by nearly three
277 orders of magnitude, in good agreement with experimental observations.¹⁰ The strong in-plane fields at
278 the corners originate from the coupling of the quasinormal modes (QNMs) of the system, as identified in

our analysis (see SI for details). Three QNMs are found within the white-light wavelength range of 550–830 nm (1.50–2.25 eV), with their field distributions and Lorentzian spectral line shapes shown in **Figure 3b**:

1. α mode: A pure out-of-plane field with $\hbar\omega_\alpha = 1.74 - 0.39i$ (unit: eV, Same as below), exhibiting a bonding-dipole field distribution that strongly couples to incident light.

2. β_1 mode: A hybrid mode with $\hbar\omega_{\beta_1} = 1.96 - 0.20i$, combining an out-of-plane longitudinal and a much stronger in-plane transverse component. It is weakly radiative and not efficiently excited by direct illumination.

3. β_2 mode: A pure in-plane mode with $\hbar\omega_{\beta_2} = 1.78 - 0.05i$, showing symmetric in-plane fields along the x - and y - axes, confined within the gap. These modes collectively generate the strong in-plane field.

While only the α mode couples efficiently to external light, the β_1 and β_2 modes are indirectly excited via near-field coupling, with coupling rates $g_{\alpha\beta_1}$, $g_{\alpha\beta_2}$ and $g_{\beta_1\beta_2}$. The strong in-plane components of the β_1 and β_2 modes drive fluorescence via resonant excitation, with the α mode acting as the excitation channel. To quantitatively analyze the coupling, we fit the simulated scattering cross-section (SCS, blue shaded area) using an analytical model³⁵ (black dashed line), allowing us to extract the coupling strengths among the three modes from the scattering curve, as shown in **Figure 3c** (see SI for details).

The resulting “hole-burning” spectral feature near 650 nm arises from destructive interference between the bright α mode and the dark β_1 and β_2 modes. The extracted coupling strengths are provided in the inset. In summary, three mechanisms produce the emission: (1) Polarization matching between the white-light-excited NPoM fields at the bottom corners of the cube and the in-plane NPL excitons, (2) resonant excitation of the NPL excitons by dark modes that generate strong in-plane fields at these corners, and (3) the indirect excitation of dark modes through coupling with a bright mode efficiently

driven by white light. No Rabi splitting appears in the dark-field spectra; the system is in the weak-coupling regime, with cavity-enhanced excitation and emission.

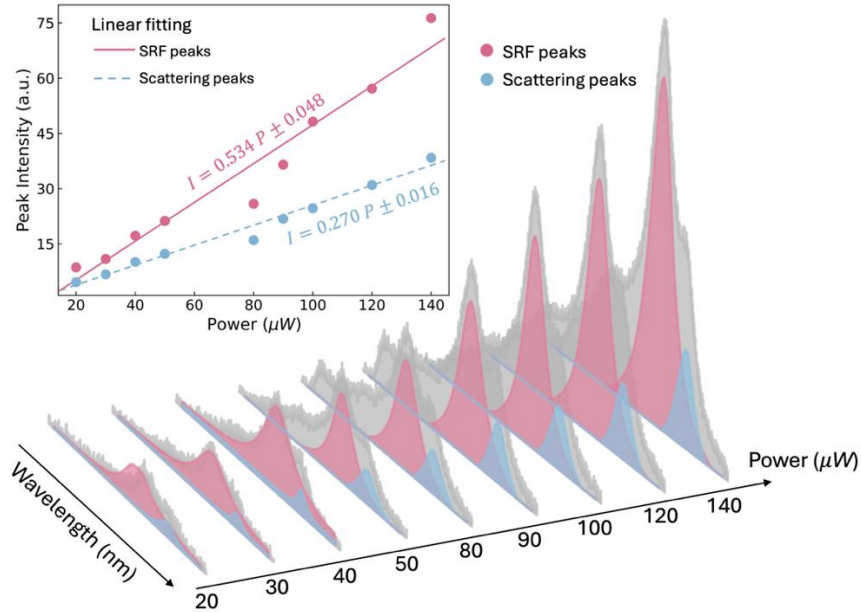


Figure 4. Waterfall plot of the dark-field spectra at excitation powers from 20 to 140 μW . Fitted curves distinguish the fluorescence peak (red, centred at ~ 650 nm) and the plasmonic scattering peak (blue, centred at ~ 710 nm). Inset: Plot of peak intensity I versus pump power P for the identified fluorescence peak at approximately 650 nm (represented by red circles) and the plasmonic scattering peak around 710 nm for reference (represented by blue circles). The linear fitting reveals that the 650 nm peak increases with incident light power at a rate 1.98 times higher than that of the scattering peak, highlighting its emissive nature.

To gain insight into the nature of these spectral features and the underlying light-matter interactions, we performed a power-dependent analysis of the NPoM system under white-light illumination. As depicted in **Figure 4**, the cavity incorporating CdSe/CdZnS core/shell NPLs prominently exhibits two spectral peaks: One at ~ 650 nm (Peak 1) and another at ~ 710 nm (Peak 2). The waterfall plot clearly shows how these peaks evolve as the excitation power is systematically increased from 20 μW to 140 μW . Fitting the spectra allows us to determine their spectral positions and intensity trends. The 650

322 nm peak intensity I grows linearly with incident power P at nearly twice the rate of the ~ 710 nm plasmonic
323 feature. This contrast supports the assignment of the 650 nm peak to an emissive process^{36–41} rather than
324 purely cavity scattering. The assignment is established independently by the quenching and detuning
325 controls (Figure 2, Figure S18) and by the spectral congruence with the laser-excited PL. These contrasting
326 behaviours are quantified in the inset of **Figure 4**, where linear fits yield a slope for Peak 1 that is ~ 2 times
327 that of Peak 2.

328 To validate that the 650 nm response reflects intrinsic NPL fluorescence, we conducted a control
329 experiment by spin-coating the same NPLs on glass. Their fluorescence intensity scales linearly with
330 power (**Figure S19**), matching the linear response of the 650 nm peak in the cavity. We further measured
331 power-dependent 405 nm-pumped PL from single NPoMs and extracted the emission peak intensity by
332 Lorentzian fitting, which shows near-linear scaling over the same excitation range (**Figure S20**).
333 Additional single-particle data (**Figure S21** and **S22**) corroborate this trend. Collectively, these findings
334 indicate that NPoMs boost the fluorescence signal while preserving its linear power dependence behaviour
335 with important implications for low-power, high-efficiency nanophotonic devices. The lamp spectrum
336 reshapes only gradually with power and shows no feature near 650 nm (**Figure S23**). A Dicke-like
337 cooperative-emission (superradiance) scenario,^{42,43} schematically illustrated in **Figure S24**, would
338 produce superlinear growth of the 650 nm peak at higher powers; within the accessible power range the
339 response is linear and unsaturated, so the emission is described by linear photoluminescence.

340 **Discussion**

341 CdSe/CdZnS nanoplatelets in a single nanogap produce a photoluminescence peak that is clearly
342 resolved within the white-light dark-field spectrum of the cavity at an irradiance of only $1\text{--}7\text{ mW cm}^{-2}$,
343 approximately seven orders of magnitude lower than that typically used for focused-laser excitation. The
344 NPoMs are 435 times brighter than the same monolayer on glass and visible to the naked eye. Second, the

cavity resolves the dipole-orientation problem of face-down platelets: the bright out-of-plane mode feeds dark modes whose in-plane corner fields, with intensity enhancements of $\sim 10^3$, drive the in-plane excitons, a design rule for two-dimensional emitters in nanogaps. Third, quenching, detuning and spectrally filtered excitation separate emission from scattering within one dark-field spectrum, a protocol that applies to any emitter-loaded nanocavity. Table 1 places the excitation conditions against previous nanocavity-enhanced emitters: the in-band irradiance here is 10^2 - 10^3 times below the lowest laser irradiance used for a single emitter in a plasmonic nanocavity³⁰, 10^3 - 10^5 times below the nanowatt-laser experiments on single quantum dots¹⁷, and 10 - 10^2 times below the white-LED illumination of a planar microcavity²²; each cavity intercepts ~ 0.1 pW of absorbable light.

Table 1. Excitation conditions of nanocavity-enhanced emitters compared with this work. In-band irradiance is the part of the incident irradiance within the emitter absorption band (the full irradiance for a laser; the 400–643 nm part of the halogen spectrum here, 0.4 of the 400-810 nm power, estimated for a 3000 K filament; ~ 0.8 of a white-LED spectrum for WS₂). Power per cavity is the in-band irradiance times the 75 nm facet area (5.6×10^{-11} cm²), used for every row so that the rows are comparable.

Work	Emitter and cavity	Excitation	Irradiance (W cm ⁻²)	In-band irradiance (W cm ⁻²)	Power per cavity	Enhancement reported
Hoang 2015 ²⁰	CdSe/ZnS QDs; Ag cube 70–80 nm on Au, ~ 1 nm gap	535 nm, 150 fs, 40 MHz; ~ 300 nm spot	1×10^3 (0.01– 10×10^3)	1×10^3	~ 60 nW	PL $\times 177$ – $2,300$; rate $\times 190$ – 880 ; QE ~ 50 %
Hoang 2016 ²²	single CdSe/ZnS QD; Ag cube on Au	488 nm CW, 30 nW to 1 μ W on ~ 300 nm spot (535 nm pulsed for lifetimes)	~ 40 (30 nW) to $\sim 1 \times 10^3$ (1 μ W)	same	~ 2 nW	PL $\times 1,900$ (single), $\times 495$ (mean); Purcell factor ≥ 540
Akselrod 2014 ¹¹	Ru dye in 8 nm gap; Ag cube ~ 80 nm on Au	535 nm, 200 fs, 20 MHz, 100 nW; 350 nm spot	$\sim 1 \times 10^2$	same	~ 6 nW	rate up to 860 ($>1,000$ for smaller gaps); QE >0.5
Wu 2019 ²¹	NaYF ₄ :Er,Yb UCNP monolayer; Ag cube on Au	980 nm CW, 7.5 mW, 15 μ m spot	4.2×10^3	same	~ 0.2 μ W	UCL $\times 3.3 \times 10^4$ (vs Au film); rate $\times 166$
Chen 2022 ⁴⁶	single NaGdF ₄ :Yb,Er UCNP; tilted Ag cube on Ag plate	976 nm CW	10^4 – 10^5 (power series, Fig. 3f)	same	~ 1 μ W	UCL $\times 7.0 \times 10^4$; 47 ns decay
Meng 2023 ⁴⁷	single sub-30 nm lanthanide nanocrystal; plasmonic nanocavity	980 nm CW	0.45	0.45	~ 25 pW	UCL $\times 2.3 \times 10^5$; 560 photons s ⁻¹

Shang 2022 ⁵	monolayer WS ₂ ; planar Si microcavity	white LED, Köhler, 85 μm spot	0.09	~ 0.07	n/a (planar)	resonant emission; no factor stated
Focused laser, this work (PL reference)	same NPoM sample	405 nm CW, 0.9 mW, $\times 100/0.9$ NA, 0.275 μm spot (SI Eq. S25)	3.8×10^5	same	$\sim 20 \mu\text{W}$	reference for the "seven orders of magnitude" statement
This work	a few (<10) CdSe/CdZnS NPLs; Ag cube 75 nm on Au, 7 nm gap	halogen lamp, dark-field 65–72°, 400–810 nm; focused sunlight	$1.1\text{--}7.4 \times 10^{-3}$	$0.4\text{--}3 \times 10^{-3}$	0.03–0.2 pW	PL $\times 435$ vs monolayer on glass; peak resolved in the dark-field spectrum

Light scattered by the cube also launches surface plasmon polaritons on the gold film, and these could excite platelets far outside the gap. Three observations exclude a significant contribution from this channel. A monolayer on gold without cubes emits nothing visible under the same illumination (**Figure 1h**), because its platelets are quenched and see a suppressed tangential field. A 55 nm cube launches surface plasmons as efficiently as a 75 nm cube yet shows a strongly suppressed 650 nm peak (**Figure S18**). And the position and width of the peak follow the individual cavity (**Figure S8**), which far-field emission from the film would not do. The platelets under the four corners of one cube share the cavity mode and add incoherently: broadband incoherent pumping, inhomogeneous platelet linewidths and a strictly linear power dependence leave no signature of collective emission, and the model treats them as independent dipoles.

These results extend the well-studied NPoM configuration with a self-assembled monolayer of flat NPLs, and the platform's demonstrated properties point directly to applications in chip-scale photonics. Dense nanocube coverage already operates as a centimetre-scale red light emitter under lamp or sunlight illumination; diluting the monolayer offers a route to few- and single-photon sources; and because any change in the nanogap environment appears directly in the emission, the same cavities act as sensors powered by ambient light.

This study establishes the direct appearance of few-emitter photoluminescence within the white-light dark-field spectrum of a plasmonic nanogap cavity as both a phenomenon and a measurement capability. A self-assembled monolayer of face-down NPLs, with atomic-level thickness precision, forms

the cavity spacer and places <10 emitters in each hotspot; quenching, detuning and filtered-excitation controls cleanly separate their emission from the scattering background, at irradiances seven orders of magnitude below focused-laser excitation. Quasinormal-mode analysis resolves the central paradox of this geometry: the bright out-of-plane cavity mode feeds dark modes whose strong in-plane corner fields drive the in-plane NPL excitons, turning an unfavourable dipole orientation into an efficient excitation pathway. The emission grows linearly and without saturation, coexisting with, and resolvable against, the conventional scattering response of the cavity. These results recast dark-field spectra of emitter-loaded nanocavities as carrying both scattering and emission, and open a route to laser-free few-emitter spectroscopy and to chip-scale photonic devices driven by ambient light or sunlight.

Methods

Sample fabrication. CdSe/CdZnS core/shell NPLs were synthesized and assembled into a monolayer on 200 nm template-stripped Au by the silicone-oil-assisted liquid–air self-assembly method, and Ag NCs (75 ± 10 nm, nanoComposix) were spin-coated on top. Full synthesis, assembly, and characterization protocols are given in the Supplementary Materials (§S1).

Single-particle dark-field and fluorescence spectroscopy. Individual NPoMs were located and measured in a custom-built microscopy setup based on an inverted microscope (Nikon Ti-U) combining white-light dark-field imaging, fluorescence imaging, and spectroscopy. White light from a halogen lamp illuminated the sample through the dark-field annulus (NA 0.91–0.95) of a Nikon CFI TU Plan Fluor BD 100×/0.90 objective, which also collected the scattered and emitted light. Spectra were acquired with an Andor Kymera 193i spectrograph (300 lines/mm grating, 500 nm blaze, centered at 600 nm) and an Andor DU920P-BV CCD cooled to -60 °C, in 10 s single scans. Fluorescence was excited with a 405 nm diode laser (Thorlabs PL205). Illumination power was measured with a Nova power meter at the objective

entrance; for the power series in Fig. 4, the power of the overfilling beam was varied from 20 to 140 μW , corresponding to sample-plane dark-field irradiances of $\approx 1.1\text{--}7.4\text{ mW cm}^{-2}$ (§S3, Eqs. S21-S23).

Numerical simulations. Full-wave scattering and quasinormal-mode analyses were performed in COMSOL Multiphysics with the MAN toolbox (§S2).

Data availability

The data supporting the findings of this study are available within the article and its Supplementary Information. Source data are provided with this paper.

Code availability

The COMSOL models, the MAN quasinormal-mode scripts and the spectral fitting code are available from the corresponding authors on request.

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Author contributions

511 D.A., J.K.W.Y., E.G.D. and H.V.D. conceived the idea and designed the experiments. J.K.W.Y. and
512 H.V.D. supervised the research. D.A. and N.V.C. performed the optical experiments and analysed the
513 data. W.Z. performed the simulations; W.Z. and L.W. developed the theory. S.A. and E.G.D. designed
514 and synthesized the core/shell NPLs. E.G.D. and S.C. assembled the NPL monolayers on the metal
515 substrates. T.T.H.D. and S.T.H. built the custom microscope. X.Liang collected the TGA data. D.A. wrote
516 the initial manuscript; N.V.C., W.Z., E.G.D., N.A.M., H.V.D., L.W. and J.K.W.Y. revised it with input
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518 **Competing interests**

519 The authors declare no competing interest.

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