

Quantum-precision-spectroscopy metrology of vacuum Rabi splitting in plasmon-single-emitter strong coupling

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Strong coupling between plasmonic nanostructures and single emitters enables the generation
of hybrid plexcitonic states at the single-photon level under ambient conditions, which is not
only crucial for investigating fundamental phenomena in plasmonic cavity quantum
electrodynamics but also serves as a cornerstone for the development of room-temperature
quantum technologies. However, accurately probing the key parameters of vacuum Rabi
splitting (VRS) and coupling constant g in such intrinsically lossy, non-Hermitian single-
emitter strong coupling systems remains challenging, severely hindering their applications.
This is primarily attributed to substantial plasmonic dissipation and the consequent damping
linewidth mismatch between plasmons and emitters. Here we demonstrate that these challenges
can be overcome by exceptional point-assisted photonic substrate engineering that spectrally
narrows the plasmon mode and enables precise linewidth matching with the emitter. Using a
deterministic plasmon-single-exciton strong coupling platform, we achieve quantum-

precision-spectroscopy metrology of the VRS and at room temperature, characterized by the measured spectral splitting (SS) faithfully reflecting the intrinsic quantum eigenlevel splitting, with vanishing or perturbatively small systematic error. We attain a relative measurement error as low as $\sim 0.86\%$, representing a 40-fold improvement compared with conventional SiO_2 substrates. Meanwhile, the g value of this strong coupling system is also directly measured with a tiny error of $\sim 0.60\%$, indicating a roughly 20-fold improvement in accuracy. Our results reveal a previously inaccessible regime of plasmon-single-emitter strong coupling and establish a powerful route towards room-temperature quantum-precision-spectroscopy metrology operating at the single-photon level.

Introduction

A single emitter strongly coupled to single-mode fields has been central subject of cavity quantum electrodynamics (cQED) and quantum optics, serving as the foundation for advanced quantum applications in many quantum information processing tasks¹⁻⁴, including quantum computing^{5,6}, quantum communications^{7,8}, and high-precision quantum measurement⁹. In the strong coupling regime, energy is coherently exchanged between the emitter and the mode fields, resulting in mode hybridization and vacuum Rabi splitting (VRS)¹⁰⁻¹⁵. Experimentally, the single-emitter strong coupling in nanocavities serves as ideal platform for qubit manipulation, with the VRS typically probed using spectroscopy¹²⁻²¹ under the assumption that the observed spectral splitting (SS) equals the VRS. This assumption generally holds for conventional cQED systems, including atomic and solid-state platforms operated under cryogenic temperatures and ultrahigh vacuum, where the dissipations are negligible¹³⁻¹⁵.

In recent years, plasmonic nanocavities have advanced strong light-matter interactions into the ambient regime²¹⁻²⁶, owing to their exceptional advantages in ultrasmall mode volume and strong spatial localization²⁷. Notably, sustained research efforts have progressively reduced the number of emitters involved in strong coupling—from the initial hundreds of thousands²⁸⁻³² to only a few³³⁻³⁷, and more recently down to the single-emitter^{16-18,38-41} and even single-exciton level^{19,42,43}. Despite these remarkable advancements, theoretical investigations in plasmon-single-emitter strong coupling systems have demonstrated that the common assumption of the measured SS accurately representing the VRS is generally invalid^{44,45}. This discrepancy arises from the heavy losses of the plasmon mode, the pronounced damping mismatch between the

plasmon and exciton, and the relatively small g value between the two coupling subsystems. As a result, precise spectroscopic detection of the intrinsic VRS in plasmon-single-exciton strong coupling systems remains highly challenging. For instance, experiments by Zhong *et al.* demonstrated that the relative deviation between the measured SS and the intrinsic VRS in a plasmon-few-exciton coupling system can reach as high as 100.6%³⁴. Additionally, our theoretical investigations⁴⁴ reveal that apparent strong-coupling signatures, such as the SS, can be clearly observed even in the absence of genuine VRS in plasmon-single-emitter coupling systems, highlighting the phenomenon of “pseudo-strong coupling”. These findings underscore that achieving precise and reliable spectroscopic measurements of the intrinsic VRS in single-exciton strong coupling systems is both fundamentally important and urgently required for advancing quantum nanophotonics.

In this work, we suggest and develop a quantum-precision-spectroscopic metrology strategy for accurately probing the VRS and g in plasmon-single-exciton strong coupling systems at room temperature. In intrinsically lossy light-matter platforms, spectroscopic fidelity is governed not only by the interaction strength but also by the non-Hermitian nature of the hybridized modes, where strong plasmonic dissipation and linewidth mismatch distort the spectral response and obscure genuine VRS. Within this non-Hermitian framework, the exceptional point (EP) defines the critical boundary between weak and strong coupling, at which both eigenfrequencies and linewidths coalesce, rendering the spectral response maximally sensitive to true light-matter hybridization. Using a deterministic single-exciton strong coupling platform based on an Au nanodimer cavity, we show that the photonic substrate engineering (Si/SiO₂) spectrally localizes the plasmon mode, substantially compresses its damping linewidth and aligns it with exciton. This shifts the EP condition to a markedly reduced critical interaction strength, from 27.75 meV to 4.75 meV. Operating in this EP-proximal regime restores quantitative correspondence between the measured SS and the intrinsic VRS, reducing the relative measurement error from approximately 35.1% to 0.86%. Meanwhile, the coupling constant g has also been directly and precisely measured with a tiny error of ~0.60%. Our results establish EP-assisted photonic substrate engineering as a viable route to quantum-precision-spectroscopy metrology of VRS and g , opening pathways toward room-temperature quantum precision measurement at the single-photon level.

Principles of quantum-precision-spectroscopy metrology of VRS

In cQED, VRS arises from the strong coupling between a single two-level quantum emitter and a single-mode field. When multiple emitters participate, collective Dicke states⁴⁶ and nonlinear effects emerge, obscuring the intrinsic light-matter interaction and preventing a faithful reflection of the fundamental VRS via SS measurements. Therefore, deterministic single-emitter strong coupling is a prerequisite for eliminating multi-emitter effects and accurately restoring the genuine VRS. To meet this requirement, we construct a deterministic plasmon-single-exciton strong coupling system consisting of a methylene blue (MB) molecule, hosting a single exciton, strongly coupled to an Au nanodimer cavity (Au NDC), as shown in Fig. 1a. Despite this deterministic single-exciton strong coupling platform, probing the VRS via SS reveals pronounced discrepancies between the measured SS and the VRS (Figs. 1a, b). This deviation is not an experimental artifact but an intrinsic feature of lossy, non-Hermitian plasmon-exciton systems—a phenomenon that has remained largely overlooked. We attribute this mismatch primarily to three factors: (1) strong plasmonic damping (Γ_p), (2) a substantial damping linewidth mismatch between the plasmon mode (Γ_p) and the exciton (Γ_e), and (3) a relatively small g value between the two coupling subsystems⁴⁷.

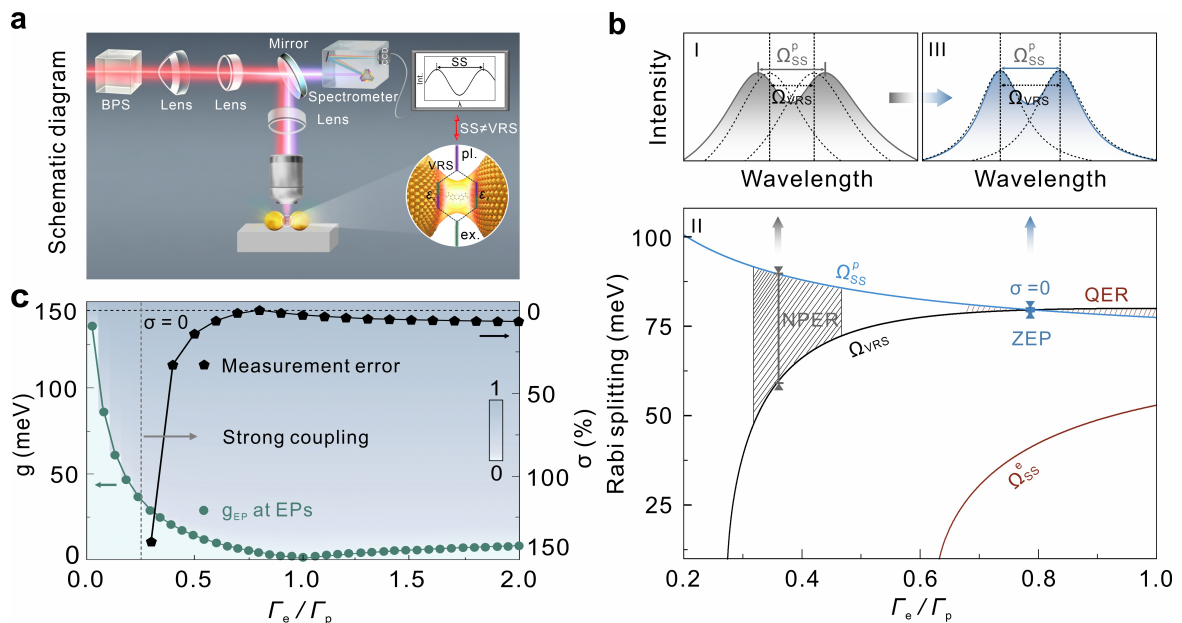


Fig. 1 | Non-Hermitian origins of measurement errors in probing VRS. **a**, Schematic diagram of spectroscopic probing of the VRS in a plasmon-single-emitter strong coupling system on a conventional substrate. **b**, Intrinsic VRS (Ω_{VRS} , black line) and the apparent SS

detected from the plasmon absorption channel ($\Omega_{\text{SS}}^{\text{p}}$, blue line) and the emitter emission channel ($\Omega_{\text{SS}}^{\text{e}}$, brown line), plotted as functions of the damping ratio Γ_e/Γ_p . The black shaded region denotes the experimentally relevant parameter space ($20\% \leq \sigma \leq 100\%$), corresponding to a non-perturbative error regime (NPER), whereas the red shaded region indicates a high-precision quasi-exact regime (QER; $0 < \sigma \leq 3\%$). ZEP denotes the zero-error point at which the measured SS coincides with the VRS. **c**, Real parts of the hybridized eigenenergies as functions of the g value at different ratios of Γ_e/Γ_p . Green markers indicate the critical interaction strengths g_{EP} at EP for each case (Supplementary Fig. S1a). Black markers denote the corresponding relative measurement error σ evaluated at $g = 40$ meV and $\Gamma_e = 60$ meV.

In spectroscopy measurements of plasmon-single-emitter strong coupling, the SS observed in scattering—predominantly originating from the plasmonic channel and commonly probed in experiments—is given by⁴⁴ (Supplementary Note 1)

$$\Omega_{\text{SS}}^{\text{p}} = 2 \left\{ g(1 + \Gamma_e/\Gamma_p) \cdot (g^2 + \Gamma_e\Gamma_p/4)^{1/2} - (g^2 + \Gamma_e\Gamma_p/4) \cdot \Gamma_e/\Gamma_p \right\}^{1/2}. \quad (1)$$

Notably, this quantity differs substantially from the intrinsic VRS of the coupled quantum system at resonance ($\varepsilon_p = \varepsilon_e$), which is defined as,

$$\Omega_{\text{VRS}} = 2\sqrt{g^2 - [(\Gamma_p - \Gamma_e)/4]^2}, \quad (2)$$

and originates from the hybridized eigenenergies of $\varepsilon_{\pm} = (\varepsilon_p + \varepsilon_e)/2 - i(\Gamma_p + \Gamma_e)/4 \pm \Omega_{\text{VRS}}/2$ ^{14,44,48}. It can be seen that the SS observed from plasmon scattering channel $\Omega_{\text{SS}}^{\text{p}}$ is

substantially larger than that extracted from the emitter emission channel, $\Omega_{\text{SS}}^{\text{e}} = 2\sqrt{g^2 - (\Gamma_p^2 + \Gamma_e^2)/8}$ ⁴⁶, which has been confirmed in recent experiment⁴⁹. More disturbingly,

both these two SSs exhibit pronounced deviations from the intrinsic VRS (Fig. 1b), reflecting the intrinsically non-Hermitian nature in the lossy strong coupling systems. Encouragingly, as the plasmonic damping linewidth is compressed to approach the exciton linewidth ($\Gamma_p \rightarrow \Gamma_e$),

the EP shifts to a substantially reduced critical interaction strength⁴⁰, $g_{\text{EP}} = |\Gamma_p - \Gamma_e|/4$ (green markers in Fig. 1c). This shift simultaneously enhances the intrinsic Ω_{VRS} while suppressing $\Omega_{\text{SS}}^{\text{p}}$, leading to a pronounced reduction in the spectral measurement error of VRS, defined as

$\sigma = \frac{\Omega_{\text{SS}}^{\text{p}} - \Omega_{\text{VRS}}}{\Omega_{\text{VRS}}} \times 100\%$. Moreover, the relation $\Omega_{\text{SS}}^{\text{p}} \approx \Omega_{\text{VRS}} \approx 2g$ holds during this process,

offering a pathway to directly measure the coupling constant g by observing $\Omega_{\text{SS}}^{\text{p}}$.

For representative experimental parameters $g = 40$ meV and $\Gamma_e = 60$ meV, the measurement error of VRS approaches zero when the damping ratio is tuned to $\Gamma_e/\Gamma_p \approx 0.79$, defined as the zero-error point (ZEP; Fig. 1b). When Γ_e/Γ_p is further adjusted within the ranges $0.79 < \Gamma_e/\Gamma_p < 1.0$ and $0.68 \leq \Gamma_e/\Gamma_p < 0.79$, the relative error remains confined to $0 < \sigma < 3\%$, defining a quasi-exact regime (QER; red shaded region in Fig. 1b). This represents a dramatic improvement over the commonly accessed experimental parameter space, where $20\% \leq \sigma \leq 100\%$, corresponding to the non-perturbative error regime (NPER), in which the deviation between SS and VRS cannot be treated as a perturbative correction. Notably, in the QER of $0.79 < \Gamma_e/\Gamma_p < 1.0$, the g value can be directly and precisely measured using SS, where $g \approx \Omega_{SS}^p/2$.

In this work, quantum-precision-spectroscopy metrology refers to operation within the ZEP and QER, where the measured SS faithfully represents the intrinsic VRS of hybridized quantum levels, with vanishing or perturbatively small systematic error. To provide direct experimental guidance for achieving ultrahigh-precision VRS measurements using Ω_{SS}^p , we summarize the damping-linewidth matching conditions required to reach ZEP ($\sigma = 0$) for different combinations of g and Γ_e . This analysis reveals an exponential dependence of the optimal damping ratio Γ_e/Γ_p on g value (Supplementary Fig. S1b).

Large error of VRS measured in plasmon-single-exciton strong coupling

To experimentally demonstrate the limitations of conventional spectral readout of the VRS in plasmon-single-emitter strong coupling, we first construct a deterministic single-exciton strong coupling system with high fidelity in a plasmonic nanocavity, using host-guest chemistry and cucurbit[7]uril (CB[7])-mediated active self-assembly (Supplementary Note 2 and Supplementary Fig. S2). This method ensures: (1) integration of only a single molecule exciton within the plasmonic nanocavity, (2) precise placement of the exciton at the maximum electric-field location, and (3) perfect alignment of the excitonic transition dipole moment (μ_e) with the mode fields. These certainty features ensure strong coupling between the plasmonic field and a deterministic exciton, enabling reliable prediction of the intrinsic Ω_{VRS} and providing a benchmark for precisely evaluating measurement errors inferred from SS.

We next verify the single-exciton nature of the hybrid system using digital bianalyte surface-

enhanced Raman scattering (bi-SERS) experiments. Figure 2 summarizes bi-SERS measurements performed on individual Au NDCs using an atomic force microscope-Raman (AFM-Raman) system (Supplementary Fig. S3), with methylene blue (MB) and proflavine (PF) serving as molecular probes. MB and PF share similar chemical structures and bind to CB[7] in an identical manner, making bi-SERS a reliable approach for identifying single-molecule events in plasmonic nanocavities^{38,50,51}. Through host-guest chemistry⁵², individual CB[7]@single MB and CB[7]@single PF complexes are prepared and mixed with colloidal Au nanoparticles at low concentrations (averaging ~ 0.43 CB[7]@single MB and ~ 0.43 CB[7]@single PF per Au nanoparticle, with an MB/PF-to-CB[7] molar ratio of 1:2). Subsequent CB[7]-mediated active self-assembly leads to the formation of Au NDCs with single molecules embedded at the gap hotspot⁵³.

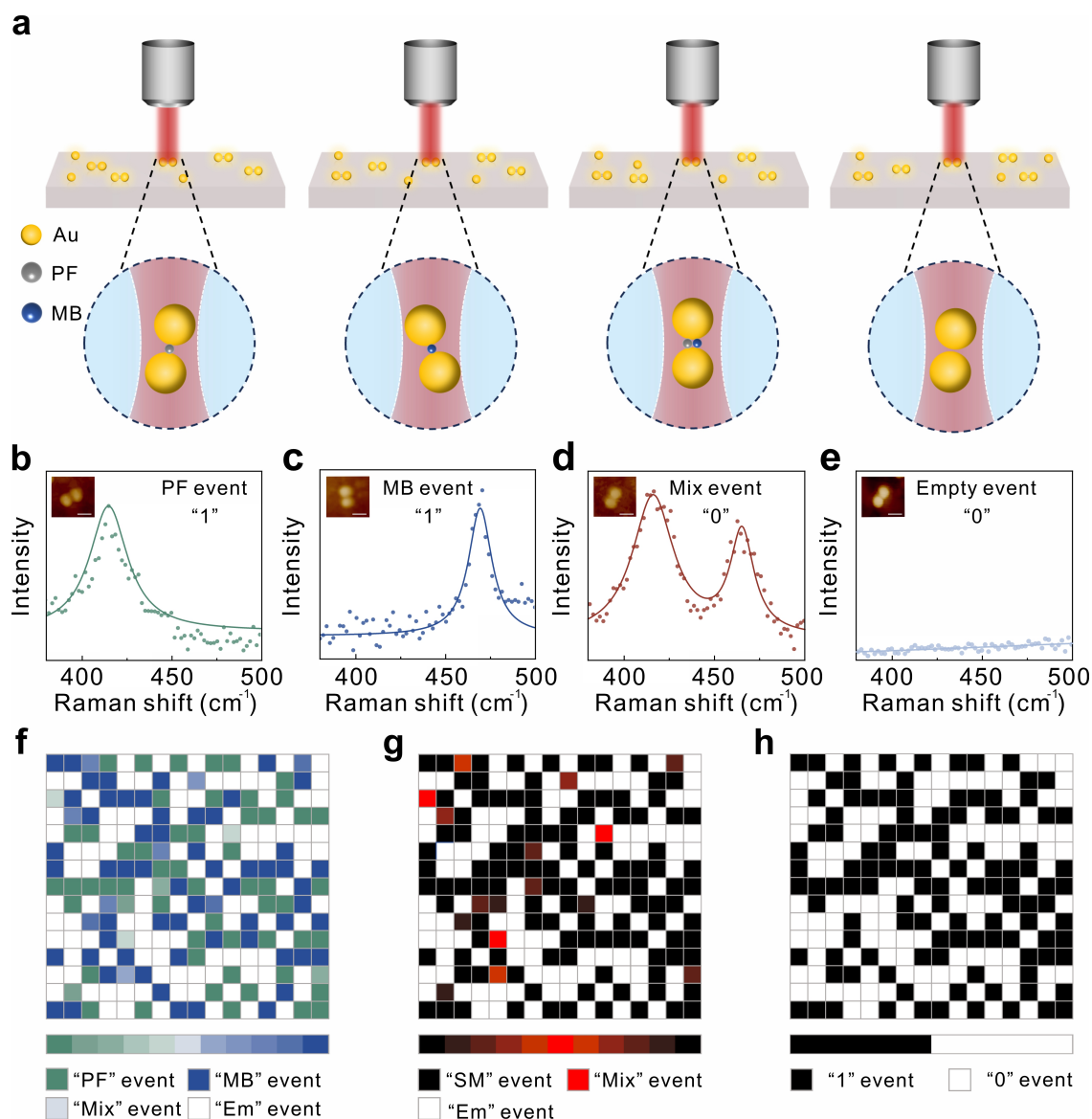


Fig. 2 | Digital bi-SERS identification of single-molecule events in Au NDCs. **a**, Schematic of surface-enhanced Raman spectroscopy (SERS) measurements on individual Au nanodimer cavities (Au NDCs) under four cases. **b-e**, Representative SERS spectra from individual Au NDCs corresponding to (b) single PF events, (c) single MB events, (d) mixed PF and MB events, and (e) empty events (no target molecules), as defined in **a**. Each acquisition voxel is classified as “1” or “0” depending on the presence or absence of a single-molecule event. Insets show AFM images of the measured samples (scale bar: 100 nm). **f**, Heatmap showing the occurrence frequencies of the four cases, constructed using $P_{\text{MB}} = I_{\text{MB}}/(I_{\text{MB}} + I_{\text{PF}})$, where I_{MB} and I_{PF} are the intensities of the SERS peaks of MB and PF, respectively. Blue squares indicate $P_{\text{MB}} = 1$, green squares indicate $P_{\text{MB}} = 0$. **g**, Heatmap derived from **f** by merging blue and green squares into black squares to represent single-molecule events. Squares with intermediate colors are mapped from red to black according to the deviation of P_{MB} from 0.5 (marked in red). **h**, Digital bi-SERS map obtained by labeling single-molecule events as black squares (“1”) and non-single-molecule events as white squares (“0”).

As illustrated in Fig. 2a, four distinct configurations can occur within the nanogaps of individual Au NDCs. When only a single PF or MB molecule is present, distinct Raman signatures are observed at 400–410 cm^{-1} (PF, Fig. 2b) and 460–470 cm^{-1} (MB, Fig. 2c), respectively, corresponding to single-molecule events. In contrast, simultaneous presence (Fig. 2d) or complete absence (Fig. 2e) of both molecules corresponds to bimolecular and empty events, respectively. To quantify these outcomes, we construct a probability heatmap based on the relative MB contribution $P_{\text{MB}} = I_{\text{MB}}/(I_{\text{MB}} + I_{\text{PF}})$ (Fig. 2f). Single MB and PF events are identified by $P_{\text{MB}} = 1$ and $P_{\text{MB}} = 0$, respectively, while intermediate values indicate bimolecular events. By merging single-molecule events into black squares (Fig. 2g) and grouping bimolecular and empty events as non-single-molecule results (Fig. 2h), we find that approximately 47.5% of measured Au NDCs exhibit single-molecule events. Excluding empty gaps, this proportion can increase to 84.6%, confirming the high fidelity of deterministic single exciton integration.

Having established the deterministic plasmon-single-exciton platform, we proceed to spectroscopic measurements of VRS (Supplementary Fig. S4). When the Au NDC/CB[7]@single MB hybrid is placed on a conventional bulk SiO_2 substrate, the intrinsic VRS is predicted to be $\Omega_{\text{VRS}} = 87.5 \text{ meV}$, calculated from $g = 51.82 \text{ meV}$, $\Gamma_e = 70 \text{ meV}$ (Supplementary Fig. S5), and $\Gamma_p = 181 \text{ meV}$ (Supplementary Fig. S6). The g value between

the plasmon mode and the single exciton is calculated using:

$$g = \frac{\epsilon_e}{\sqrt{2\epsilon_0\epsilon_p V_{\text{eff}}}} \boldsymbol{\mu}_e \cdot \mathbf{f}_p(\mathbf{r}) \quad (3)$$

where the effective mode volume $V_{\text{eff}} = 52.5 \text{ nm}^3$ (Methods; Supplementary Note 3, Fig. S7, and Table S1), the transition dipole moment $\mu_e = 0.09 \text{ e}\cdot\text{nm}^{39,54}$, and the normalized electric field (EF) at the location of the exciton $f_p(\mathbf{r}) = \frac{E_p(\mathbf{r})}{|E_p(\mathbf{r})|_{\text{max}}} = 1$ are employed.

By tuning the gap plasmon resonance into alignment with the MB exciton absorption at 1.879 eV (660 nm) through adjustment of the Au nanoparticle radius to 42 nm, the SS becomes observable in dark-field scattering measurements (Supplementary Fig. S8). Varying the particle radius from 36 to 46 nm tunes the plasmon mode across the 635-680 nm range (Supplementary Fig. S6), yielding a clear anticrossing in the dispersion with $\Omega_{\text{SS}}^p = 118.2 \text{ meV}$. However, this value deviates substantially from the intrinsic $\Omega_{\text{VRS}} = 87.5 \text{ meV}$, corresponding to a large relative error of $\sigma \approx 35.1\%$. This discrepancy highlights a fundamental limitation of conventional spectral readout of VRS in plasmon-single-exciton strong coupling.

Quantum-precision-spectroscopy metrology of VRS

To enable a quantum-precision-spectroscopy metrology of the VRS in the Au NDC/CB[7]@single-MB hybrid system, it is essential to reduce the Γ_p and match it with the excitonic linewidth Γ_e . It pushes the coupling system toward the ZEP (Fig. 1b), facilitating the quantum-precision determination of the VRS. We achieve this through precise EP-assisted photonic substrate engineering that spectrally slims the plasmon mode without compromising its nanoscale field confinement. In specific, we fabricate a Si/SiO₂ photonic substrate that tailors the electromagnetic environment above its top surface by engineering optical pathways (OPs)⁵⁵ (i.e. radiative channels, Fig. 3a(I) and Supplementary Fig. S9), arising from constructive and destructive interference between incident and reflected optical fields⁵⁶. The gray line in Fig. 3b presents the measured reflectance spectra of the Si/SiO₂ substrate, comprising a 2.58- μm -thick SiO₂ layer on a Si wafer. Multiple transverse-magnetic photon modes (TM₁-TM₅) are observed across the spectral range of 550-800 nm. The calculated multiplication factor (M factor) spectrum, $F_m(\mathbf{r}, \omega, \hat{\boldsymbol{\mu}}_p) = \rho(\mathbf{r}, \omega, \hat{\boldsymbol{\mu}}_p)/\rho_0$, represents the ratio of the projected local density of states (PLDOS) to that in vacuum (ρ_0) (Methods), as illustrated by the red curve in Supplementary Fig. S9a. It exhibits five distinct Lorentzian peaks with

$F_m > 1$, corresponding to five open OPs at TM₁–TM₅. When an Au NDC is positioned on the photonic substrate (Fig. 3a(II)), its gap plasmon mode spectrally overlaps with the TM₃ photon mode, resulting in strong hybridization between the plasmon mode and the engineered OP at TM₃. This interaction reshapes the plasmonic spectral response and modifies its radiative and non-radiative decay channels.

The blue line in Fig. 3b shows the normalized scattering spectrum of an Au NDC supported on the Si/SiO₂ photonic substrate, revealing that the plasmonic damping linewidth is compressed to $\Gamma_p = 89$ meV, from 181 meV that measured on SiO₂ substrate, where OPs are absent (Supplementary Fig. S6b). The experimentally measured linewidth narrowing agrees well with our analytical predictions based on equation (4) (Methods) and is further corroborated by full-wave finite element simulations (Supplementary Note 4):

$$S(\omega) \propto -F_m(\mathbf{r}, \omega, \hat{\mu}_p) \cdot \text{Im} \left\{ \hbar\omega - \varepsilon_p + i\frac{\Gamma_p}{2} - \sum_{j=1}^N \frac{g_j^2}{\hbar\omega - \varepsilon_{cj} + i\frac{\Gamma_{cj}}{2}} \right\}^{-1}. \quad (4)$$

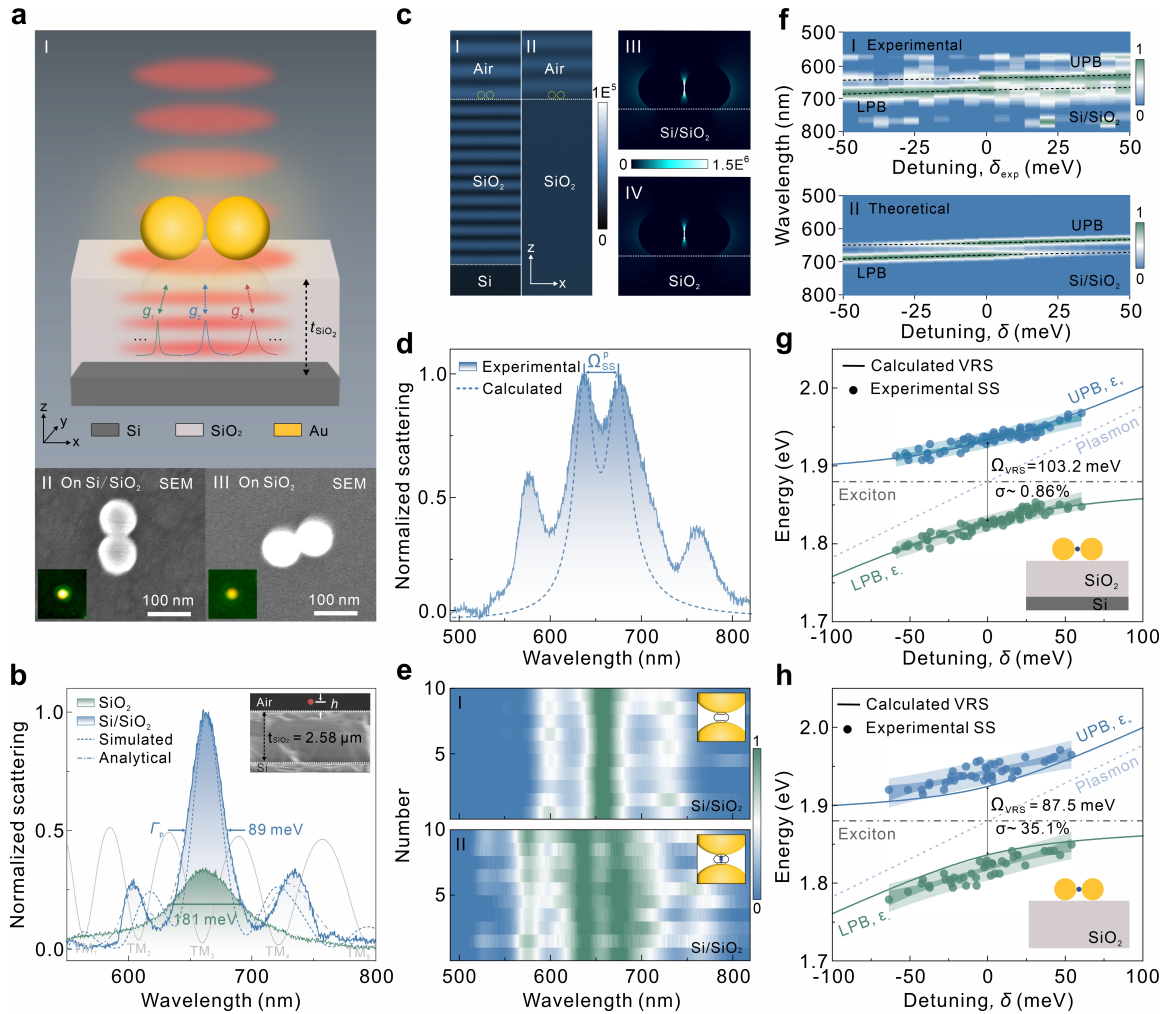
This pronounced suppression of plasmonic damping linewidth arises from resonant coupling among the lossy plasmon mode, the photon modes, and the vacuum reservoir, mediated by the engineered OPs. In addition to linewidth suppression, the photonic substrate places the Au NDC in an electromagnetic environment with maximized EFs due to constructive interference (Fig. 3c(I)). As a result, the nanogap EF is slightly enhanced by about 1.4 times relative to the Au NDC on the bulk SiO₂ substrate (Fig. 3c(III, IV)), where destructive interference minimizes the local fields (Fig. 3c(II)). It leads to an increase in scattering intensity, manifested as stronger brightness of the Au NDCs measured on Si/SiO₂ photonic substrate (insets in Fig. 3a(II, III)).

When an individual Au NDC/CB[7]@single MB hybrid is positioned on the Si/SiO₂ photonic substrate with $t_{\text{SiO}_2} = 2.58$ μm , the damping linewidth of the gap plasmon mode, Γ_p , is markedly narrowed, resulting in $\Gamma_e/\Gamma_p \approx 0.79$ (Supplementary Fig. 9) and pushing the system close to the ZEP ($\Gamma_e/\Gamma_p = 0.82$). This linewidth matching substantially decreases the critical interaction strength at EP from $g_{\text{EP}} = 27.75$ meV on SiO₂ to $g_{\text{EP}} = 4.75$ meV on the Si/SiO₂ photonic substrate. Consequently, the VRS of this plasmon-single-exciton strong coupling system increases from $\Omega_{\text{VRS}} = 87.5$ meV to $\Omega_{\text{VRS}} = 103.2$ meV (Supplementary Table S1). Figure 3d displays a representative scattering spectrum obtained from a single Au NDC/CB[7]@single MB hybrid on Si/SiO₂ substrate, demonstrating an experimental SS of

271 $\Omega_{SS}^p = 104.1$ meV. It agrees well with the theoretical result calculated using (Supplementary
 272 Note 1)¹⁹,

$$273 \quad \zeta(\omega) \propto -\text{Im} \frac{(\hbar\omega - \varepsilon_e + i\Gamma_e/2)}{(\hbar\omega - \varepsilon_p + i\Gamma_p/2) \cdot (\hbar\omega - \varepsilon_e + i\Gamma_e/2) - g^2}, \quad (5)$$

274 with $g = 51.87$ meV, $\bar{\Gamma}_p = 89.6$ meV (Fig. 3e(I)), and $\Gamma_e = 70$ meV. Moreover, the observed
 275 Rabi splitting fully satisfies the critical strong-coupling condition of $g^2 > \Gamma_e^2/8(1 + \Gamma_p/2\Gamma_e)$ ⁴⁴,
 276 confirming that the system operates within the strong coupling regime. Importantly, this
 277 approach demonstrates a good repeatability across individual Au NDC/CB[7]@single MB
 278 hybrids at resonance, as shown in Fig. 3e(II).



279
 280 **Fig. 3 | Spectral compressing enables quantum-precision-spectroscopy measurements of**
 281 **VRS. a, (I)** Schematic of an Au NDC positioned in an engineered electromagnetic environment
 282 engineered by a Si/SiO₂ photonic substrate. (II, III) Scanning electron microscope (SEM)
 283 images of Au NDCs measured on (II) Si/SiO₂ and (III) SiO₂ substrates, respectively. The insets
 284 are corresponding dark-field (DF) images. **b, Normalized scattering spectra of individual Au**

NDCs (radius 42 nm) on the photonic substrate and on a bulk SiO₂ substrate, respectively. The dashed blue curve corresponds to numerical simulation results, while the dot-dashed pale blue curve represents analytical prediction calculated using equation (4). The measured reflectance of the Si/SiO₂ photonic substrate is overlaid in gray. Inset: side-view scanning electron microscope (SEM) image of the photonic substrate. **c**, Left: EF distributions of the Si/SiO₂ photonic substrate (I) and bulk SiO₂ substrate (II). Right: Cross-sectional (*x*-*z*) EF distributions of the Au NDC positioned on each substrate. **d**, Normalized scattering spectra of an Au NDC/CB[7]@single MB hybrid on Si/SiO₂ photonic substrate. The dashed curve is the theoretical prediction calculated using equation (5). **e**, (I) Normalized scattering spectra from individual Au NDCs, whose gaps are filled by single CB[7] molecules that are empty, or (II) encapsulating single MB molecules, measured on the Si/SiO₂ substrates with $t_{\text{SiO}_2} = 2.58 \mu\text{m}$. **f**, (I) Normalized scattering spectra of individual Au NDC/CB[7]@single MB hybrids measured on the Si/SiO₂ substrates with t_{SiO_2} of around 2.58 μm , ordered according to the experimental detuning δ_{exp} . (II) Normalized scattering spectra calculated using equation (5) based on parameters extracted from experimental measurements, arranged by the theoretical detuning δ . **g**, **h**, Hybridized state energies extracted from individual Au NDC/CB[7]@single MB hybrids measured on **g** Si/SiO₂ photonic substrates t_{SiO_2} of around 2.58 μm and **h** SiO₂ substrates, respectively. Solid curves represent theoretical states of the hybridized eigenenergies, $\varepsilon_{\pm}$. Shaded regions indicate the 95% confidence intervals. UPB: upper plexciton branch; LPB: lower plexciton branch.

To verify that the observed SS originates from genuine plasmon-exciton coupling rather than plasmonic artifacts, we performed laser-induced photobleaching experiments (Supplementary Note 5 and Fig. S10). Following photobleaching of the MB molecule, the SS features in the scattering spectra disappear, directly providing that the observed SSs arise from strong coupling between the molecular excitons and the plasmonic nanocavities. Additionally, the plasmonic resonances of the Au NDCs can be well tuned from approximately 639 to 679 nm by adjusting the Au nanoparticle radius (39-44 nm) and the SiO₂ layer thickness ($t_{\text{SiO}_2} = 2.52$ -2.65 μm) in the Si/SiO₂ photonic substrates (Supplementary Figs. S11–S13). This good tunability enables precise spectral overlap of the substrate-engineered gap plasmon mode with the exciton absorption at ~ 660 nm, facilitating a systematic investigation of energy dispersions in individual Au NDC/CB[7]@single MB hybrids (Supplementary Fig. S14). Figure 3f(I) presents normalized scattering spectra of individual Au NDC/CB[7]@single MB hybrids on the Si/SiO₂ photonic substrate with t_{SiO_2} of around 2.58 μm , ordered by the experimental detuning $\delta_{\text{exp}} = \varepsilon_+ + \varepsilon_- - 2\varepsilon_e$. A pronounced anticrossing behavior is observed, which

319 closely follows the theoretical results calculated using equation (5) and ordered according to
 320 theoretical detuning $\delta = \varepsilon_p - \varepsilon_e$ (Fig. 3f(II)). Finally, the energy dispersions of the plexcitonic
 321 states extracted from the measured SSs yield $\Omega_{ss}^p = 104.1$ meV, with a relative measurement
 322 error of $\sigma \approx 0.86\%$ in probing the VRS (Fig. 3g). This uncertainty is approximately 40-fold
 323 smaller than that obtained for the same system on the bulk SiO₂ substrate (Fig. 3h).

324 To further demonstrate the importance of OP manipulation in accurate probing the VRS, we
 325 performed scattering measurements on a large number of individual Au NDC/CB@single MB
 326 hybrids on Si/SiO₂ photonic substrates with different t_{SiO_2} . By precisely tuning the thickness
 327 of SiO₂ layer (Fig. 4a), the photon modes and OPs created in the electromagnetic environment
 328 can be deterministically modulated. Figure 4b compares the reflectance of the bulk SiO₂
 329 substrate and the Si/SiO₂ photonic substrates with $t_{SiO_2} = 1.68, 2.58$, and 2.82 μm , respectively.
 330 Unlike the bulk SiO₂ substrate, which does not support photon modes and OPs in the
 331 electromagnetic environment above its top surface (Figure 4b (I)), each Si/SiO₂ photonic
 332 substrate hosts a photon mode and a corresponding OP at $\lambda \approx 660$ nm (Figure 4b (II-IV)).
 333 However, these OPs exhibit different radiative linewidths varying from 114.2 meV to 69.8 meV.
 334 When the bare Au NDCs, supporting gap plasmon resonances at approximately 660 nm, are
 335 placed on these substrates, their damping linewidths can be modulated in different degrees by
 336 the OPs (Figure 4c). As t_{SiO_2} is increased from 1.68 to 2.82 μm , the mean damping linewidth
 337 (at $\lambda \approx 660$) of individual Au NDCs decreases from $\tilde{\Gamma}_p = 115.3 \pm 5.3$ meV to 69.9 ± 1.3 meV
 338 (Supplementary Figs. S9, S15 and S17). Consequently, when individual Au
 339 NDC/CB[7]@single MB hybrids are located on the Si/SiO₂ photonic substrates with $t_{SiO_2} =$
 340 1.68 μm and 2.82 μm , the minimum errors in measuring the VRS decrease to $\sigma = 6.12\%$ and
 341 2.89%, respectively (Supplementary Fig. S16 and S17), which are larger than that measured
 342 on Si/SiO₂ photonic substrate with $t_{SiO_2} = 2.58$ μm (Figure 3h). Nevertheless, these
 343 uncertainty remains decreased by a factor of about 5.7 and 12.1, respectively, compared to
 344 those measured on a bulk SiO₂ substrate.

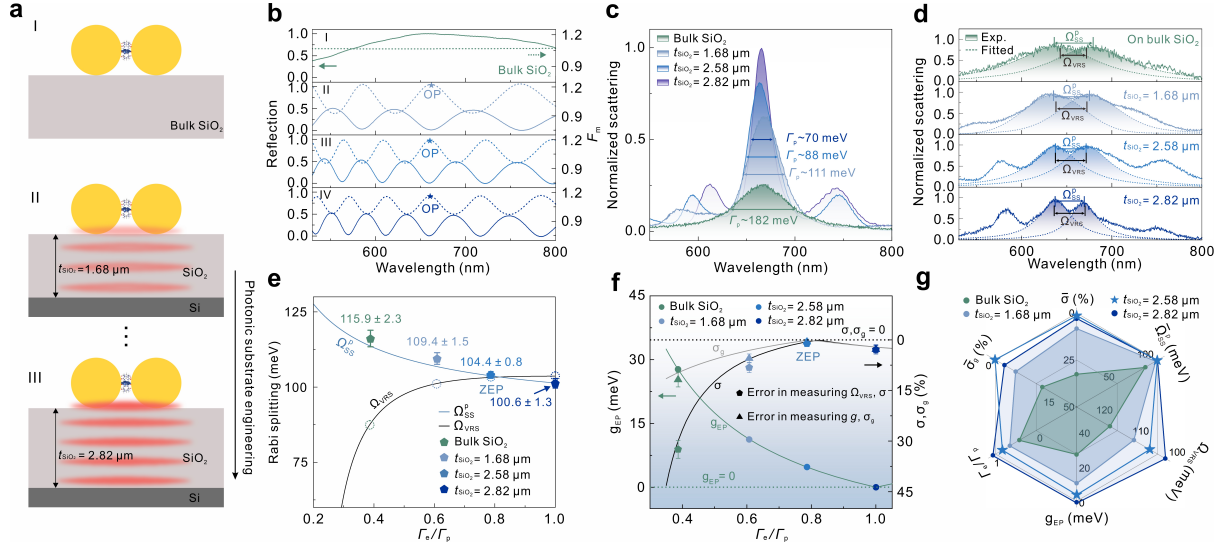


Fig. 4 | High precisions of VRS metrology enabled by EP-assisted photonic substrate engineering. **a**, Schematics of Au NDCs supported on bulk SiO₂ and on Si/SiO₂ photonic substrates with t_{SiO_2} varying from 1.68 μm to 2.82 μm . **b**, Measured reflectance and simulated F_m spectrum of (I) bulk SiO₂ and Si/SiO₂ photonic substrates with different t_{SiO_2} of (II) 1.68 μm , (III) 2.58 μm and (IV) 2.82 μm , respectively. **c**, Normalized scattering spectra of individual Au NDCs on the four substrates. All spectra are normalized to the maximum scattering intensity from the Au NDC on Si/SiO₂ substrate with $t_{\text{SiO}_2} = 2.82 \mu\text{m}$. **d**, Normalized scattering spectra of individual Au NDC/CB[7]@single MB hybrids measured on the four substrates in **b**. The dashed curves represent Lorentzian fits applied to determine the Ω_{SS}^p . **e**, Statistical Ω_{SS}^p for individual Au NDC/CB[7]@single MB hybrids measured on the four substrates. Dashed circles denote the theoretically expected Ω_{VRS} for the four cases. Black and pale blue curves are calculated Ω_{VRS} and Ω_{SS}^p , as functions of the linewidth ratio Γ_e/Γ_p . **f**, Experimental and theoretical (solid black and gray lines) measurement errors of VRS (σ , pentagons) and g value (σ_g , triangles) for individual Au NDC/CB[7]@single MB hybrids on the four substrates in **b**. The colored markers are the g_{EP} at EP for these four cases. **g**, Radar plot summarizing the key parameters of Γ_e/Γ_p , $\bar{\sigma}$, $\bar{\sigma}_g$, $\bar{\Omega}_{\text{SS}}^p$, Ω_{VRS} , and g_{EP} for measurements mentioned above.

Figure 4d presents the representative scattering spectra of individual Au NDC/CB[7]@single MB hybrids on bulk SiO₂ and Si/SiO₂ photonic substrates with $t_{\text{SiO}_2} = 1.68, 2.58$, and $2.82 \mu\text{m}$. The spectra illustrate a progressive clarity with the deepening of SS and increasingly smaller deviations between the measured SS and the intrinsic VRS, as the Γ_p is reduced. Figure 4e summarizes the statistical mean values of the measured SS on these four photonic substrates, demonstrating $\bar{\Omega}_{\text{SS}}^p = 115.9 \pm 2.3 \text{ meV}$, $109.4 \pm 1.5 \text{ meV}$, $104.4 \pm 0.8 \text{ meV}$, and 100.6 ± 1.3

meV, respectively. This trend aligns closely with our theoretical predictions, and gradually converges toward the intrinsic Ω_{VRS} at the ZEP (Figure 4e). When individual Au NDC/CB[7]@single MB hybrids are positioned on the aforementioned four substrates, the ratio of Γ_e/Γ_p can be precisely tuned to values of 0.39, 0.61, 0.79, and 1.0, respectively. In these cases, the relative error $\bar{\sigma}$ in probing Ω_{VRS} demonstrates a significant decrease, from $\bar{\sigma} = 32.5 \pm 2.6\%$ measured on SiO_2 to $\bar{\sigma} = 8.2 \pm 1.5\%$, $1.1 \pm 0.8\%$ and $2.9 \pm 1.2\%$ measured on the three Si/SiO₂ photonic substrates (Figure 4f).

Furthermore, it also presents an effective strategy for directly measuring the g value of the plasmon-single-emitter strong coupling system. For ideal strong coupling systems with negligible losses, the g value is typically calculated using equation (3) or directly measured via SS, where $\Omega_{\text{SS}} = \Omega_{\text{VRS}} = 2g$. However, for lossy, non-Hermitian plasmon-single-emitter systems, this relationship does not hold because of substantial plasmonic dissipation and the discrepancy between the linewidths of plasmonic and excitonic states. In this scenario, the g value is typically calculated according to equation (3), while its accuracy highly depends on the precise determination of the emitter's position within the nanocavity and the alignment of its transition dipole moment relative to the mode field. Unfortunately, for most plasmon-emitter strong coupling systems, it is difficult to ensure both of these two determinacies⁴².

When Γ_p is suppressed and approaches to Γ_e , the g value gradually satisfies the relation $\Omega_{\text{SS}} \approx \Omega_{\text{VRS}} \approx 2g$ (equations (1) and (2), providing a way to directly measure g with high precision. By defining the relative measurement error of g as $\sigma_g = \frac{g^{\text{exp}} - g}{g} \times 100\%$, where $g^{\text{exp}} = \Omega_{\text{SS}}/2$ and g is calculated from equation (3), the statistical σ_g measured across different individual Au NDC/CB[7]@single MB hybrids reduce from $\bar{\sigma}_g = 11.8 \pm 2.2\%$ measured on SiO_2 to $\bar{\sigma}_g = 5.5 \pm 1.3\%$, $0.6 \pm 0.8\%$, and $2.8 \pm 1.3\%$ (triangles in Figure 4f) on the three photonic substrates. Notably, when the Γ_p is reduced to result in $\Gamma_e/\Gamma_p \approx 0.79$, the relative σ_g can be reduced to $\sim 0.60\%$, closing to zero and indicating a 20-fold improvement in measurement accuracy compared to that measured on SiO_2 substrate. To provide a comprehensive comparison, the statistical measurement errors of $\bar{\sigma}$ and $\bar{\sigma}_g$ combined with the other four parameters are summarized in a radar plot, as shown in Figure 4g. This analysis

highlights that the EP-assisted photonic substrate engineering substantially reduces the plasmonic damping linewidth Γ_p , yielding a closer match to the exciton linewidth Γ_e . As a result, the plasmon-exciton coupling parameter g_{EP} decreases systematically from 27.75 meV on bulk SiO₂ to 11.25 meV, 4.75 meV and 0 meV for the Si/SiO₂ photonic substrates with $t_{\text{SiO}_2} = 1.68, 2.58$ and $2.82 \mu\text{m}$. Correspondingly, the intrinsic VRS increases from $\Omega_{\text{VRS}} = 87.5$ meV to 101.1 meV, 103.2 and 103.6 meV (circles in Figure 4f). In this process, we realize the quantum precision measurement of VRS and g .

Conclusion

We have established a robust, deterministic strategy for achieving quantum-precision-spectroscopy metrology of the VRS and g value in lossy, non-Hermitian plasmon-single-emitter strong coupling systems. The central concept is to suppress and precisely engineer the damping linewidth of the plasmon mode so that it is optimally matched to the exciton linewidth, thereby minimizing spectral broadening and measurement uncertainty. This is accomplished through the precise EP-assisted photonic substrate engineering, which tailors the electromagnetic environment surrounding the coupled systems.

Using an Au NDC hosting a single MB molecule via CB[7]-mediated active assembly, we realized a deterministic plasmon-single-exciton strong coupling platform with well-defined geometry and coupling parameters. By integrating this hybrid system onto a Si/SiO₂ photonic substrate and precisely tuning the SiO₂ thickness, we generated controllable OPs that reshape the photonic PLDOS. At an optimized SiO₂ thickness of $2.58 \mu\text{m}$, the plasmon damping linewidth was suppressed to 1.27 times the exciton linewidth. This linewidth matching dramatically reduced the effective plasmon-exciton damping imbalance and enabled a substantial enhancement in the resolvability of the intrinsic VRS. As a result, we achieved a record-low relative measurement errors of $\sigma \approx 0.86\%$ and $\sigma_g \approx 0.60\%$ in determining the VRS and g value of a plasmon-single-exciton system, representing improvements in precision of 40-fold and 20-fold, respectively, compared to measurements performed on conventional SiO₂ substrates. These improvements are not limited to a single device but is statistically robust across many individual hybrids, underscoring the generality and reproducibility of the approach. This robustness facilitates the realization of quantum-precision spectroscopy metrology for plasmon-single-emitter strong coupling systems operating at room temperature.

Beyond precise VRS and g value determination, our results establish photonic substrate engineering as a powerful and versatile tool for linewidth control in lossy plasmonic systems. The demonstrated ability to systematically tune plasmon damping, coupling parameters, and measurement precision through substrate design opens new opportunities for studying quantum electrodynamical phenomena at the single-emitter level, including ultrastrong coupling, non-Hermitian physics, dissipative quantum dynamics, and ultrafast coherent energy exchange measurements (Supplementary Fig. S19) at room temperature.

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Methods

Fabrication and characterization of Au NDCs and Au NDC/CB[7]@single MB hybrids

Au NDC/CB[7]@single MB hybrids were fabricated by first centrifuging and washing 4 mL of as-synthesized smooth Au nanoparticles (Supplementary Note 6) twice with deionized water, followed by redispersion in 1 mL of deionized water. Subsequently, 60 μ L of a CB[7]@MB emitter solution (3.33×10^{-8} M) was added and allowed to hybridize for 4 h. During this process, CB[7] molecules act as molecular linkers that mediate the assembly of individual Au nanoparticles into nanodimer cavities while hosting MB molecules within the nanogap, enabling deterministic formation of Au NDC/CB[7]@single MB hybrids in which a single MB molecule is precisely positioned at the gap center with its transition dipole moment optimally aligned to the gap-plasmon mode (Supplementary Fig. S2d). Based on the quantities used, approximately 1.2×10^{12} Au nanoparticles and about 6.0×10^{11} MB monomers participated in the hybridization, corresponding to an average loading of $\bar{n}_{\text{MB}} \approx 0.86$ CB[7]@single MB emitters and $\bar{n}_0 \approx 0.86$ unoccupied CB[7] molecules per Au nanoparticle (MB: CB = 1: 2), ensuring single-exciton coupling to individual Au nanodimers. TEM images were acquired using a JEOL JEM-F200 microscope operated at 100 kV, and absorption spectra were measured with a UV-vis-NIR spectrophotometer (Cary 5000) using 3 mL of deionized water as the reference.

Fabrication and characterization of Si/SiO₂ photonic substrates

Bulk silicon substrates (MKNANO, Nanjing) were cleaned by sonication in acetone for 10 min, followed by sequential sonication in isopropyl alcohol and deionized water for 10 min each. SiO₂ layers were subsequently deposited by inductively coupled plasma chemical vapor deposition (ICP-CVD; PlasmonPro System100 ICP180-CVD, Oxford Instruments). Scanning electron microscopy (SEM) images of the photonic substrates were acquired using Zeiss Auriga-39-34 and JEOL JSM-7610F Plus microscopes operated at an accelerating voltage of 5 kV. Reflectance spectra of the bare Si/SiO₂ photonic substrates were measured using a Filmetrics F20 system (Filmetrics Inc., USA).

Dark-field scattering and SERS measurements

The experimental setup for dark-field scattering measurements is shown in Supplementary Fig. S4. A 10 μ L droplet of the Au NDC/CB[7]@single MB hybrid solution was deposited onto the

Si/SiO₂ photonic substrate and allowed to stand for 5 min, after which the droplet was removed, the surface rinsed with deionized water, and dried under a nitrogen stream. This procedure resulted in the immobilization of isolated individual Au NDC/CB[7]@single MB hybrids on the substrate surface. To establish a one-to-one correspondence between scattering spectra and individual hybrids, dark-field optical microscopy was combined with SEM imaging of the same field of view.

Dark-field scattering measurements were performed using a custom-built optical system (Olympus BX53M) equipped with a monochromator (Acton SpectraPro 2360), a 100 W quartz tungsten halogen light source, and a thermoelectrically cooled CCD camera (Princeton Instruments Pixis 100B_eXcelon, Acton Inc., USA) operated at -70 °C. Scattered light was collected in a backscattering geometry using a high-numerical-aperture dark-field objective (100×, NA = 0.80). Simultaneously, color dark-field images were acquired with a high-resolution digital color camera (ARTCAM-300MI-C, ACH Technology Inc.) positioned at the microscope imaging plane.

Bi-SERS measurements were conducted using an atomic force microscope–Raman (AFM–Raman) integrated system (NTEGRA Spectra PNL, NT-MDT Co., Russia), as schematically illustrated in Supplementary Fig. S4. Raman spectra were excited with a 633 nm laser and acquired with an integration time of 2 s at a pump intensity of 60 μW/μm².

Mode volume calculation

Owing to the sub-nanometre gap of the Au nanodimer cavity (Au NDC), the plasmonic EF ($|E_p(\mathbf{r}, \omega)|$) and the corresponding electromagnetic energy density $w(\mathbf{r}, \omega)$ are predominantly confined within the interparticle gap region (Supplementary Fig. S2). As a result, an Au NDC with a 0.9 nm gap can be treated as an optical nanocavity, and its mode volume V_m can be evaluated using^{23,38,57}

$$V_m(\omega) = \frac{\int w(\mathbf{r}, \omega) d^3r}{\max[w(\mathbf{r}, \omega)]}, \quad (6)$$

with the electromagnetic energy density given by

$$w(\mathbf{r}, \omega) = \frac{\epsilon_0}{2} \left[\text{Re}[\epsilon(\mathbf{r}, \omega)] + \frac{2\omega \text{Im}[\epsilon(\mathbf{r}, \omega)]}{\gamma_p} \right] |E_p(\mathbf{r}, \omega)|^2, \quad (7)$$

where ϵ_0 is the free-space permittivity, $\epsilon(\mathbf{r}, \omega)$ is the relative dielectric constant at the position of $\mathbf{r}$.

All calculations were performed using three-dimensional finite element method (3D-FEM) simulations. To accurately evaluate the volume integral of $w(\mathbf{r}, \omega)$ for the gap-plasmon mode, the EF distribution was computed over a three-dimensional domain and numerically integrated. The mode volume reaches its minimum at $\lambda = 660$ nm, yielding $V_m = 52.5$ nm³, which is in close agreement with the value ($V_m = 53.5$ nm³) independently estimated from Purcell factor simulations using the same geometry (Supplementary Note 3). While, this method based on equations (6, 7) is more suitable for calculating the mode volume of a nanocavity excited by the plane wave, as we have utilized in our experiments. In the simulations, the dispersive permittivities of Si and SiO₂ were taken from experimentally measured refractive index data reported by Aspnes and Studna (1983)⁵⁸ and Rodríguez-de Marcos *et al.* (2016)⁵⁹, respectively, while the permittivity of gold was adopted from the experimental measurements of Johnson and Christy⁶⁰.

Theoretical model for plasmonic damping linewidth manipulating

We consider a metal nanostructure that supports a well-defined bosonic dipolar surface plasmon resonance (SPR) mode, described by annihilation and creation operators p and p^+ , respectively, while higher-order multipolar modes are neglected. Under weak external excitation by an incident field $\mathbf{E}_0$, the interaction Hamiltonian of the plasmon mode can be written as^{44,55}

$$H_{dir} = \boldsymbol{\mu}_p \cdot (\mathbf{E}_0 + \sum_o \mathbf{E}_o) \cdot (p^+ + p) \quad (8)$$

here $\boldsymbol{\mu}_p$ is the transition dipole moment of the SPR mode and $\sum_o \mathbf{E}_o$ denotes the total EF contributed by the OPs. In practice, the excitation field is much weaker than the OP-induced fields ($|\mathbf{E}_0| \ll |\mathbf{E}_\alpha|$), allowing $\mathbf{E}_0$ to be neglected in equation (8).

According to Fermi's golden rule, the absorption spectrum of the plasmon mode is given by

$$S(\omega) \propto -\text{Im} \frac{A_p \langle i | p; p^+ | i \rangle}{\varepsilon_{fi} - \omega - i0^+} = -\text{Im} \{ A_p \ll p; p^+ \gg_{\omega+i0^+} \}, \quad (9)$$

where $\ll p; p^+ \gg_{\omega+i0^+}$ is the retarded Zubarev Green's function^{19,61}, and

$$A_p = |\boldsymbol{\mu}_p \cdot \sum_o \mathbf{E}_o|^2 \propto F_m(\mathbf{r}, \omega, \hat{\boldsymbol{\mu}}_p) \cdot \rho_0. \quad (10)$$

where, $F_m(\mathbf{r}, \omega, \hat{\boldsymbol{\mu}}_p) = \rho(\mathbf{r}, \omega, \hat{\boldsymbol{\mu}}_p) / \rho_0$ is the multiplication enhancement factor (M factor) of the projected local density of optical states (LDOS) $\rho(\mathbf{r}, \omega, \hat{\boldsymbol{\mu}}_p)$ along the plasmonic dipole orientation $\hat{\boldsymbol{\mu}}_p$, and ρ_0 denotes the LDOS in vacuum. The Hamiltonian that describes the

interactions between the plasmon mode and the photon modes engineered by the photonic substrate can be rewritten as⁵⁵,

$$H = (\varepsilon_p p^+ p + H_{\text{decay1}}) + (\sum_{j=1}^N \varepsilon_{cj} c_j^+ c_j + H_{\text{decay2}}) - \sum_{j=1}^N g_j [p^+ c_j + c_j^+ p], \quad (11)$$

where, c_j and c_j^+ are bosonic annihilation and creation operators of the j th photon mode, g_j is the coupling constant between the plasmon mode and this photon mode, while $\varepsilon_p(\varepsilon_{cj})$ and $\Gamma_p(\Gamma_{cj})$ denote the energy and damping linewidth of the plasmon mode (the j th photon mode), respectively; H_{decay1} and H_{decay2} are Hamiltonians reflecting the inelastic interactions of the plasmon mode and the photon modes with a continuum of modes, respectively.

By explicitly evaluating the Green's function and the Heisenberg equation in equation (11), the $\ll p; p^+ \gg_{\omega+i0^+}$ can be deduced as,

$$\ll p; p^+ \gg_{\omega+i0^+} = \left\{ \hbar\omega - \varepsilon_p + i\frac{\Gamma_p}{2} - \sum_{j=1}^N \frac{g_j}{\hbar\omega - \varepsilon_{cj} + i\frac{\Gamma_{cj}}{2}} \right\}^{-1} \quad (12)$$

Lastly, the absorption spectrum of the plasmonic nanostructure in the engineered electromagnetic environment can be deduced as,

$$\begin{aligned} S(\omega) &\propto -\text{Im}\{A_p \ll p; p^+ \gg_{\omega+i0^+}\} \\ &= -F_m(\mathbf{r}, \omega, \hat{\boldsymbol{\mu}}_p) \cdot \text{Im} \left\{ \hbar\omega - \varepsilon_p + i\frac{\Gamma_p}{2} - \sum_{j=1}^N \frac{g_j^2}{\hbar\omega - \varepsilon_{cj} + i\frac{\Gamma_{cj}}{2}} \right\}^{-1} \end{aligned} \quad (13)$$

i.e., equation (3) in the main text. We note that the scattering spectrum of the engineered plasmon mode can also be described by equation (13) if one measures its optical response from the same plasmonic nanostructure and ignores the intensity difference⁶². Equation (13) explicitly shows that the spectral response of the plasmon mode can be effectively engineered through the OP-induced modification of the projected LDOS, captured by the M-factor spectrum $F_m(\mathbf{r}, \omega, \hat{\boldsymbol{\mu}}_p)$. In the weak-coupling limit, the M factor reduces to the conventional Purcell factor⁶³, which can be accurately evaluated using full-wave numerical simulations, as described below.

F_m spectrum simulation

Two-dimensional finite-difference time-domain (2D-FDTD) simulations were performed using a point electric dipole source and a point detector. Bloch boundary conditions were applied

along the x direction, while perfectly matched layers (PMLs) were used along the z direction. In the simulations, an x -polarized point electric dipole source was positioned 42 nm above the Si/SiO₂ photonic substrate, corresponding to the radius of the Au nanoparticles forming the Au nanodimer cavity when placed on the substrate. The dispersive optical constants of all materials employed in the simulations were identical to those used in the mode-volume (V_m) calculations described above.

Data availability

The data that support the plots within this paper and other findings of this study are available from the corresponding author upon reasonable request.

Code availability

Codes used to generate simulated data are available from the corresponding author upon request.

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Author contributions

W.L., L.G., L.W., and R.L. conceived the idea and supervised the project. S.F. and Q.L. carried out the theoretical calculations and numerical simulations. S.F., X.F., and T.L. performed the optical measurements and morphological characterization. D.K.C., W.Y.L., and J.W. fabricated the samples. S.F., X.F., H.Z. and Y.K. contributed to the development of the experimental setup and conducted optical measurements. S.F., G.W., and L.D. performed the bi-SERS measurements. X.H.W. and B.W. provided theoretical and experimental guidance. W.J.Z. contributed by providing the article framework, engaging in data discussions and writing the manuscript. W.L., L.G., L.W., and R.L. led the interpretation of the results and were responsible for writing, revising, and finalizing the manuscript. All authors discussed the results and commented on the manuscript.

Competing interests

The authors declare that they have no competing interests.

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