

RESEARCH ARTICLE

Open Access



Scanning-exciton optical nanoscopy using a single quantum dot

Zhiyuan Wang^{1,2,3†}, Jiahao Han^{1,2†}, Xiaoqi Hou^{4,5†}, Jianwei Tang^{1,2*} , Weixi Liu⁶, Weiwang Xu¹, Zhaohua Tian¹, Hongli Chen^{1,2}, Jinkang Cao^{1,2}, Yaocheng Shi⁶, Haiyan Qin^{4*} and Xue-Wen Chen^{1,2*}

Abstract

Mapping light fields and local density of optical states (LDOS) around nanostructured materials is instrumental for advancing both fundamentals and practical applications in nano-optics, nanomaterial science, and quantum technologies. In particular, LDOS governs key processes such as spontaneous emission, light scattering, van der Waals interactions, and nanoscale heat transfer, yet it remains inaccessible with conventional optical microscopy techniques. Here, we develop scanning-exciton optical nanoscopy to simultaneously map the light intensity and LDOS near plasmonic and dielectric photonic structures with a few-nanometer resolution. By grafting a rationally designed core-shell-shell quantum dot to a silica nanotip, we obtain an ultra-photostable and sensitive scanning quantum probe carrying excitons that are generated and decay at the rates proportional to the local light intensity and LDOS, respectively. We first demonstrate the correlative dual-parameter sensing capability using a model plasmonic structure, and subsequently apply it to uncover previously unobservable nanoscale coupling physics occurring in a plasmonic trimer. Furthermore, we report the first optical mapping of LDOS over a photonic-crystal nanocavity, visualizing the resonant nanocavity mode and its position-dependent coupling with a quantum emitter, essential for quantum photonic applications. Our work demonstrates a transformative technique towards bridging the gap between surface morphology and far-field optical response, and constitutes a platform for exploring light-matter interactions at deep-nanoscale and single-quanta level.

Keywords Scanning near-field optical microscopy, Local density of optical states, Colloidal quantum dot, Plasmonic nanostructure, Photonic-crystal nanocavity

[†]Zhiyuan Wang, Jiahao Han and Xiaoqi Hou have contributed equally to this work.

*Correspondence:

Jianwei Tang
jianwei_tang@hust.edu.cn

Haiyan Qin
hattieqin@zju.edu.cn

Xue-Wen Chen
xuewen_chen@hust.edu.cn

¹ School of Physics, Wuhan National Laboratory for Optoelectronics, Institute for Quantum Science and Engineering and Hubei Key Laboratory of Gravitation and Quantum Physics, Huazhong University of Science and Technology, Wuhan 430074, China

² Wuhan Institute of Quantum Technology, Wuhan 430206, China

³ College of Physics and Optoelectronics, Taiyuan University of Technology, Taiyuan 030024, China

⁴ Zhejiang Key Laboratory of Excited-State Energy Conversion and Energy Storage, Department of Chemistry, Zhejiang University, Hangzhou 310058, China

⁵ School of Chemistry and Materials Science, Hangzhou Institute for Advanced Study, University of Chinese Academy of Sciences, Hangzhou 310024, China

⁶ Centre for Optical and Electromagnetic Research, State Key Laboratory of Extreme Photonics and Instrumentation, College of Optical Science and Engineering, Zhejiang University, Hangzhou 310058, China

1 Introduction

Recent breakthroughs in nanofabrication technology and material science have enabled precise control over light fields and their interactions with matter down to 10 nm scale or even to atomic scale [1–4], unlocking a broad range of transformative applications. Meanwhile, the burgeoning field of quantum nanophotonics integrates concepts and techniques from nanophotonics and quantum physics [5, 6], leveraging delicate on-chip coupling between nanoengineered light fields and solid-state single quantum systems [7–11]. Accordingly, the ability to precisely map light fields and the LDOS which tells the atom–field coupling rate is key to uncovering the physical insights and pushing the boundaries of these exciting fields. Over the past several decades, substantial efforts have been devoted to acquiring these two types of critical information. Scanning near-field optical microscopy (SNOM) is now widely used to image the light field near photonic nanostructures at tens of nanometer resolution [12–16]. However, except from some special situations [17], SNOM lacks access to LDOS—a key parameter governing light-matter interactions [18–20], van der Waals forces [21], nanoscale heat transfer [22], and more. For LDOS mapping, researchers have to resort to electron beam based approaches [23], such as electron energy-loss spectroscopy [24, 25] and cathodoluminescence [26]. Although these techniques have their own merits, they operate under vacuum, rely on complex instrumentation, and lack access to light-field information.

A promising optical scheme to simultaneously map light fields and LDOS in ambient conditions is to replace the physical probe of standard SNOM by a quantum emitter, ideally an atom [27, 28]. In principle, a scanning atom probe could be brought arbitrarily close to the positions of interest and detect the local light intensity via absorption at atomic resolution. Further, the atom probe, as a quantum sensor, can sense the LDOS via its fluorescence lifetime change [18]. Earlier experimental demonstrations employed single emitters embedded in solid-state hosts, such as single molecules in organic microcrystals [29] and nitrogen-vacancy centers in nanodiamonds [30–33], highlighting the potential of this technique. However, such single-emitter probes confront a number of issues in ambient conditions. For instance, single-molecule probes in organic microcrystals suffer from relatively large size [29], photobleaching and photoluminescence (PL) blinking at room temperature [34]. Nanodiamond probes with single nitrogen-vacancy centers [30–33, 35] have a broad emission spectrum (over 100 nm linewidth),

small absorption cross-section ($\sim 10^{-17}$ – 10^{-16} cm²), and highly inhomogeneous quantum efficiencies with significant nonradiative decays. As a result, nanodiamond probes typically show small changes in the fluorescence lifetime [32, 33]. These issues significantly jeopardize the use of emitter-host quantum systems as reliable and sensitive LDOS probes [35]. Different from the emitter-host systems, colloidal quantum dots (QDs) are sub-10 nm semiconductor nanocrystals that act like atoms due to quantum confinement [36] and typically have a much narrower, tunable emission spectrum and orders of magnitude larger absorption cross-section [37, 38], showing their potential as sensitive probes with negligible background [39–42]. Indeed, single QDs positioned with microfluidic flow control were shown to probe LDOS around a silver nanowire [43], although they cannot be freely scanned in all three dimensions with a nanometer resolution. However, conventional colloidal QDs suffer from poor photostability at the single-dot level—particularly in ambient conditions—and are prone to severe PL blinking and photobleaching. This limitation substantially undermines the applicability of a single QD as a scanning nanoscopic light source, especially for LDOS imaging. Taken together, despite extensive experimental endeavors over the past three decades, an optical microscopy technique to robustly map nanoscale light fields and LDOS remains elusive.

Here, we develop scanning-exciton optical nanoscopy (SEON) with an ultra-photostable scanning QD probe and demonstrate simultaneous mapping of the light intensity and LDOS near nanostructures with unprecedented fidelity and a spatial resolution of a few nanometers. We realize an air-photostable and robust scanning-exciton probe by grafting a single QD (~ 6.6 nm) with an optimized core/shell/shell structure on the apex of a 40 nm-diameter silica nanotip. Here the monoexciton within the QD serves as the key sensing unit for the light intensity and LDOS. The dual-parameter sensing capability of SEON is demonstrated using a single gold nanosphere, with the first direct visualization of both the illumination-dependent light intensity distribution and the illumination-independent LDOS profile. Such simultaneous dual-parameter sensing enables a correlative analysis of the two types of information around the nanostructure to reveal the intricate physics previously inaccessible. We apply SEON to reveal previously unobservable intricate coupling physics in a plasmonic trimer system, and, for the first time, to optically map the LDOS and nanocavity mode over an integrated photonic-crystal nanocavity structure.

2 Results

2.1 Photostable scanning quantum-dot probe for dual-parameter sensing

We rationally design and synthesize high-quality CdSe/CdS/ZnS (core/shell/shell) QDs to meet the stringent requirements as an effective SEON probe (Supplementary Material Section 1.1). The energy band alignment diagram of the QDs is illustrated in Fig. 1a, where the band-edge hole wavefunction is confined in the CdSe core, while the band-edge electron wavefunction is delocalized over the CdSe/CdS region. The ZnS shell isolates the electron wavefunction from the environment and thus substantially improves the QD's photostability in ambient conditions. A small CdSe core is selected to suppress the biexciton quantum yield and increase the spatial resolution as the light absorption and emission predominantly occur in the core region [44]. To achieve target wavelength and stability with minimal total size, we epitaxially grow a thin intermediate CdSe shell and an outer ZnS shell. Transmission electron microscopy images show that the QDs exhibit a non-spherical shape due to the formation of the hexahedral nanocrystal and have an overall size of about 6.6 nm and a CdSe core of 3 nm (Supplementary Material Section 1.2). The QDs feature an emission peak at 605 nm and a spectral linewidth of 17 nm (Supplementary Material Section 1.3). QDs with different emission peaks can be synthesized by tuning the core and shell sizes. Our QDs possess monoexciton quantum efficiencies of over 96% (Supplementary Material Section 1.4). The tunable narrow-linewidth emission with near-unity quantum yield enables high-sensitive LDOS mapping for a given nanophotonic structure.

As schematically illustrated in Fig. 1a, a QD is grafted at the apex of a 40 nm-diameter silica nanotip formed by pulling a standard single-mode fiber (Supplementary Material Section 2.1). We observe no appreciable changes of the PL lifetime and intensity from the same QD before and after it is grafted to the silica nanotip apex (Supplementary Material Section 2.2). The fiber base is glued onto a tuning fork mounted on a three-dimensional (3D) piezoelectric nanopositioner which enables 3D closed-loop scanning of the QD at sub-nanometer resolution. SEON relies on mono-excitonic dynamics within the QD: Under light illumination, an exciton in the QD can be generated and recombined with the rates proportional to the local light intensity and LDOS respectively. Thereby, we realize SEON to simultaneously map two distinct types of optical properties, i.e., light intensity and LDOS, around the same material structure. The detailed experimental setup for SEON can be found in Supplementary Material Section 3.

Next, we thoroughly characterize the emission properties of a scanning quantum-dot probe, including the

photon statistics, photostability, and LDOS sensitivity. Figure 1b shows the measured second-order photon correlation function, which exhibits a pronounced anti-bunching dip at zero delay, confirming that the nanotip contains a single emissive QD [45]. Here a non-zero normalized anti-bunching dip of 0.1 is due to a nonvanishing biexciton quantum yield [46], corresponding to the ratio of biexciton to monoexciton quantum yields $\eta_{\text{BX}}/\eta_{\text{X}} = 0.1$. Importantly, in our experiment we operate under weak excitation conditions, resulting in negligible biexciton contribution [46]. Figure 1c displays a typical PL intensity time trace of five minutes with on-time fraction over 98%. Notably, the scanning QD probe can continuously emit for over three hours without degradation (Supplementary Material Section 4). The detected fluorescence has a signal-to-background ratio of 55. We further demonstrate that the excited-state lifetime of the QD is highly sensitive to LDOS variations, highlighting its near-unity monoexciton quantum yield. When the QD probe is positioned above a glass coverslip, we observe distinct PL decay curves at varied QD-substrate distances as shown in Fig. 1d. Moreover, all the PL curves exhibit monoexponential decay dynamics, which is crucial for accurately extracting the decay rate with a small number of photons (Supplementary Material Section 5). The extracted position-dependent decay rates are plotted in Fig. 1e and agrees well with the calculated radiative decay rate [19]. Here in the calculations the emission dipole moment of the QD probe [47] is modelled as a prolate spheroid consisting of three orthogonal incoherent dipoles characterized by the parameters (R, θ, ϕ) (Supplementary Material Section 6). All these measurements demonstrate that our QD probe possesses the following favorable properties: outstanding photostability in air, near-unity quantum efficiency, monoexponential PL decay, and low background.

2.2 Dual-parameter nanoimaging on a model plasmonic structure

To faithfully demonstrate SEON's capability, we first carry out experiments with a model plasmonic structure, i.e., a gold nanosphere (AuNS). Its optical response under excitation and LDOS distribution are well-established in theory. However, simultaneous mapping of its distinct distributions of the light intensity and LDOS under varied illuminations has not been visualized in experiment. Here we perform SEON (Supplementary Material Section 3.1) to experimentally illustrate how the light field around an AuNS (76.4 nm in diameter; see Supplementary Material Section 7 for AFM topographic image and scattering spectrum) is highly illumination dependent while the LDOS is independent. As sketched in Fig. 2, the excitation (532 nm wavelength; ~ 100 ps pulse width;

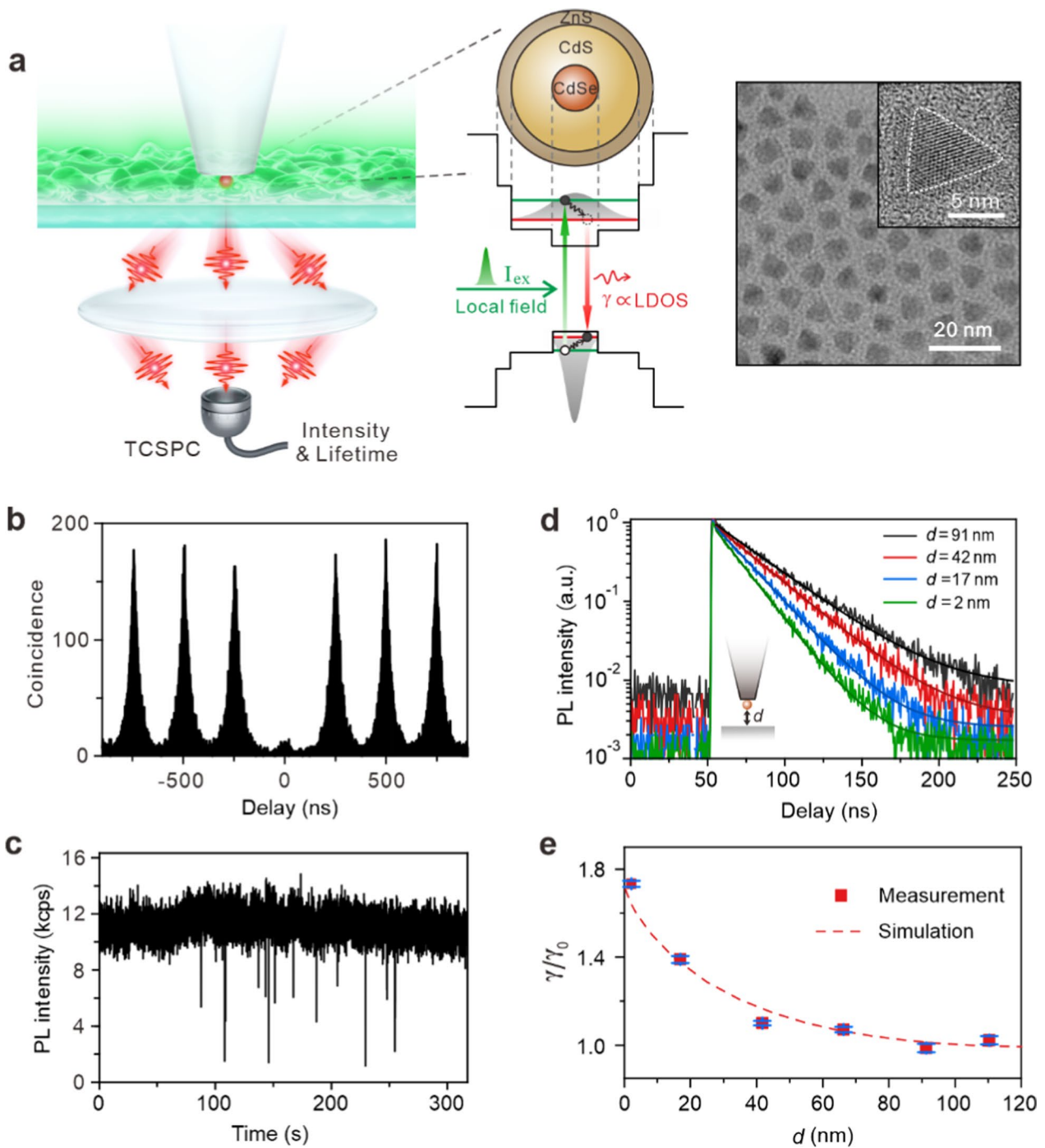


Fig. 1 Photostable scanning quantum-dot probe with dual-parameter sensing capability. **a** Schematics of the scanning-exciton optical nanoscopy (SEON), energy band alignment scheme of the CdSe/CdS/ZnS core/shell/shell quantum dot (QD), and transmission electron microscopy (TEM) image of the QDs. The inset shows a high-resolution TEM image of one QD. Excitons in a QD are excited upon light absorption, and decay via emitting photons, which are registered with a time-correlated single-photon counting (TCSPC) unit for light intensity and fluorescence lifetime mapping. **b** Second-order photon correlation function of the fluorescence from the scanning QD probe. The pronounced anti-bunching dip at zero delay confirms the probe carries only one QD. **c** A representative PL intensity time trace of the QD probe under constant excitation, exhibiting an on-time fraction over 98%. The photon counts per time bin (20 ms) is 224 ± 18 . **d** PL decay curves of the QD at varied distances from a glass substrate. All decay curves are well described by a monoexponential decay model, with an Adjusted R^2 of 0.99 indicating strong model-data consistency. **e** Measured (symbols) and calculated (dashed line) normalized decay rate as a function of the QD-substrate distance. The error bars represent the fitting uncertainties (1σ). The QD probe's dipole parameters (R, θ, ϕ) = (3.1, 106° , 137°)

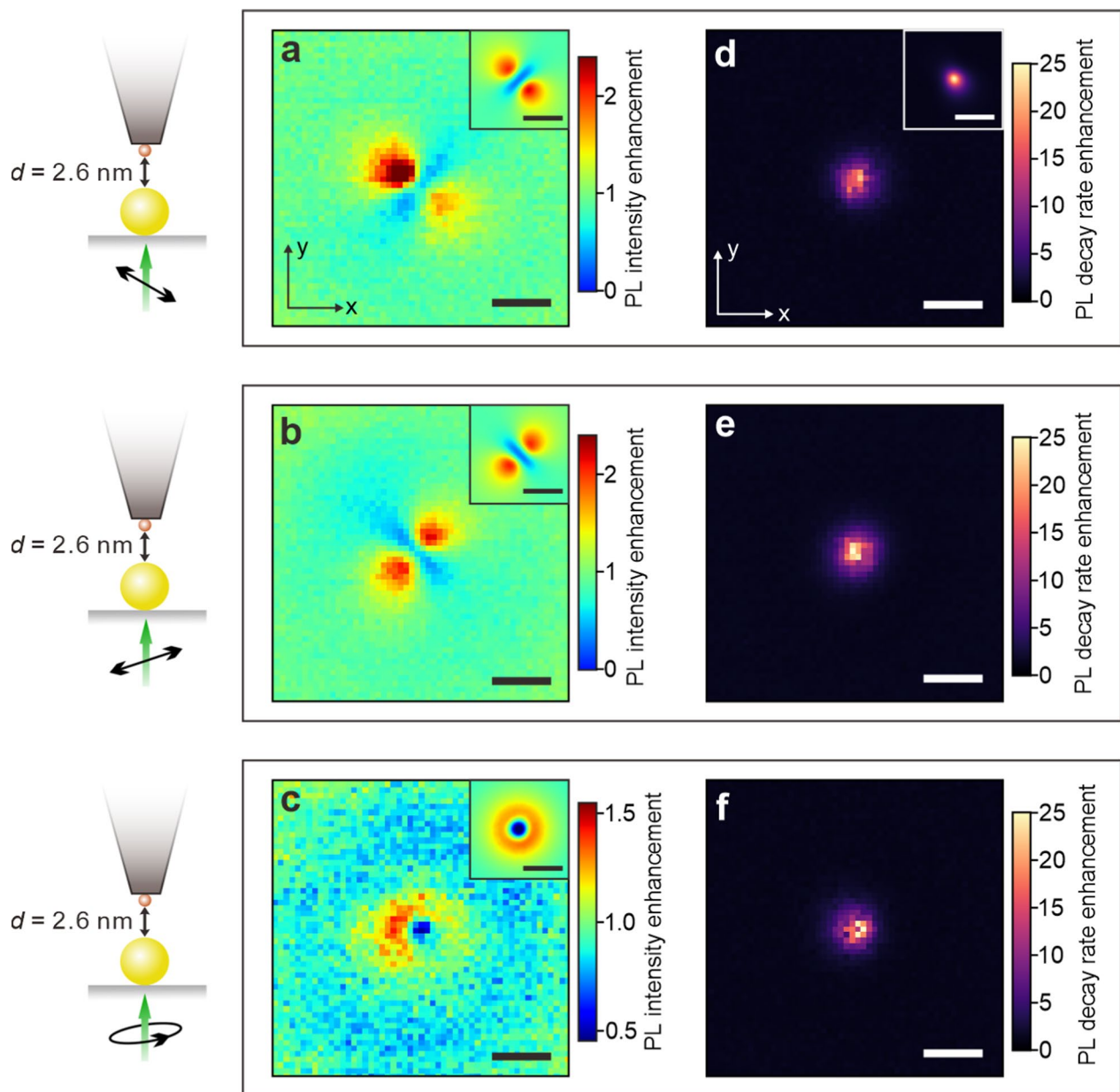


Fig. 2 Simultaneous mapping of the light intensity and LDOS on a single gold nanosphere (AuNS). **a–c** Images of PL intensity enhancement obtained via SEON on an AuNS under three types of in-plane excitation polarizations, i.e., two orthogonal linear polarizations (**a**, **b**) and a circular polarization (**c**). The vertical QD–AuNS distance d is 2.6 nm. **d–f** The corresponding images of PL decay rate enhancement, showing their independence of the excitation polarization. The insets display the simulated distributions. The QD probe's dipole parameters $(R, \theta, \phi) = (2.8, 77^\circ, 52^\circ)$. Scale bars, 100 nm

4.03 MHz repetition rate) is launched from the glass substrate side and generates an enhanced near field pattern according to the polarization of the illumination. We scan the QD over the AuNS at a constant height from the substrate. The QD–substrate distance can be well-controlled and kept within 1 nm precision (Supplementary Material Section 8). PL intensity and decay rate enhancement

mappings are constructed by averaging multiple scanning frames (Supplementary Table 1 lists scanning parameters for all mappings presented in the manuscript). The “PL intensity enhancement” and “PL decay rate enhancement” are dimensionless normalized ratios I/I^0 and γ/γ^0 , respectively. Here I^0 and γ^0 respectively denote the

averaged intensity and decay rate of the same QD probe measured in a reference region far away from the AuNS.

In Fig. 2, measurements are performed for three types of excitation polarizations, including two orthogonal in-plane linear polarizations and an in-plane circular polarization. Our measurements show that the fluorescence intensity distributions are distinct for the three types of illuminations while the LDOS mappings keep consistent. For in-plane linear polarizations, the enhanced near field spots occur on two sides of the AuNS along the polarization direction (Fig. 2a, b)—a signature of the AuNS's in-plane dipolar plasmonic resonance—in good agreement with the simulated results (insets at the upper right corner). Excitation with an in-plane circular polarization generates a donut-shaped intensity profile (Fig. 2c)—arising from uniform excitation of the dipolar plasmonic mode across all in-plane directions. Similar intensity profiles are observed for a variety of QD-substrate distances (Supplementary Material Section 9). Contrary to the PL intensity images, the corresponding decay rate mappings are nearly identical across all polarizations, all showing only one enhancement spot above the AuNS as depicted in Fig. 2d, e, f.

These experiments validate SEON's capability for simultaneous dual-parameter sensing around the same structure, which will enable correlative analysis to reveal intricate physical phenomena at the deep nanoscale. Such capability is unattainable with single-parameter-sensing techniques, such as conventional SNOM techniques (due to the lack of LDOS sensitivity) or electron-beam-based spectroscopies (due to the inaccessibility of optical fields).

2.3 Revealing interplay of fluorescence enhancement and quenching

We further perform SEON on an AuNS (79 nm in diameter) under out-of-plane linearly polarized excitation, achieved by launching transverse magnetic (TM)-polarized illumination in total internal reflection (TIR) configuration, as schematically illustrated in Fig. 3a. Figure 3b, c present PL intensity and decay rate mappings at a series of QD-AuNS distances. The PL intensity mappings now shows a single enhancement spot with an additional arc-shaped fringe. This fringe arises from the interference between the evanescent field of the TIR illumination and the scattered field from the AuNS. Reproducible via numerical simulation, this fringe highlights the probe's ability to resolve subtle features of the nanoscale light field with unprecedented fidelity.

The PL intensity and PL decay rate enhancement factors directly above the AuNS are extracted from the mappings and plotted as functions of QD-AuNS distance in Fig. 3d, e respectively. The decay rate increases

monotonically with decreasing distances. In contrast, the PL intensity reaches its maximum at a finite distance, which echoes previous study [48]. This is a demonstration of the interplay of fluorescence enhancement and quenching revealed by SEON at varied heights. At very short QD-AuNS distances, coupling to high-order multipolar plasmonic modes (with extremely low radiation efficiency) becomes dominant; thus, suppression of the emitter's radiation efficiency outweighs the excitation enhancement, leading to a decrease in PL intensity. Note that the couplings to highly radiative dipolar modes and poorly radiative multipolar modes both contribute to the LDOS, causing the monotonical increase of the decay rate as the probe approaches the AuNS.

2.4 Spatial resolution of the SEON

Given that both absorption and emission of the QD probe predominantly occur within the 3 nm CdSe core [44] and the probe can be controlled at sub-nanometer precision with piezoelectric nanopositioners, high spatial resolution is expected. To explicitly characterize the resolving capability of SEON, we quantitatively evaluated the vertical and lateral resolutions from the measurements presented in Fig. 3.

The field above the AuNS decays sharply with increasing distance from the AuNS surface in the vertical direction. Therefore, characterizing the drastic changes of this sharply decaying light field within a few nanometers can readily demonstrate high vertical resolution. As shown in Fig. 3e, when the QD-AuNS distance decreases from 6 to 2 nm, SEON clearly captures a surge in the decay rate enhancement factor: from 9.1 to 25.3, corresponding to a 178% change over a 4 nm vertical range. This significant signal distinction confirms that vertical positions separated by only 4 nm can be reliably resolved, demonstrating a vertical resolution better than 4 nm. This is further validated by the successful capture of a complete “enhancement-to-quenching” transition of PL intensity over the same 4 nm vertical interval (Fig. 3d), reflecting the probe's ability to resolve subtle nanoscale vertical variations.

For lateral resolution, since the field distribution above the AuNS is relatively broad in the lateral direction, we adopt a convolution fitting method (detailed in Supplementary Material Section 10)—a widely accepted strategy for resolution quantification in scanning probe optical nanoscopy. Specifically, the experimentally measured lateral profile is treated as the convolution of the simulated “true” near-field profile and the probe's lateral response function (approximated as a Gaussian, whose full-width at half-maximum (FWHM) denotes

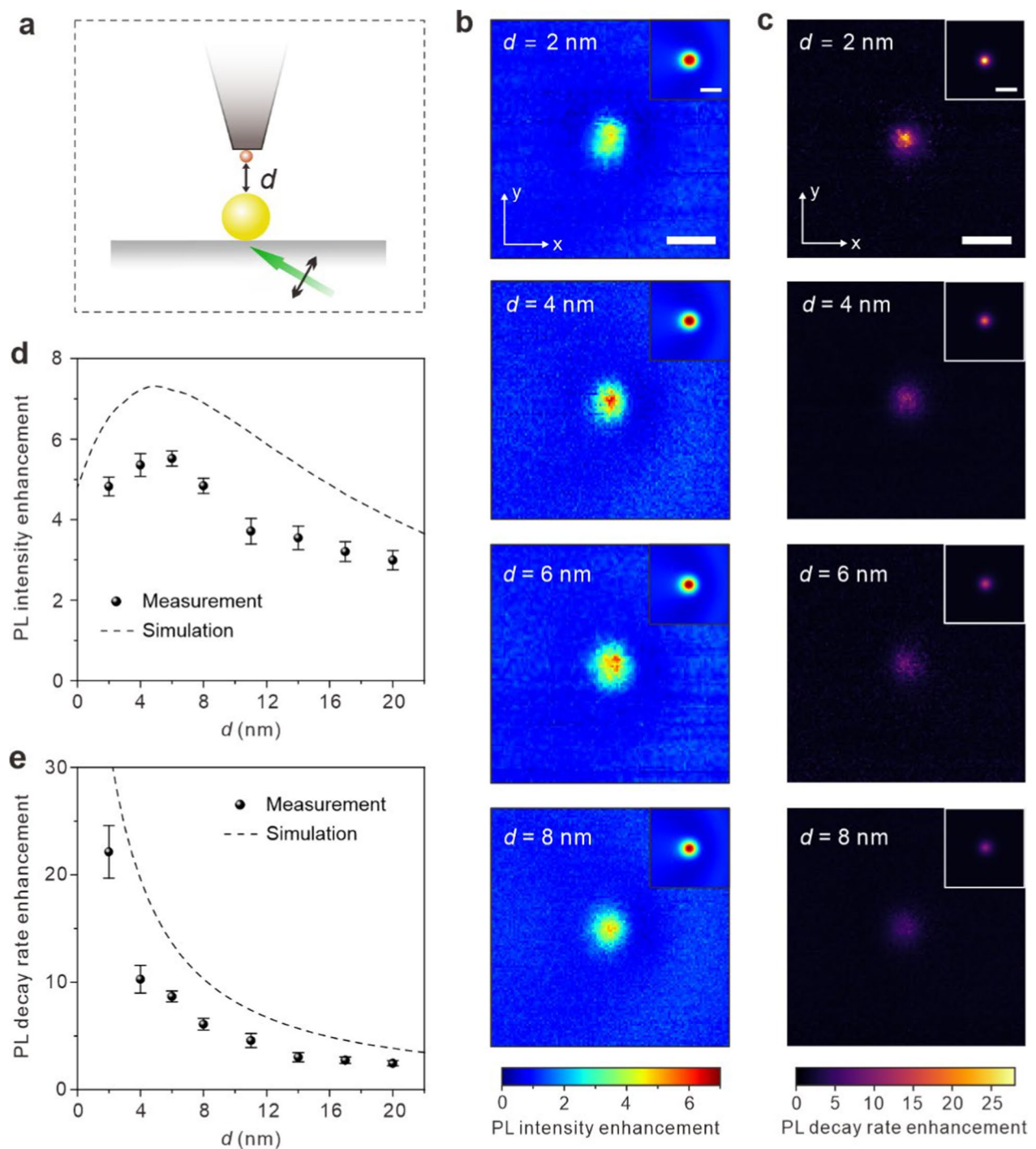


Fig. 3 Scanning-exciton optical nanoscopy on a single gold nanosphere (AuNS) under vertically polarized excitation. **a** Illustration of the total-internal-reflection (TIR) illumination scheme with a transverse magnetic (TM) polarization. **b, c** PL intensity (**b**) and decay rate (**c**) enhancement mappings at a series of QD-AuNS distances ($d = 2, 4, 6, \text{ and } 8 \text{ nm}$). The insets display the simulated distributions. The QD probe's dipole parameters (R, θ, ϕ) = (1.4, 37°, 205°). Scale bars, 100 nm. **d, e** PL intensity (**d**) and decay rate (**e**) enhancement factors directly above the AuNS as a function of QD-AuNS distance. Each data point represents the mean value of the top 10 highest-value pixels in each mapping. Error bars denote the standard deviation (1 σ). The dashed curves are the simulated results

the lateral resolution). Fitting the experimental decay rate profile acquired 2 nm above the AuNS (top panel of Fig. 3c) to the convolved simulated profile yields a Gaussian FWHM of 4 nm (Supplementary Material Section 10), which defines the lateral resolution of SEON.

2.5 Revealing nanoscale coupling physics in a plasmonic trimer

We next apply SEON on a cluster of three nearly touching AuNSs which are assembled via AFM nanomanipulation [49, 50] (Supplementary Material Section 11), to uncover previously inaccessible nanoscale coupling physics. Figure 4a displays the AFM topographic image of the plasmonic trimer and the cross-sectional plot along the dashed line tells the heights of AuNSs #1, #2 and #3 are 78 nm, 79 nm and 82 nm, respectively. The SEON measurement is performed under the out-of-plane polarized illumination as in Fig. 3. During the scan, the QD-substrate distance is maintained at 86 nm, i.e., the vertical distances between the QD and AuNSs #1, #2 and #3 are 8 nm, 7 nm and 4 nm, respectively. The PL intensity mapping (Fig. 4b) displays three well-separated nanoscale hot spots with different enhancement factors, superimposed with arc-shaped fringes. The PL decay rate mapping (Fig. 4c) also exhibits three hot spots, but without fringes. This indicates that the fringes in the intensity mapping is

caused by interference between the illumination and the scattering.

Notably, while both the intensity and decay rate mappings show three hot spots, the magnitude ranking of these hot spots differs significantly. A detailed analysis of this discrepancy shall reveal the complex interplay of competing physical mechanisms. In the PL decay rate mapping (Fig. 4c), the three hot spots above AuNSs #1, #2 and #3 exhibit monotonically increasing values, which is consistent with the decreasing QD-AuNS distances. Interestingly, in the PL intensity mapping (Fig. 4b), the intensity rankings of the three hot spots are completely different. The PL intensity hot spot above AuNS #1, which has the largest QD-AuNS distance and thus the smallest excitation enhancement, exhibits a higher fluorescence intensity than that above AuNS #3. This can be attributed to fluorescence quenching that occurs when the QD is too close to AuNS #3. More intriguingly, the PL intensity hot spot above the medium-size AuNS (#2) has the lowest value. This seemingly unexpected phenomenon is a result of the destructive interference between the multiple scatterings and the illumination. Our numerical simulation (Supplementary Material Section 12), which treats the AuNSs as perfect spheres and takes into account the mutual couplings, can qualitatively reproduce the distributions of PL intensity and decay rate, as displayed by the insets of Fig. 3b, c, respectively. This experiment

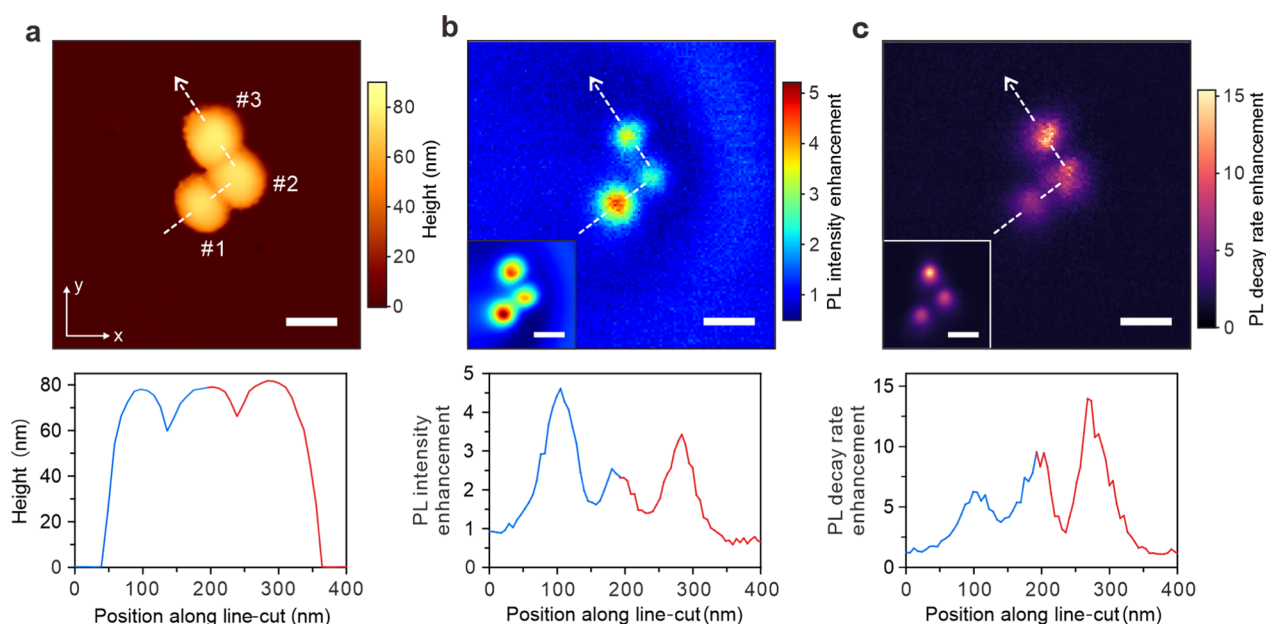


Fig. 4 Revealing nanoscale coupling mechanisms in a plasmonic trimer. Atomic-force microscopy (AFM) topographic mapping (a), PL intensity enhancement mapping (b), and decay rate enhancement mapping (c) for a plasmonic trimer consisting of three nearly touching AuNSs. The measured heights of AuNSs #1, #2 and #3 are 78 nm, 79 nm and 82 nm, respectively. Cross-sectional plots along the dashed lines are shown below the mappings. The insets in (b) and (c) are the simulated distributions. Scale bars, 100 nm. The QD probe's dipole parameters (R, θ, ϕ) = (2.9, 107°, 90°)

exemplifies SEON's ability to reveal intricate nanoscale coupling physics, involving multiple scatterings and their interference with the illumination, as well as spontaneous emission enhancement and quenching of an emitter. Such level of mechanistic disentanglement is impossible with single-parameter-sensing techniques.

2.6 Dual-parameter mapping on a waveguide-interfaced photonic-crystal nanocavity

Photonic-crystal nanocavities are essential components in both nanophotonics and quantum photonics due to their ability to confine light in subwavelength mode volumes with high quality factors, which enable low-threshold lasing, Purcell enhancement of spontaneous emission, and cavity quantum electrodynamics (cQED) experiments [7, 8]. Integrating photonic nanocavities with waveguides to form photonic circuits offers

flexible on-chip control over light emission, routing, and interaction at the single-photon level. To uncover the underlying physics and drive progress in this exciting field, it is crucial to experimentally map the light field and LDOS around the photonic-crystal nanocavities with nanometer resolution.

Here we demonstrate the first mappings by performing SEON on a photonic-crystal nanobeam cavity coupled to a single-mode waveguide [51], as illustrated in Fig. 5a. The photonic-crystal nanocavity consists of a suspended Si_3N_4 nanobeam with a periodic array of air holes with gradually decreasing sizes (see Supplementary Material Section 13 for fabrication). Figure 5b displays the AFM topographic mapping for the cavity. As inferred from an isolated peak in a measured transmission spectrum shown in Fig. 5c, the cavity should support a resonant cavity mode around 605 nm—a wavelength matched to

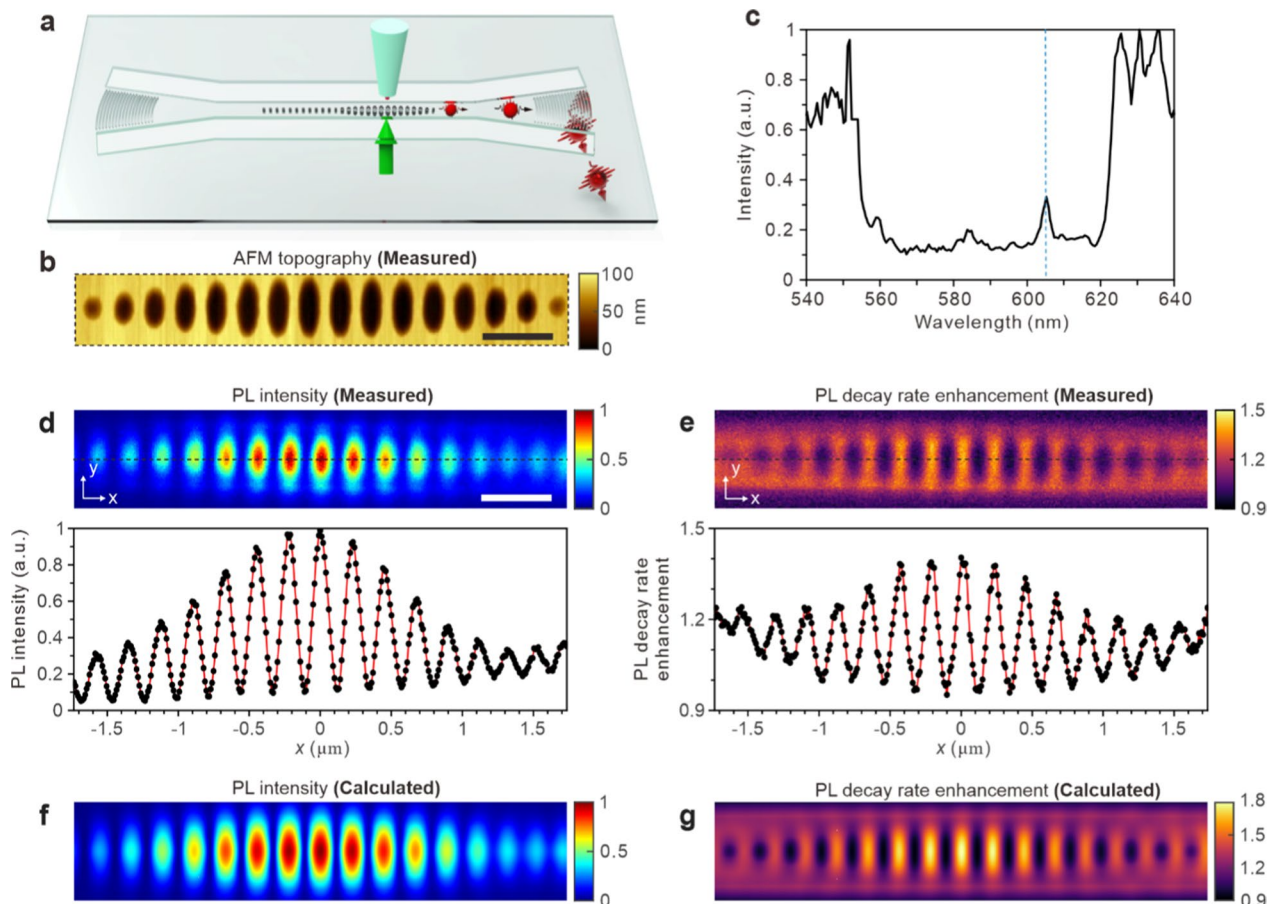


Fig. 5 Dual-parameter mapping on a waveguide-interfaced photonic-crystal nanocavity. **a** Layout of the photonic-crystal nanobeam cavity structure and a schematic illustration of the SEON experiment. **b** Atomic-force microscopy topographic mapping for the nanocavity. **c** The transmission spectrum of the waveguide-interfaced photonic-crystal nanocavity. The blue dashed line indicates the resonance of the fundamental mode of the nanocavity. **d** PL intensity mapping for the nanocavity. Cross-sectional plot along the dashed line is shown below the mapping. **e** PL decay rate enhancement mapping for the nanocavity. Cross-sectional plot along the dashed line is shown below the mapping. The QD-sample distance is kept to 30 nm. **f** Simulated PL intensity distribution for the nanocavity. **g** Simulated PL decay rate enhancement mapping for the nanocavity. Scale bars, 500 nm. The QD probe's dipole parameters (R, θ, ϕ) = (2.2, 93°, 101°)

the emission peak of our QD probe to ensure efficient QD-cavity coupling for high-fidelity mapping. For SEON measurement, the excitation beam denoted by a green arrow in Fig. 5a is launched vertically from the substrate side to excite the QD probe. The QD is maintained at a constant height of 30 nm above the sample surface and sample scan is applied here. There are two collection channels for QD fluorescence: (1) the grating coupler channel, where a fraction of QD fluorescence enters the nanocavity, unidirectionally funnels to the waveguide, and ultimately couples out of the chip via a grating coupler; (2) the free space channel, where the remaining fluorescence is radiated into free space and directly collected by a lens. These two channels are separately registered via TCSPC and analyzed (see Supplementary Material Section 3.2 for the full optical setup).

The grating coupler channel exhibits strongly position-dependent collection efficiency, arising from spatially varying QD-cavity coupling—with high efficiency at cavity mode hotspots and near-background levels elsewhere. Figure 5d represents the PL intensity mapping from the grating coupler channel, which reflects the spatial profile of the cavity mode. To ensure robust decay rate analysis across the entire scanning area, we combine signals from both channels to generate the PL decay rate enhancement mapping (Fig. 5e).

Notably, both the PL intensity mapping and the PL decay rate enhancement mapping are in excellent agreement with simulations (Fig. 5f, g), and the experimental observations are reproducible (Supplementary Material Section 14). The measured PL intensity distribution closely matches the spatial profile of the resonant nanocavity mode, as the signal reflects the spatially varying coupling efficiency of an emitter into the very resonant mode. In contrast, the measured PL decay rate enhancement distribution reveals a more complex physical picture. It includes contributions not only from the nanocavity resonant mode but also from the non-resonant local dielectric background owing to the dielectric constant contrast between the silicon–nitride backbone and the air holes. The latter is a direct manifestation of the LDOS modification by its immediate material's property. Thus, our measured LDOS mapping displays the characteristics of the nanocavity mode with the material structural pattern superimposed. These results highlight the unique capability of SEON to characterize the resonant mode of the waveguide-interfaced photonic-crystal nanocavity as well as its coupling with a nearby quantum emitter. The reproducibility of the experimental measurements and their consistency with the simulations show again the high fidelity and robustness of the SEON mappings for relatively large integrated photonic systems.

3 Discussions

We have demonstrated scanning-exciton optical nanoscopy for simultaneous correlative mapping of light intensity and LDOS with unprecedented fidelity and a spatial resolution of few nanometers. The dual-parameter sensing capability has been faithfully validated with a model plasmonic structure, where distinct light-intensity patterns with consistent LDOS patterns are observed under varied illumination configurations. We have performed scanning-exciton optical nanoscopy on a plasmonic trimer and a waveguide-interfaced photonic-crystal nanocavity system, showcasing its unique ability to uncover the intricate nanoscale physics with simultaneous correlative knowledge of the light intensity and LDOS, which is inaccessible with existing techniques. This technique outperforms earlier single-emitter scanning probe microscopies based on nanodiamonds containing single nitrogen-vacancy centers and organic microcrystals embedded with single dye molecules, by virtue of a suite of advantages. These include near-unity quantum yield (more sensitive to LDOS change), orders-of-magnitude larger absorption cross-section (lower background), small size (better accessibility and less distortion), and a narrow tunable emission (higher spectral resolution and adaptability). Moreover, our single-QD probes also possess outstanding photostability in air.

Our scanning-exciton optical nanoscopy establishes a foundational platform for exploring and scrutinizing light-matter interactions at the deep nanoscale, with wide-ranging implications for functional nanomaterials, quantum optics, integrated photonics, and nanoplasmonics. It opens the door to a range of experiments that require delicate nano-controlled studies, such as controlled Förster energy transfer or dipole–dipole coupling between single quantum systems [52], and coupling between an emitter and extremely localized optical field [53], pushing the boundaries of exploring light-matter interactions at the single-emitter level. The technique could be possibly used for direct experimental visualization of atomic-scale field localization [54]. An immediate extension of the technique would be to develop a reflection-mode SEON, i.e., excitation and collection through the same tapered fiber, by overcoming challenges such as the autofluorescence background from the fiber and the low collection efficiency of the emission. Further extensions could integrate multicolor QD probes for wavelength-multiplexed LDOS mapping, or combine this technique with ultrafast spectroscopy to resolve dynamical processes in quantum materials.

Supplementary Information

The online version contains supplementary material available at <https://doi.org/10.1186/s43593-026-00128-x>.

Supplementary Material 1.

Acknowledgements

We gratefully acknowledge financial support from the National Natural Science Foundation of China (Grant Number U25D9003, 12525414, 62235006, 12374349, 22173079, 22105011, U24A20318, 62135011), the Fundamental Research Funds for the Central Universities (2023BR003, 2024BRB002), and the Hubei Provincial Talent Program. X.-W. C and H.Q. thank Xiaogang Peng for his continuous support and encouragement.

Author contributions

X.-W.C. conceived the research. X.-W.C., H.Q. and J.T. designed the experiment. Z.W., J.H., J.T., X.-W.C., W.X., H.C. and J.C. performed the scanning-exciton optical nanoscopy experiments and analyzed the data. X.H. synthesized and characterized the quantum dots under the guidance of H.Q. W.L. fabricated the photonic-crystal nanobeam cavity structure under the guidance of Y.S. Z.T., J.H., J.T. and X.-W.C. performed numerical calculations and analysis. All authors contributed to discussion of the results. The paper was written by X.-W.C. and J.T. and was commented by H.Q. and X.H. The project was supervised by X.-W.C., J.T. and H.Q.

Funding

National Natural Science Foundation of China, U25D9003, Xue-Wen Chen, 12525414, Xue-Wen Chen, 62235006, Xue-Wen Chen, 12374349, Jianwei Tang, 22173079, Haiyan Qin, 22105011, Xiaoqi Hou, U24A20318, Yaocheng Shi, 62135011, Yaocheng Shi, Fundamental Research Funds for the Central Universities, 2023BR003, Xue-Wen Chen, 2024BRB002, Jianwei Tang, Hubei Provincial Talent Program, Jianwei Tang.

Data availability

The data that support the findings of this study are available within the article and its Supplementary Information, and from the corresponding authors upon request.

Declarations

Competing interests

X.-W.C., W.X., and Z.W. filed a patent based on the results presented in this work. The remaining authors declare no competing interests.

Received: 27 October 2025 Revised: 29 January 2026 Accepted: 17 February 2026

Published online: 18 March 2026

References

- H. Benisty, J.-J. Greffet, P. Lalanne, *Introduction to Nanophotonics* (Oxford University Press, 2022)
- Y. Yang, Y. Jeon, Z. Dong, J.K.W. Yang, M. Haddadi Moghaddam, D.-S. Kim, D.K. Oh, J. Lee, M. Hentschel, H. Giessen, D. Kang, G. Kim, T. Tanaka, Y. Zhao, J. Bürger, S.A. Maier, H. Ren, W. Jung, M. Choi, G. Bae, H. Chen, S. Jeon, J. Kim, E. Lee, H. Kang, Y. Park, D. Du Nguyen, I. Kim, P. Cencillo-Abad, D. Chanda, X. Jing, N. Liu, I.V. Martynenko, T. Liedl, Y. Kwak, J.-M. Nam, S.-M. Park, T.W. Odom, H.-E. Lee, R.M. Kim, K.T. Nam, H. Kwon, H.-H. Jeong, P. Fischer, J. Yoon, S.-H. Kim, S. Shim, D. Lee, L.A. Pérez, X. Qi, A. Mihi, H. Keum, M. Shim, S. Kim, H. Jang, Y.S. Jung, C. Rossner, T.A.F. König, A. Fery, Z. Li, K. Aydin, C.A. Mirkin, J. Seong, N. Jeon, Z. Xu, T.G. Hu, H. Kwon, H. Jung, H. Alijani, I. Aharonovich, J. Kim, J. Rho, Nanofabrication for Nanophotonics. *ACS Nano* **19**, 12491–12605 (2025)
- J.J. Baumberg, J. Aizpurua, M.H. Mikkelsen, D.R. Smith, Extreme nanophotonics from ultrathin metallic gaps. *Nat. Mater.* **18**, 668–678 (2019)
- D.G. Baranov, M. Wersall, J. Cuadra, T.J. Antosiewicz, T. Shegai, Novel nanostructures and materials for strong light matter interactions. *ACS Photonics* **5**, 24–42 (2018)
- A. González-Tudela, A. Reiserer, J.J. García-Ripoll, F.J. García-Vidal, Light-matter interactions in quantum nanophotonic devices. *Nat. Rev. Phys.* **6**, 166–179 (2024)
- D.D.A. Clarke, O. Hess, Near-field strong coupling and entanglement of quantum emitters for room-temperature quantum technologies. *Photonix* **5**, 33 (2024)
- S. Noda, M. Fujita, T. Asano, Spontaneous-emission control by photonic crystals and nanocavities. *Nat. Photonics* **1**, 449–458 (2007)
- Vučковиć, J. in *Quantum Optics and Nanophotonics* (eds Claude Fabre, Vahid Sandoghdar, Nicolas Treps, & Leticia F. Cugliandolo) (Oxford University Press, 2017).
- A.W. Elshaari, W. Pernice, K. Srinivasan, O. Benson, V. Zwiller, Hybrid integrated quantum photonic circuits. *Nat. Photonics* **14**, 285–298 (2020)
- R. Uppu, L. Midolo, X. Zhou, J. Carolan, P. Lodahl, Quantum-dot-based deterministic photon-emitter interfaces for scalable photonic quantum technology. *Nat. Nanotechnol.* **16**, 1308–1317 (2021)
- E. Pelucchi, G. Fagas, I. Aharonovich, D. Englund, E. Figueroa, Q. Gong, H. Hannes, J. Liu, C.-Y. Lu, N. Matsuda, J.-W. Pan, F. Schreck, F. Sciarrino, C. Silberhorn, J. Wang, K.D. Jöns, The potential and global outlook of integrated photonics for quantum technologies. *Nat. Rev. Phys.* **4**, 194–208 (2022)
- N. Rotenberg, L. Kuipers, Mapping nanoscale light fields. *Nat. Photonics* **8**, 919–926 (2014)
- X. Chen, D. Hu, R. Mescall, G. You, D.N. Basov, Q. Dai, M. Liu, Modern scattering-type scanning near-field optical microscopy for advanced material research. *Adv. Mater.* **31**, 1804774 (2019)
- R. Hillenbrand, Y. Abate, M. Liu, X. Chen, D.N. Basov, Visible-to-THz near-field nanoscopy. *Nat. Rev. Mater.* **10**, 285–310 (2025)
- S.G. Stanciu, D.E. Tranca, G. Zampini, R. Hristu, G.A. Stanciu, X. Chen, M. Liu, H.A. Stenmark, L. Latterini, Scattering-type scanning near-field optical microscopy of polymer-coated gold nanoparticles. *ACS Omega* **7**, 11353–11362 (2022)
- S. Hasegawa, K. Imura, Three-dimensional visualization of chiral nano-optical field around gold nanoplates via scanning near-field optical microscopy. *Nano Lett.* **25**, 376–382 (2025)
- C. Chicanne, T. David, R. Quidant, J.C. Weeber, Y. Lacroute, E. Bourillot, A. Dereux, G. Colas des Francs, C. Girard, Imaging the local density of states of optical corrals. *Phys. Rev. Lett.* **88**, 097402 (2002)
- E.M. Purcell, Spontaneous emission probabilities at radio frequencies. *Phys. Rev.* **69**, 681 (1946)
- R. Carminati, A. Caze, D. Cao, F. Peragut, V. Krachmalnicoff, R. Pierrat, Y. De Wilde, Electromagnetic density of states in complex plasmonic systems. *Surf. Sci. Rep.* **70**, 1–41 (2015)
- A. Karabchevsky, A. Mosayyebi, A.V. Kavokin, Tuning the chemiluminescence of a luminol flow using plasmonic nanoparticles. *Light Sci. Appl.* **5**, e16164–e16164 (2016)
- N.G. Van Kampen, B.R.A. Nijboer, K. Schram, On the macroscopic theory of Van der Waals forces. *Phys. Lett. A* **26**, 307–308 (1968)
- J.-P. Mulet, K. Joulain, R. Carminati, J.-J. Greffet, Enhanced radiative heat transfer at nanometric distances. *Microscale Thermophys. Eng.* **6**, 209–222 (2002)
- A. Polman, M. Kociak, F.J. de García Abajo, Electron-beam spectroscopy for nanophotonics. *Nat. Mater.* **18**, 1158–1171 (2019)
- F.J. de García Abajo, M. Kociak, Probing the photonic local density of states with electron energy loss spectroscopy. *Phys. Rev. Lett.* **100**, 106804 (2008)
- A. Hörl, G. Haberer, A. Trügler, F.-P. Schmidt, U. Hohenester, G. Kothleitner, Tomographic imaging of the photonic environment of plasmonic nanoparticles. *Nat. Commun.* **8**, 37 (2017)
- R. Sapienza, T. Coenen, J. Renger, M. Kuttge, N.F. van Hulst, A. Polman, Deep-subwavelength imaging of the modal dispersion of light. *Nat. Mater.* **11**, 781 (2012)
- S.K. Sekatskii, V.S. Letokhov, Single fluorescence centers on the tips of crystal needles: first observation and prospects for application in scanning one-atom fluorescence microscopy. *Appl. Phys. B* **63**, 525–530 (1996)
- V. Sandoghdar, J. Mlynek, Prospects of apertureless SNOM with active probes. *J. Opt. A Pure Appl. Opt.* **1**, 523–530 (1999)

29. J. Michaelis, C. Hettich, J. Mlynek, V. Sandoghdar, Optical microscopy using a single-molecule light source. *Nature* **405**, 325–328 (2000)
30. S. Kuhn, C. Hettich, C. Schmitt, J.P.H. Poizat, V. Sandoghdar, Diamond colour centres as a nanoscopic light source for scanning near-field optical microscopy. *J. Microsc.* **202**, 2–6 (2001)
31. A. Cuche, A. Drezet, Y. Soneffraud, O. Faklaris, F. Treussart, J.F. Roch, S. Huant, Near-field optical microscopy with a nanodiamond-based single-photon tip. *Opt. Express* **17**, 19969–19980 (2009)
32. A.W. Schell, P. Engel, J.F.M. Werra, C. Wolff, K. Busch, O. Benson, Scanning Single Quantum Emitter Fluorescence Lifetime Imaging: quantitative analysis of the local density of photonic states. *Nano Lett.* **14**, 2623–2627 (2014)
33. R. Beams, D. Smith, T.W. Johnson, S.H. Oh, L. Novotny, A.N. Vamivakas, Nanoscale fluorescence lifetime imaging of an optical antenna with a single diamond NV center. *Nano Lett.* **13**, 3807–3811 (2013)
34. E. Betzig, R.J. Chichester, Single molecules observed by near-field scanning optical microscopy. *Science* **262**, 1422–1425 (1993)
35. A. Mohtashami, A.F. Koenderink, Suitability of nanodiamond nitrogen-vacancy centers for spontaneous emission control experiments. *New J. Phys.* **15**, 24 (2013)
36. A.P. Alivisatos, Semiconductor Clusters, Nanocrystals, and Quantum Dots. *Science* **271**, 933 (1996)
37. W. Xu, X. Hou, Y. Meng, R. Meng, Z. Wang, H. Qin, X. Peng, X.-W. Chen, Deciphering charging status, absolute quantum efficiency, and absorption cross section of multicarrier states in single colloidal quantum dots. *Nano Lett.* **17**, 7487–7493 (2017)
38. J. Puthenpurayil, O.H. Cheng, T. Qiao, D. Rossi, D.H. Son, On the determination of absorption cross section of colloidal lead halide perovskite quantum dots. *J. Chem. Phys.* **151**, 154706 (2019)
39. Y. Ebenstein, T. Mokari, U. Banin, Quantum-dot-functionalized scanning probes for fluorescence-energy-transfer-based microscopy. *J. Phys. Chem. B* **108**, 93–99 (2004)
40. N. Chevalier, M.J. Nasse, J.C. Woehl, P. Reiss, J. Bleuse, F. Chandezon, S. Huant, CdSe single-nanoparticle based active tips for near-field optical microscopy. *Nanotechnology* **16**, 613–618 (2005)
41. K. Matsuzaki, S. Vasant, H.W. Liu, A. Dutschke, B. Hoffmann, X. Chen, S. Christiansen, M.R. Buck, J.A. Hollingsworth, S. Gotzinger, V. Sandoghdar, Strong plasmonic enhancement of biexciton emission: controlled coupling of a single quantum dot to a gold nanocone antenna. *Sci. Rep.* **7**, 42307 (2017)
42. R. Jazi, T.P.L. Ung, P. Maso, G. Colas Des Francs, M. Nasilowski, B. Dubertret, J.P. Hermier, X. Quélin, S. Buil, Measuring the orientation of a single CdSe/CdS nanocrystal at the end of a near-field tip for the realization of a versatile active SNOM probe. *Phys. Chem. Chem. Phys.* **20**, 16444–16448 (2018)
43. C. Ropp, Z. Cummins, S. Nah, J.T. Fourkas, B. Shapiro, E. Waks, Nanoscale imaging and spontaneous emission control with a single nano-positioned quantum dot. *Nat. Commun.* **4**, 1447 (2013)
44. F. García-Santamaría, Y. Chen, J. Vela, R.D. Schaller, J.A. Hollingsworth, V.I. Klimov, Suppressed auger recombination in “giant” nanocrystals boosts optical gain performance. *Nano Lett.* **9**, 3482–3488 (2009)
45. P. Michler, A. Imamoglu, M.D. Mason, P.J. Carson, G.F. Strouse, S.K. Buratto, Quantum correlation among photons from a single quantum dot at room temperature. *Nature* **406**, 968–970 (2000)
46. G. Nair, J. Zhao, M.G. Bawendi, Biexciton quantum yield of single semiconductor nanocrystals from photon statistics. *Nano Lett.* **11**, 1136–1140 (2011)
47. S.A. Empedocles, R. Neuhauser, M.G. Bawendi, Three-dimensional orientation measurements of symmetric single chromophores using polarization microscopy. *Nature* **399**, 126–130 (1999)
48. P. Anger, P. Bharadwaj, L. Novotny, Enhancement and quenching of single-molecule fluorescence. *Phys. Rev. Lett.* **96**, 113002 (2006)
49. J. Tang, J. Xia, M. Fang, F. Bao, G. Cao, J. Shen, J. Evans, S. He, Selective far-field addressing of coupled quantum dots in a plasmonic nanocavity. *Nat. Commun.* **9**, 1705 (2018)
50. Y. Meng, D. Huang, H. Li, X. Feng, F. Li, Q. Liang, T. Ma, J. Han, J. Tang, G. Chen, X.-W. Chen, Bright single-nanocrystal upconversion at sub 0.5 W cm^{-2} irradiance via coupling to single nanocavity mode. *Nat. Photonics* **17**, 73–81 (2023)
51. Q.M. Quan, P.B. Deotare, M. Loncar, Photonic crystal nanobeam cavity strongly coupled to the feeding waveguide. *Appl. Phys. Lett.* **96**, 203102 (2010)
52. A.R. Clapp, I.L. Medintz, H. Mattoussi, Förster resonance energy transfer investigations using quantum-dot fluorophores. *ChemPhysChem* **7**, 47–57 (2006)
53. H. Groß, J.M. Hamm, T. Tufarelli, O. Hess, B. Hecht, Near-field strong coupling of single quantum dots. *Sci. Adv.* **4**, eaar4906 (2018)
54. Y.-H. Ouyang, H.-Y. Luan, Z.-W. Zhao, W.-Z. Mao, R.-M. Ma, Singular dielectric nanolaser with atomic-scale field localization. *Nature* **632**, 287–293 (2024)