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Programmable Raman Scattering through Quasi-BIC Pump Gating and Stokes-Sideband Routing

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ABSTRACT: Surface-enhanced Raman scattering (SERS) is usually engineered as a broadband near-field amplification process, in which plasmonic hot spots increase both excitation and emission fields. However, beyond their limited spectral selectivity, conventional plasmonic metasurfaces generally lack deterministic control over the photonic states that mediate Raman scattering. Here, we introduce a hybrid plasmonic–photonic platform that redefines SERS as a momentum-programmable and polarization-switchable inelastic scattering process. Gold nanoparticle hot spots provide molecular-scale electromagnetic enhancement, while dispersive quasi-bound states in the continuum (qBICs) supported by a non-Hermitian photonic crystal slab independently control pump and Stokes channels through the momentum- and polarization-dependent local density of optical states. When a qBIC is resonant with the laser, Raman generation is switched on by critical-coupling-mediated pump gating; when a dispersive photonic branch intersects selected Stokes-shifted frequencies, individual vibrational sidebands are preferentially amplified and routed along the associated momentum-selective radiative channel. Under double-resonant conditions, this hierarchical coupling produces global Raman enhancement factors approaching 250 beyond the baseline SERS substrate enhancement factor ($\sim 10^5$) and selectively promotes otherwise weak conventional SERS bands into dominant spectral features, with sideband-specific amplitude increase exceeding 2,000. These results establish qBIC SERS as a programmable hybrid nanophotonic regime in which Raman spectra are enhanced, optically gated, spectrally filtered, and routed in momentum space with large polarization contrast.

INTRODUCTION

Raman scattering provides chemically specific access to molecular vibrations, but its intrinsically weak cross-section has long limited its sensitivity and applicability^{1,2}. Surface-enhanced Raman scattering (SERS) overcomes this limitation by exploiting plasmonic nanostructures that concentrate electromagnetic fields into deeply subwavelength hot spots^{3,4}, thereby enhancing both the excitation field and the emission of Stokes photons⁵⁻⁷. This strategy has enabled single-molecule sensitivity and a broad range of applications in chemical sensing, biosensing, and molecular spectroscopy⁸⁻²¹. However, the broadband nature of plasmonic hot spots also limits their selectivity: conventional SERS substrates do not deterministically control pump activation, sideband selection, or far-field routing.

A different route emerges if Raman scattering is viewed as a two-stage optical process governed by both the local pump intensity and the local density of optical states (LDOS) at the Stokes-shifted frequency, which may be independently engineered²²⁻²⁶. In this direction, cavity-controlled SERS has been demonstrated using mechanically tunable Fabry-Pérot cavities coupled to plasmonic nanoantennas, enabling sideband-resolved Raman enhancement through active cavity reconfiguration²⁷. Recently, nanoantennas have been integrated in waveguides for sideband-resolved excitation and collection²⁸. These results reveal the potential of photonic environments (reservoirs) to amplify or suppress sideband inelastic light scattering. However, in closed, mechanically tunable resonators, control remains largely device-specific and poorly suited to scalable sensing architectures such as microfluidic or lab-on-chip platforms. In addition, photonic reservoir engineering, in principle, may embrace further functionalities, beyond sideband enhancement. Indeed, both momentum and polarization may act as native control variables, enabling Stokes-sideband routing or even polarization switching within the same integrated platform. Thus far, however, the broader landscape of photonic-reservoir engineering has remained largely underexplored.

In this direction, bound states in the continuum²⁹⁻³³ (BICs) provide an open photonic platform with exceptional potential. In ideal form, BICs are non-radiative eigenstates embedded in the radiation continuum. In realistic nanophotonic structures, they appear as quasi-BICs (qBICs) with finite linewidth, high quality factor (Q), strong field enhancement, and tunable dispersion in frequency, momentum, and polarization³⁴⁻⁴⁰. These properties have made qBIC metasurfaces powerful tools for biological sensing⁴¹⁻⁵², lasing⁵³⁻⁵⁵, vortex generation^{56,57}, and nonlinear optics^{58,59}. Thereby, unlike conventional closed

35 cavities, in principle the photonic reservoir reconfiguration of the inelastic light scattering can be engineered within a planar, far-field-accessible geometry.

Here, we introduce a qBIC-enhanced SERS platform that reorganizes Raman scattering into a programmable inelastic-light process, enabling pump gating of the SERS spectrum, Stokes sideband-specific enhancement, sideband-resolved directional routing, and polarization switching. Gold
40 nanoparticle (AuNP) hot spots are integrated with a non-Hermitian photonic crystal slab (PhCS) supporting dispersive qBIC modes. The AuNPs provide the molecular-scale near-field concentration required for strong local Raman generation, providing a baseline SERS substrate enhancement factor $EF \sim 10^5$. The PhCS supplies narrowband, momentum-addressable LDOS control. This hierarchical architecture separates the Raman process into two independently programmable stages. A qBIC resonant
45 with the laser acts as a pump gate, switching the Raman response on or off through critical-coupling-mediated excitation enhancement. Then, the same or a distinct dispersive qBIC branch can enhance selected Stokes-shifted frequencies when its dispersion intersects specific molecular vibrations, producing sideband-selective, enhanced Raman emission, routed along specific directions in the momentum space. When both conditions are simultaneously satisfied, the system enters a double-
50 resonant regime in which pump injection and Stokes emission are enhanced cooperatively. The qBIC contribution enhances the baseline SERS response by roughly two orders of magnitude on average, but its most distinctive effect is sideband selectivity. When the high- Q photonic reservoir amplifies both pump and Stokes populations, it allows weak or nearly silent conventional SERS bands to be selectively promoted into dominant spectral features. In the strongest cases, the sideband-specific LDOS
55 amplification exceeds 2,000, boosting the effective SERS enhancement by more than three orders of magnitude and reaching values above 2×10^8 . This extreme response is produced when the local sites of Raman generation are surrounded by a qBIC reservoir that directs the final radiative outcome of the inelastic scattering process.

60 Biphenyl-4-thiol (BPT) molecules adsorbed on AuNPs provide the SERS reporter signal locally probing the momentum- and polarization-dependent LDOS. Angle-resolved SERS band diagrams, matched to local photoluminescence (PL) measurements, show that strong, sideband-selective Raman enhancement is deterministically driven by the PhCS high- Q dispersion. The SERS intensity maps quantitatively reproduce the modeled momentum-dependent pump–Stokes matching conditions. The normalized

65 contrast between neighboring Raman sidebands provides a self-referenced probe of the spectral gradient of the LDOS. Polarization-resolved measurements further reveal momentum-dependent switching between distinct Raman generation pathways associated with TE-like and TM-like qBIC dispersions.

RESULTS

70 Theoretical model

The central mechanism of our platform is that Raman scattering is no longer governed by a single broadband enhancement factor, but by a hierarchy of independently addressable optical channels. Spontaneous Raman scattering can be viewed as a two-step process. First, the incident laser drives molecular polarization through the local pump field. Second, the molecule emits a Stokes photon into
75 the available optical modes at the Stokes-shifted frequency. Therefore, to leading order, the detected Raman signal factorizes into the intrinsic molecular vibrational response $S_{\text{mol}}(\Omega)$, the local pump intensity $|E_{\text{pump}}(\omega_L, \theta, \sigma)|^2$, and the optical LDOS at the Stokes frequency $\rho_{\text{Stokes}}(\omega_L - \Omega, \theta, \sigma)$,

$$I_{\text{Raman}}(\Omega, \theta, \sigma) \propto S_{\text{mol}}(\Omega) |E_{\text{pump}}(\omega_L, \theta, \sigma)|^2 \rho_{\text{Stokes}}(\omega_L - \Omega, \theta, \sigma), \quad (1)$$

80

where Ω is the Raman shift, ω_L is the laser frequency, θ denotes the in-plane momentum coordinate set by the incidence angle or, for reciprocity, by the detection angle, and σ is the excitation polarization. This factorization provides the physical basis for separating Raman generation into an excitation channel and an emission channel. The complete quantitative expression used to calculate the Raman maps,
85 including the broadband plasmonic Lorentzian-like response, qBIC susceptibilities, detuning functions, and overlap roll-off factors, is given in Methods (“Theory of pump-gated and sideband-selective SERS”).

Within this framework, the hybrid system can be understood as the product of four distinct contributions. In compact form, the photonic reservoir-enhanced SERS response can be written as

90

$$I_{\text{SERS}}(\Omega, \theta, \sigma) \sim S_{\text{mol}}(\Omega) \times G_{\text{pl}}(\omega_L) G_{\text{pl}}(\omega_S) \times G_{\text{pump}}^{\text{BIC}}(\omega_L, \theta, \sigma) \times G_{\text{Stokes}}^{\text{BIC}}(\omega_S, \theta, \sigma), \quad (2)$$

with Stokes frequency $\omega_S = \omega_L - \Omega$. As schematically indicated in **Fig. 1a**, the first term is the intrinsic molecular spectrum $S_{\text{mol}}(\Omega)$, which determines the vibrational fingerprints of the Raman reporter (*e.g.*,

95 BPT). The second is the broadband plasmonic enhancement (G_{pl}) provided by the AuNP hot spots at both the pump and Stokes wavelengths. The third is the qBIC pump gate ($G_{\text{pump}}^{\text{BIC}}$), which controls whether the excitation field is efficiently coupled into the hybrid hot-spot region. The fourth is the Stokes-mode LDOS ($G_{\text{Stokes}}^{\text{BIC}}$), which determines which Raman sidebands are preferentially enhanced, suppressed, or routed through the far field. This expression highlights the key conceptual departure from conventional
 100 SERS. Plasmonics provides the baseline electromagnetic gain, while the qBICs provide a programmable photonic layer that controls the excitation and emission steps independently.

This decomposition leads to distinct operating regimes, highlighted in **Fig. 1b**. The panel illustrates a representative SERS enhancement map of BPT calculated using Eq. (2) for a single qBIC dispersive
 105 branch evolving with the incidence/observation angle. In the simulation, Δ_p represents the detuning (with respect to the laser frequency) of the branch as its resonance shifts with the observation angle, fitted to $c \sin^2 \theta + b \sin \theta + a$. The photonic branch used in this calculation is chosen to reproduce the first experimental configuration discussed below, so that Δ_p can be directly mapped onto the measured qBIC dispersion. When no qBIC channel is spectrally matched, the system behaves as a sparse plasmonic SERS
 110 substrate, and the detected signal is dominated by the broadband AuNP response (often barely detectable when the spatial density of AuNPs is small). When a qBIC is resonant with the pump laser, the excitation channel is opened: the local pump intensity is enhanced, and the Raman spectrum is globally switched on (point 1 in **Fig. 1b**). In the map, the Rayleigh channel at zero Stokes shift dominates near $\Omega = 0$, marking the resonantly enhanced pump gate (experimentally, this elastic component will be strongly
 115 suppressed by the notch filter). This pump-gating process is most efficient near the maximal local field buildup at the critical coupling condition^{33,34}, where the radiative coupling rate of the quasi-BIC is matched to the intrinsic loss rate of the hybrid plasmonic–photonic mode. This regime can be reached either by intrinsically breaking modal in-plane inversion symmetry, or by preserving the native symmetric design and opening the radiative channel extrinsically through oblique excitation³⁵. We adopt
 120 the second route making the incidence angle a functional control parameter. For the first experimental configuration with single branch qBIC reservoir, simulations predict critical coupling of the pump mode at approximately 3° (**Extended Data Fig. 1**).

Sideband selectivity then arises when a dispersive qBIC mode overlaps with a Stokes-shifted frequency.
 125 The inclined dispersive branch cuts across the vibrational manifold of BPT and selectively amplifies

those Raman lines that satisfy spectral matching (see point 2 in **Fig. 1b**). Depending on the specific PhCS design, this Stokes-matched mode can be either the same dispersive branch that also gates the pump, as in the single-branch configuration, or a distinct photonic branch, as in the multibranch configuration discussed in the last section. In both cases, spectral matching enhances the LDOS at the selected Stokes frequency and preferentially amplifies the corresponding vibrational sideband.

Finally, when pump gating and Stokes-sideband matching occur simultaneously, the system enters a double-resonant regime in which Raman generation and Raman emission are cooperatively enhanced (1×2). This further motivates the use of extrinsic symmetry breaking in this work, as permanent asymmetry would fix the pump-coupling condition by design. Instead, extrinsic control enables highlighting the cooperative pump–Stokes enhancement and momentum-space multiplexing.

The bottom line is that the qBIC resonance frequency and radiative coupling depend on in-plane momentum and polarization: this traces an open LDOS landscape that can be addressed externally by changing angle or input polarization. As schematically shown in **Fig. 1c**, the outcome is that the same hybrid platform can activate or suppress Raman scattering, select individual Stokes sidebands, redistribute Raman intensity across vibrational lines, and route inelastic emission through polarization-dependent momentum channels.

Experimental demonstration

The typical architecture of AuNP-decorated PhCS is introduced in **Fig. 2a**. The photonic host consists of a holey square lattice in silicon nitride, while the plasmonic guests are mainly 40-nm AuNP single or dimers randomly distributed on its surface. The scanning electron micrograph reveals that the typical architecture is that of one or a few dimers in each single unit cell. The sparse density was intended not to quench the local cavity Q -factor because of the large gold ohmic losses. Morphological details on AuNP clustering, coverage density and contribution to the observed signal can be found in Methods (“SEM and dark-field analysis of AuNP surface coverage and clustering”) and **Extended Data Fig. 2**. Dark-field spectra of dimers reveal a broadband plasmonic response centered near 633 nm, spectrally overlapping the main photonic branch of the PhCS, indicating good coupling with the dispersive qBIC photonic environment seen by the emitters. The NPs provide SERS generation with baseline enhancement factor

EF $\sim 1.03 \times 10^5 \pm 30\%$, as described in Methods (“Estimated baseline SERS substrate EF”). Single particles were expected not to contribute much to the SERS signal^{60,61}.

Numerical simulations based on rigorous coupled-wave analysis (RCWA) of the local field distribution in the unit cell make the hybridization mechanism explicit (**Fig. 2c,d**). Measured dielectric dispersion was included in the simulation (Methods). Without AuNPs, the optical field follows the extended dielectric qBIC mode profile, with a local intensity enhancement, relative to the incident plane wave, exceeding 200. Introducing the AuNP dimer perturbs the qBIC condition of the nearly lossless silicon nitride and redistributes part of the delocalized photonic field into the nanoscale dimer gap. This produces a plasmonic hot spot embedded within the qBIC field profile and its associated reservoir of photonic states. The attenuation of the maximum field intensity owes to radiative leakage and ohmic losses introduced by the AuNPs.

The corresponding simulated transmittance band diagram vs angle θ (momentum) is presented in **Fig. 3a**. The hybridization of the plasmonic and dielectric responses opens radiative access to a qBIC branch around normal incidence under ideal NP distribution, also producing a TE-like/TM-like hybrid dispersion indicated by the arrows. The measured angle-resolved transmittance on the AuNP-decorated PhCS reproduces the main calculated dispersive features (**Fig. 3b**). This comparison confirms that the introduction of AuNPs does not destroy the qBIC dispersion, but rather perturbs its photonic landscape.

The experimental configuration illustrated in **Fig. 3c** was designed to go beyond far-field characterization. A global transmittance spectrum measures the spatially averaged optical response of the patterned slab, but it does not by itself establish that the specific AuNPs probed in SERS experience the same local frequency–momentum LDOS landscape. To address this point, we used the angle- and polarization-resolved photoluminescence emitted locally under 532-nm excitation as a pointwise probe of the dispersive photonic environment. This PL signal arises from the combined contribution of radiative defects in silicon nitride and luminescence from AuNPs, and is reshaped by the same qBIC LDOS that governs the Raman emission channels. PL and SERS maps were therefore correlated by switching the excitation wavelength from 532 nm to 633 nm while interrogating the same microscopic region and preserving control over incidence angle and polarization. This procedure directly links the local photonic dispersion sampled by the AuNP ensemble to the subsequent angle- and polarization-dependent SERS

response. The point-probed PL dispersion reported in **Fig. 3d** reproduces the main qBIC branch observed in the angle-resolved transmittance map of **Fig. 3b**, demonstrating that the photonic reservoir sampled by the plasmonic hot spots is not disconnected from the extended slab band structure measured in transmission. Rather, the SERS-active regions inherit the same dispersive photonic LDOS that governs the expected far-field response of the PhCS. The PL local reference dispersion can be used to distinguish deterministic qBIC-mediated Raman control from uncontrolled plasmonic variability and position-dependent hotspot fluctuations.

Figure 4 shows the experimental evidence of momentum-dependent, pump-gated and sideband-selective SERS generation obtained with the single-branch PhCS. The upper row of **Fig. 4a** tracks the pointwise probed PL of the dispersive qBIC branch as the incidence angle is varied. At small angles, the resonance is narrow and weakly emissive, as expected for a qBIC with a high radiative quality factor and limited coupling to the far field. As the angle increases, the mode progressively broadens, the qBIC becomes more radiatively accessible and its PL intensity rises.

The lower row of **Fig. 4a** shows the Raman consequence of this radiative opening. In the gate-on condition, the pump is resonantly injected through the qBIC near critical coupling, enabling efficient field delivery to the AuNP hot spots and activating Raman generation. Consequently, the full BPT SERS spectrum becomes visible, with a pronounced enhancement relative to the baseline measured outside the PhCS. Before this angular window is opened near normal incidence, the SERS signal is strongly attenuated, although the AuNP dimers and the molecular reporter remain present. In this gate-off regime, the overall Raman yield is suppressed; however, an individual vibrational sideband can still emerge if its Stokes-shifted frequency overlaps with the dispersive photonic LDOS at the corresponding angle.

The 300 cm^{-1} sideband illustrates this transition particularly clearly: as the angle is varied, this mode evolves from a dominant Raman peak (because of favorable pump and Stokes matching) to an almost silent spectral feature. Similar behavior is also observed for the BPT bands near 1200 and 1590 cm^{-1} . The 1590 cm^{-1} band is especially informative, because it appears as the only Raman feature enhanced above the background floor when the dispersive LDOS selectively intersects its Stokes-shifted emission frequency.

In the present collinear pump–collection geometry, this condition is sampled by rotating the sample, so that pump coupling and Stokes collection momentum are scanned together. Since the collinear pump is far detuned from the qBIC, the same sideband-selective angular dependence can be related to the reciprocal configuration in which the sample is kept fixed (with normal incidence pump) and the far-field emission is collected at different observation angles.

In the momentum window where the pump channel is efficiently coupled through the qBIC, and the dispersive photonic branch overlaps a specific Stokes frequency as the 300 cm⁻¹ sideband, the enhancement of the Stokes intensity can be dramatic. To quantify the specific sideband enhancement we compare SERS spectra collected inside and outside the PhCS in **Fig. 4b**. As described in Methods, given a baseline EF = 1.03×10⁵, the multiplicative cascaded PhCS contribution at a given Stokes frequency and detection angle can be evaluated as

$$F_{\text{PhCS}}(\Omega, \theta) = \frac{I_{\text{SERS}}^{\text{PhCS}}(\Omega, \theta)}{I_{\text{SERS}}^{\text{AuNP}}(\Omega)}, \quad (3)$$

where $I_{\text{SERS}}^{\text{AuNP}}$ is the reference intensity measured on AuNPs outside the patterned region, and $I_{\text{SERS}}^{\text{PhCS}}$ is the SERS intensity measured on the AuNP-decorated PhCS under identical excitation and collection conditions. Importantly, F_{PhCS} is strongly angle-, wavelength-, and also polarization-dependent. Under resonant pump condition, F_{PhCS} reaches values approaching 250 (±35) for the dominant BPT sideband at 1590 cm⁻¹. The concurrent LDOS matching under double-resonant condition promotes the weak conventional SERS sideband at 300 cm⁻¹ into a dominant spectral feature, reaching a sideband-specific enhancement factor exceeding 2,000, reviving this nearly silent vibrational feature on demand and turning it into a prominent Raman channel. This largest amplification also showed the strongest site-to-site variability, decreasing to a few hundred at other probed positions because of local variations in AuNP clustering. Since F_{PhCS} multiplies the baseline SERS response, the total effective enhancement achieved on the PhCS reaches on average $F_{\text{PhCS}} \times \text{EF} \sim 2.5 \times 10^7$, with more than three orders of magnitude increase for the 300 cm⁻¹ reaching values above 2×10^8 . Notably, this mature engineering of the pump–Stokes photonic environment improves the enhancement by more than two orders of magnitude compared with our previous report⁶².

The full angle-resolved SERS maps in **Fig. 4c** provide the most complete validation. In the pump-gated region, the qBIC is critically coupled near the laser frequency and the Rayleigh/pump channel is strongly

enhanced. In the dispersive Stokes-enhancement region, the photonic branch crosses selected Raman sidebands and selectively amplifies them. The strongest Raman signal is observed where pump gating and Stokes-sideband matching overlap. The signal fades when either the pump gate closes or the Stokes branch moves away from the molecular transitions. The agreement between the reproducible angular map following the calculated/experimental qBIC dispersion rules out a purely morphological explanation based on random AuNP variability. This reversible activation and suppression provide the experimental signature of hierarchical Raman control, in which broadband plasmonic enhancement is gated and spectrally redistributed by the local photonic reservoir modulated by the LDOS evolution with momentum. The key observation is that the Raman signal appears only when the photonic mode opens an efficient excitation pathway or provides a favorable optical environment for the emitted Stokes photons.

Polarization Multiplexing and LDOS-gradient Readout

Applying the same approach to a multibranch PhCS, we further show that Raman generation is governed by the polarization channel that couples most efficiently to the local qBIC landscape. The data reported in **Fig. 5** describe how several photonic resonances may contribute to the pump and Stokes LDOS, and may shape the SERS spectrum in different ways. To elucidate the mechanism, we compare the *s*- and *p*-polarized SERS spectra at specific detection angles (where *s* and *p* denote polarizations perpendicular and parallel to the scattering plane, respectively), then correlate them with the locally probed associated PL maps (**Fig. 5a-c**), and finally correlate the PL maps with the global far-field transmittance response (**Fig. 5d**).

Figure 5a shows a representative spectral cut at $\theta = 5^\circ$. The upper panel reports the local PL spectra measured under *s*- and *p*-polarized excitation. The vertical line at $\sim 0 \text{ cm}^{-1}$ marks the fixed 633 nm pump laser. At this angle, the two polarizations show different spectral alignments with respect to the pump line, indicating that the pump experiences a different local photonic reservoir. In addition, the *s*- and *p*-polarization version of the same LDOS branch are generally slightly or strongly detuned depending on the specific angle. This difference is directly transferred to the SERS response shown in the lower panel. Specific Stokes bands, including those near 1090 and 1590 cm^{-1} , are selectively enhanced depending on the incident polarization, with a polarization-gating multiplexing ratio M_{PG} reaching values as high as ~ 25 for selected vibrational lines (Methods).

The physical meaning of this large contrast is important. It does not primarily arise from a change in the
280 intrinsic molecular Raman spectrum, which is fixed by the BPT reporter, nor can it be explained only as
the random vector alignment with the gap hot spot axis at the emission stage, since the same position
signal evolves according to the LDOS. Instead, it reflects whether a given polarization can efficiently
open the pump-assisted qBIC channel at that specific momentum. When one polarization is spectrally
matched to the pump mode, Raman generation is activated; when the orthogonal detuned pump
285 polarization is selected, the excitation channel is suppressed. Thus, polarization acts as an additional
control parameter for switching the Raman process on or off.

The local PL maps in **Fig. 5b** clarify how this polarization control emerges. Under *s*-polarized excitation,
the dominant dispersive branch follows a TE-like evolution. Under *p*-polarized excitation, the response
290 is not purely TM-like, but displays a hybrid character, with spectral features that contain contributions
from both TE-like and TM-like photonic modes. This hybridization is expected in the AuNP-decorated
PhCS, because the plasmonic NPs locally perturb the symmetry of the dielectric slab and mediate
coupling between otherwise distinct polarization channels. However, even though the local response is
hybridized, the two polarization channels do not become identical. Their dispersive slopes, spectral
295 positions, and pump detunings remain different, and these differences are sufficient to produce distinct
Raman enhancement pathways.

This point is illustrated further in **Fig. 5c**, where the spectral cut at $\theta = 7.5^\circ$ shows a different situation
from **Fig. 5a**. At this angle, the pump-region alignment of the *s*- and *p*-polarized spectra becomes more
300 similar, but the corresponding Stokes-side resonances remain differently detuned. As a consequence,
both polarizations may contribute to pump activation, while the Stokes-side LDOS still selects different
Raman bands. The resulting Stokes spectra are therefore not simple scaled copies of each other. Instead,
the 1090 and 1590 cm^{-1} vibrational modes are reshaped differently for *s*- and *p*-polarized excitation,
demonstrating that polarization affects not only the intensity of Raman generation, but also the spectral
305 redistribution of the emitted Raman signal.

For comparison, **Fig. 5d** reports the global far-field, angle-resolved transmittance maps of the PhCS
under *s*-, *p*-polarized, and summed hybrid excitation conditions. The summed response reproduces the

310 mixed features observed in the local fluorescence maps, confirming that the local LDOS sampled by the
 AuNP hot spots remains connected to the extended PhCS band structure. At the same time, the
 comparison also shows why the local measurement is essential: the Raman reporters do not probe an
 ideal, purely TE-like or TM-like band, but the hybrid local LDOS produced by the combined action of
 the PhCS and AuNPs. Therefore, the relevant optical environment for SERS is not a single universal
 LDOS landscape, but a polarization-resolved manifold of dispersive states. **Extended Data Fig. 4** further
 315 corroborates the data interpretation, where the full theoretical and experimental dispersion-enhanced
 Raman maps are compared also for this second case, where multiple branches intervene in shaping the
 SERS spectrum, and again the correlation rules out arbitrary hot spot fluctuations. In addition, it also
 illustrates the resolved TE and TM-like responses, highlighting the detuning effects on both pump and
 Stokes modes.

320 We define a polarization contrast metric to quantify the effect in **Fig. 5e**. When the same Raman mode
 at a shift Ω (e.g, at 1590 cm^{-1}) lies at different detunings for the distinct polarizations, it experiences
 different Stokes LDOS values. The relative intensity I_{SERS} , therefore, changes as the branch is tuned
 through momentum space. The dispersive polarization contrast C_{pol} , defined in Methods, quantifies this
 325 effect by tracking how the balance between *s*- and *p*-driven Raman channels evolves as the resonances
 move across the fixed pump and Stokes frequencies. Because the fixed 633 nm pump laser intersects
 these polarization-resolved manifolds at different angles, the preferred Raman excitation pathway can
 invert as the momentum is varied. In one angular window, *s*-polarized excitation can more efficiently
 gate the Raman process; in another window, *p*-polarized excitation can become dominant. This behavior
 330 transforms the device into a polarization–momentum multiplexer for Raman generation.

An additional consequence of the LDOS picture is that Raman sideband can be used to probe the photonic
 LDOS itself. When two Raman modes at shifts Ω_1 and Ω_2 lie at different detunings from the same narrow
 qBIC branch, they experience different Stokes LDOS values. If the two Raman peaks are close enough
 335 that the molecular response and plasmonic background vary slowly between them, their normalized
 difference mainly probes the spectral variation of the Stokes LDOS. The resulting LDOS-gradient
 contrast D_{12} , defined in Methods, and shown for the 1590 cm^{-1} band in **Fig. 5f**, acquires a dispersive
 shape as the qBIC branch crosses the Raman sideband. This provides a self-referenced Raman observable
 that is sensitive not simply to the magnitude of the LDOS, but to its spectral gradient (**Fig. 5f**). By

340 comparing two nearby Raman bands through a normalized intensity difference, common-mode fluctuations in pump power, molecular density, and broadband plasmonic enhancement can be partially rejected. This self-referenced observable may be particularly useful for sensing in flowing liquids, heterogeneous biointerfaces, or time-dependent surface binding assays, because small shifts of the photonic mode can be converted into differential changes between Raman sidebands.

345

DISCUSSION

This work introduces a control layer for SERS in an open resonator, in which the Raman response is both amplified and reshaped by the dispersive optical environment of a single or multibranch PhCS supporting high- Q modes. Any photonic eigenmode supporting the qBIC traces a strong photonic reservoir
350 experienced by local SERS nanoantennas, function of momentum, polarization and frequency. This photonic reservoir controls the coupling of the pump field into the hot spots and the density of optical states available to the emitted Stokes photons. In this configuration, Raman scattering becomes sensitive to momentum, polarization, and spectral position of the photonic modes.

Conventional plasmonic substrates enhance the Raman spectrum through broadband electromagnetic
355 amplification, but they offer limited control over which vibrational bands dominate the detected signal. As a result, chemically rich spectra can become congested, weak diagnostic peaks can remain hidden below stronger bands. The qBIC architecture demonstrated here adds a photonic selection rule on top of the plasmonic enhancement through a programmable redistribution of Raman intensity across selected spectral channels.

360 The strongest Raman signals are observed in the angular range where the qBIC dispersion provides simultaneous pump-channel access and favorable Stokes-sideband matching. Under these double-resonant conditions, the photonic contribution produces stable and reproducible sideband-specific enhancement factors approaching 250 beyond the baseline AuNP SERS response, with weak-sideband selective amplification exceeding 2,000 under optimal conditions. The spectral redistribution of the
365 Raman signal follows the measured and calculated photonic dispersion, with reproducible angle-dependent correlation with the qBIC branch. The combined agreement among transmittance, pointwise probed PL, analytical modeling, and angle-resolved SERS maps demonstrates that the observed modulation owes to qBIC-mediated LDOS engineering.

While Fabry–Pérot cavity–antenna systems and integrated nanobeam–antenna hybrids have
370 demonstrated the separation of pump enhancement from the Stokes optical reservoir and enabled
sideband-resolved SERS^{27,28}, the present work differs from these approaches by replacing a closed or
waveguide-addressed cavity with single or multibranch LDOS reservoirs based on open qBIC dispersion
that largely expand the capability of manipulating Raman scattering.

375 The ability to control Raman sidebands may have direct implications for practical spectroscopy. In
mixture analysis, biological media, and surface assays, the most useful molecular information is often
not the strongest Raman peak, but a weaker line that distinguishes one species, conformation, binding
state, or local chemical environment from another. A dispersive qBIC can act as an optical filter that
enhances such selected vibrational bands while suppressing less informative spectral regions. This may
380 be useful in functional systems, for separating overlapping molecular fingerprints, or emphasizing
reporter bands chosen for a specific diagnostic task. In this sense, the platform provides a possible route
to engineer the information content of the Raman spectrum.

A second implication concerns optical multiplexing. In the multibranch configuration, different qBIC
modes couple to the pump and Stokes frequencies depending on angle and polarization. This produces
385 polarization-dependent Raman activation and large polarization-gating ratios, showing that the same
molecular layer can be read through different optical pathways. Such polarization–momentum
multiplexing could be valuable for lab-on-chip and microfluidic sensing, where different analytes,
reporters, or sensing regions may be encoded not only by spectral position, but also by the angular or
polarization channel through which Raman photons are generated and collected. Unlike closed or
390 mechanically tunable cavity–antenna systems, this control is obtained in an open planar geometry that
can be more easily incorporated in typical sensing devices.

From a fundamental perspective, the platform also connects Raman scattering to non-Hermitian photonic
band engineering. The qBIC modes used here are open resonances whose linewidth, radiative coupling,
and polarization structure evolve with momentum. Raman scattering therefore becomes a probe of an
395 engineered LDOS landscape rather than only of a molecular vibrational spectrum. This opens
possibilities for studying inelastic light scattering near exceptional points³⁴, topological polarization
singularities⁶³, or designed radiative-coupling landscapes⁶⁴. Future extensions could explore stimulated
Raman regimes¹⁸, stronger molecular optomechanical coupling^{3,65}, momentum-dependent single-

molecule strong coupling⁶⁶, or active tuning of the qBIC dispersion to dynamically reconfigure the Raman response.

In previous work, we investigated the cascaded enhancement added from a narrow qBIC resonance to a plasmonic substrate⁶². Here, instead, we introduce a substantial control layer, demonstrating that Raman generation can be activated, suppressed, spectrally filtered and routed by changing the excitation/detection angle and input polarization, without cavity reconfiguration. This approach reorganizes Raman scattering into a programmable inelastic-light process, providing a route toward programmable vibrational spectroscopy and differential molecular sensing, where sensitivity, selectivity and optical routing can be engineered within the same hybrid plasmonic–photonic control of inelastic light–matter interactions.

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Main Figures and Captions

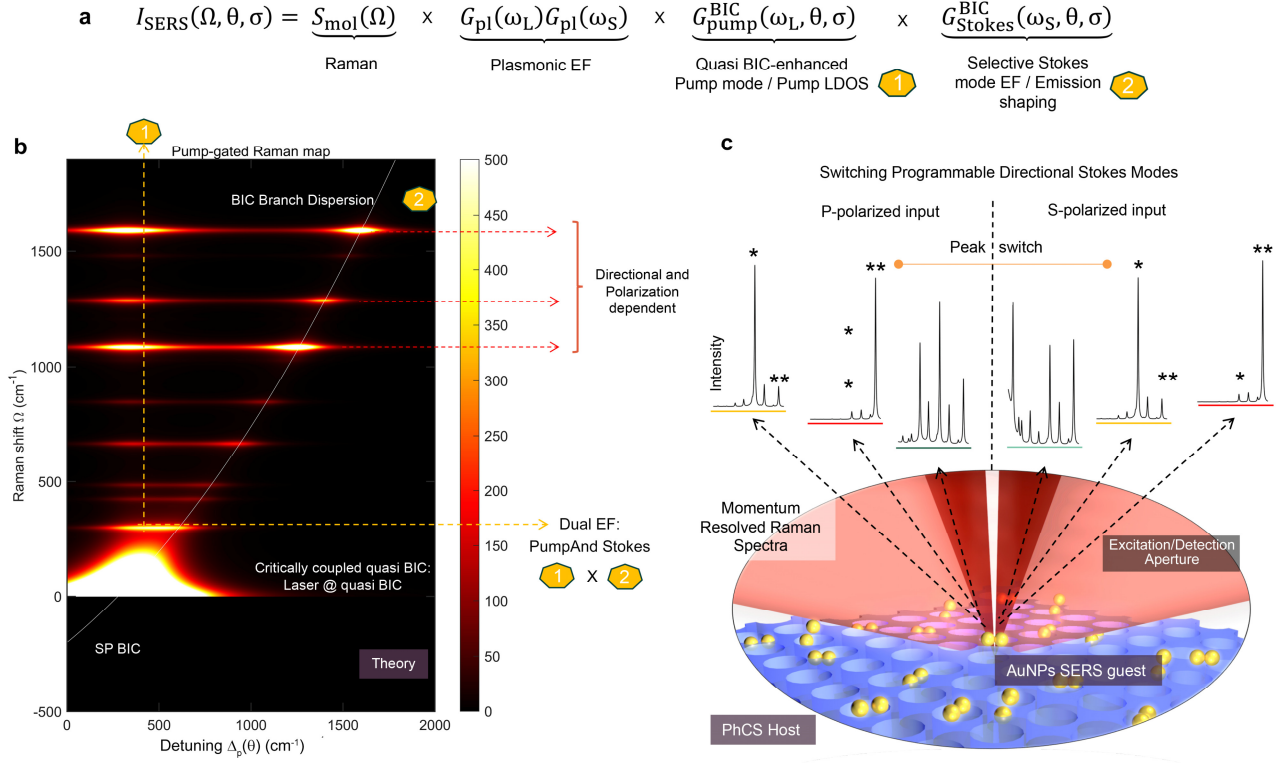


FIG. 1. Pump-gated quasi-BIC concept for programmable SERS enhancement and directional Stokes emission. **a**, Decomposition of the enhanced SERS signal $I_{\text{SERS}}(\Omega, \theta, \sigma)$ where $S_{\text{mol}}(\Omega)$ is the intrinsic Raman spectrum; $G_{\text{pl}}(\omega_L)G_{\text{pl}}(\omega_S)$ represents the plasmonic SERS enhancement; $G_{\text{pump}}^{\text{BIC}}(\omega_L, \theta, \sigma)$ is the quasi-BIC-enhanced pump LDOS (symbol 1); and finally $G_{\text{Stokes}}^{\text{BIC}}(\omega_S, \theta, \sigma)$ describes selective coupling to dispersive Stokes modes (symbol 2). The LDOS terms (1) and in particular (2) are further polarization dependent (σ), in agreement with the PhCS dispersion. **b**, Calculated pump-gated Raman map of BPT Raman reporter as a function of Raman shift Ω and detuning Δ_p . Near the quasi-BIC condition, the pump field is strongly enhanced while the dispersive BIC branch selectively amplifies Raman lines when it intersects molecular transitions, eventually enabling simultaneous pump and Stokes enhancement for specific conditions, (1)×(2). **c**, Conceptual illustration of polarization-controlled switching between directional Stokes channels. Changing the excitation polarization activates different LDOS-enhanced emission pathways, producing momentum-resolved Raman spectra with programmable spectral and directional characteristics.

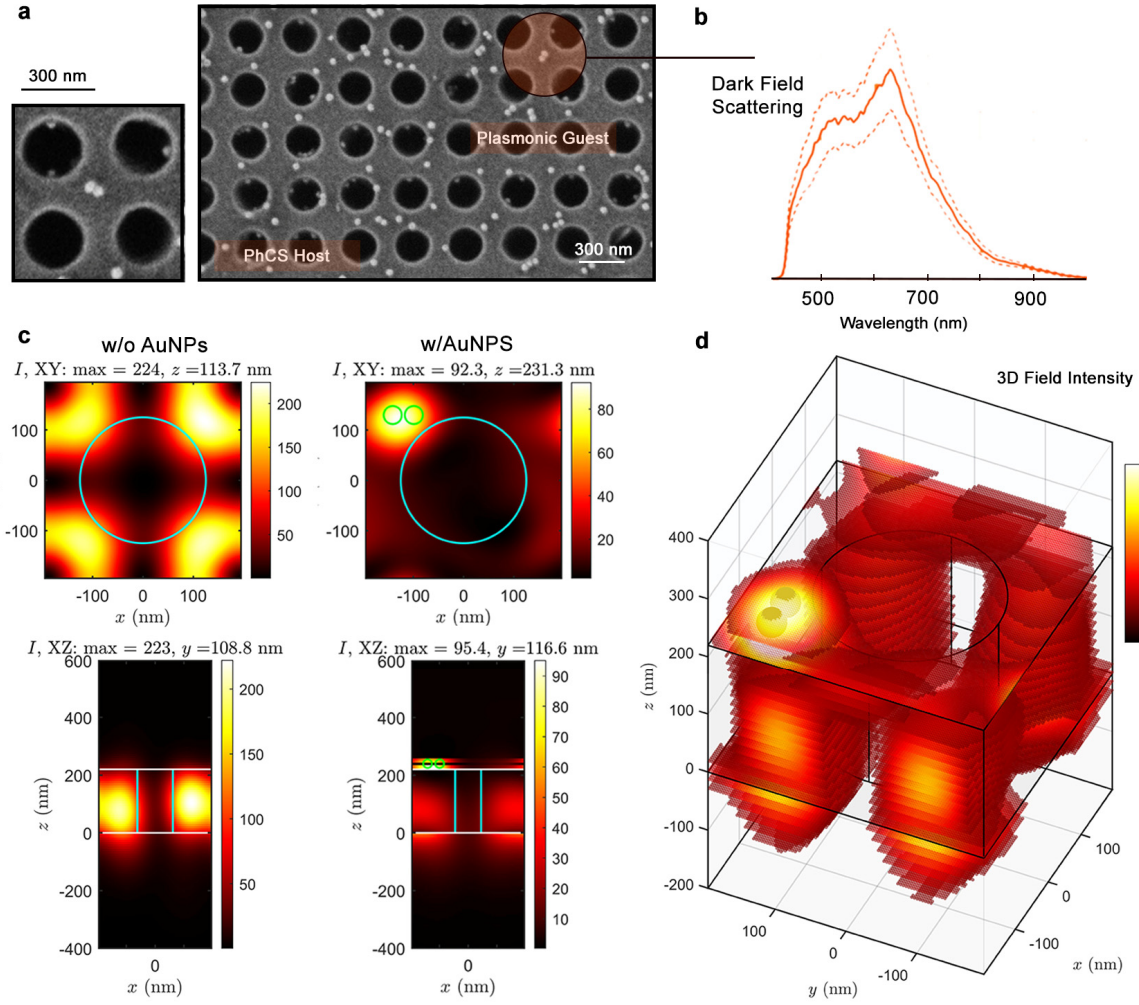


FIG. 2. Experimental platform and numerical field maps. **a**, Left: Scanning electron microscopy (SEM) images of the PhCS with a periodic array of nanoholes, where AuNP dimers and monomers are randomly distributed on the surface, acting as plasmonic guests coupled to the PhCS host. **b**, Dark-field scattering spectra of the AuNP dimer (average $\pm$ st.dev.) from 101 sampled positions. **c**, Simulated electric field intensity in the PhCS unit cell distributions without AuNPs and with AuNPs, highlighting the emergence of localized hotspots at the AuNP dimer and fully hybridized dielectric–plasmonic cavity. **d**, three-dimensional visualization of the field intensity. In the presence of the AuNPs, a strongly localized hotspot develops at the dimer while still remaining embedded within the broader quasi-BIC field landscape, with acceptable field loss of a factor 2–3. The resulting cavity is therefore neither purely plasmonic nor purely dielectric: it is a genuinely hybrid non-Hermitian resonator in which strong local field concentration and narrow dispersive LDOS control coexist in the same optical volume. This coexistence is what makes independent pump and Stokes programming possible. A main transverse-electric (TE-like) character compatible with the gap-hot post transverse geometry of the dimer is also well evident. The far-field signature of the same physics is visible in the angle-resolved transmittance maps. Numerical simulations are compared with experimental results in **Fig. 3a-b**.

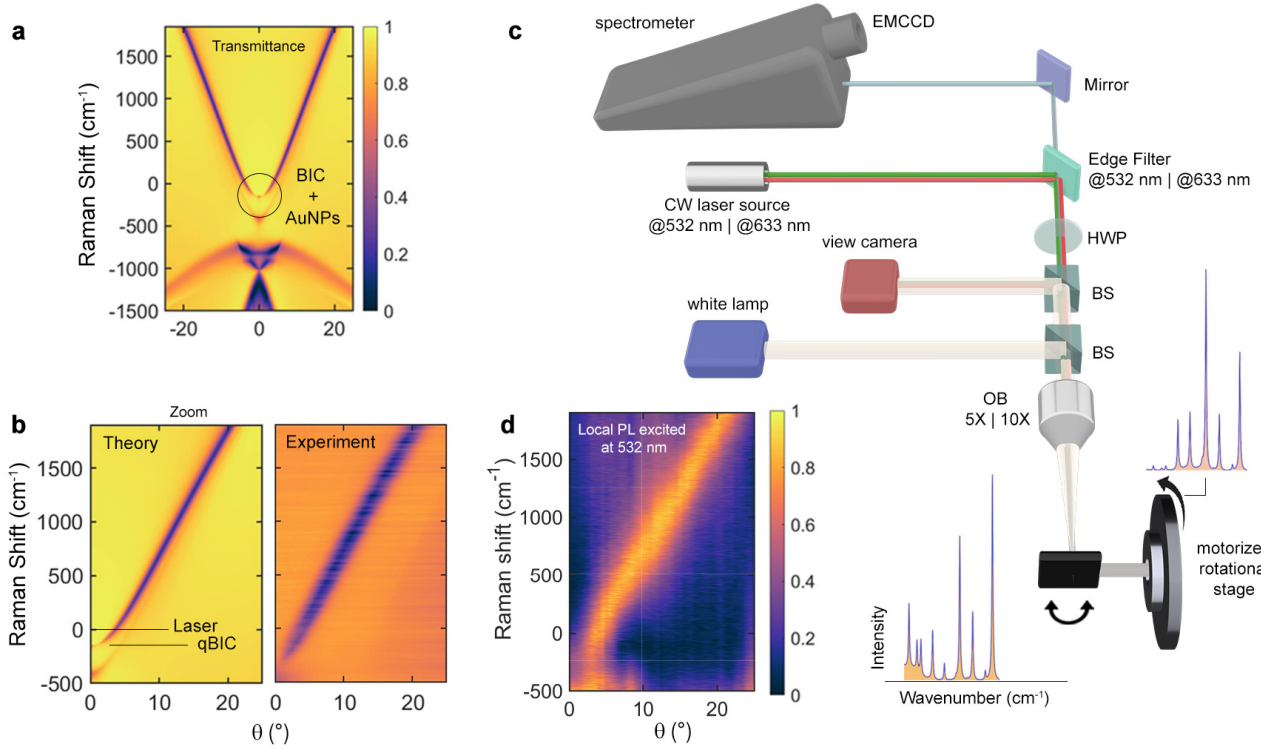


FIG. 3. Hybrid plasmonic-qBIC platform for programmable SERS. **a**, Calculated angle-resolved transmittance map showing the qBIC dispersion. The presence of AuNPs perturbs the BIC condition, enabling radiative coupling at normal incidence and emergence of a TE-like/TM-like hybrid band. **b**, Comparison between theoretical and experimental angle-resolved transmittance in units of Raman shift from 633 nm. **c**, Experimental setup for programmable SERS measurements, including dual excitation (local PL probe at 532 nm and SERS probe at 633 nm), polarization control (via half-wave plate, HWP), angle-resolved detection via a motorized rotational stage, and EMCCD-based spectral acquisition. BS: beam splitter; OB: microscope objective. **d**, Local (point-probed) photoluminescence (PL) map under 532 nm excitation, in units of Raman shift from 633 nm, confirming angle-dependent, specific local dispersion consistent with the overall quasi-BIC dispersion used to correlate local SERS emission with PhCS LDOS shaping.

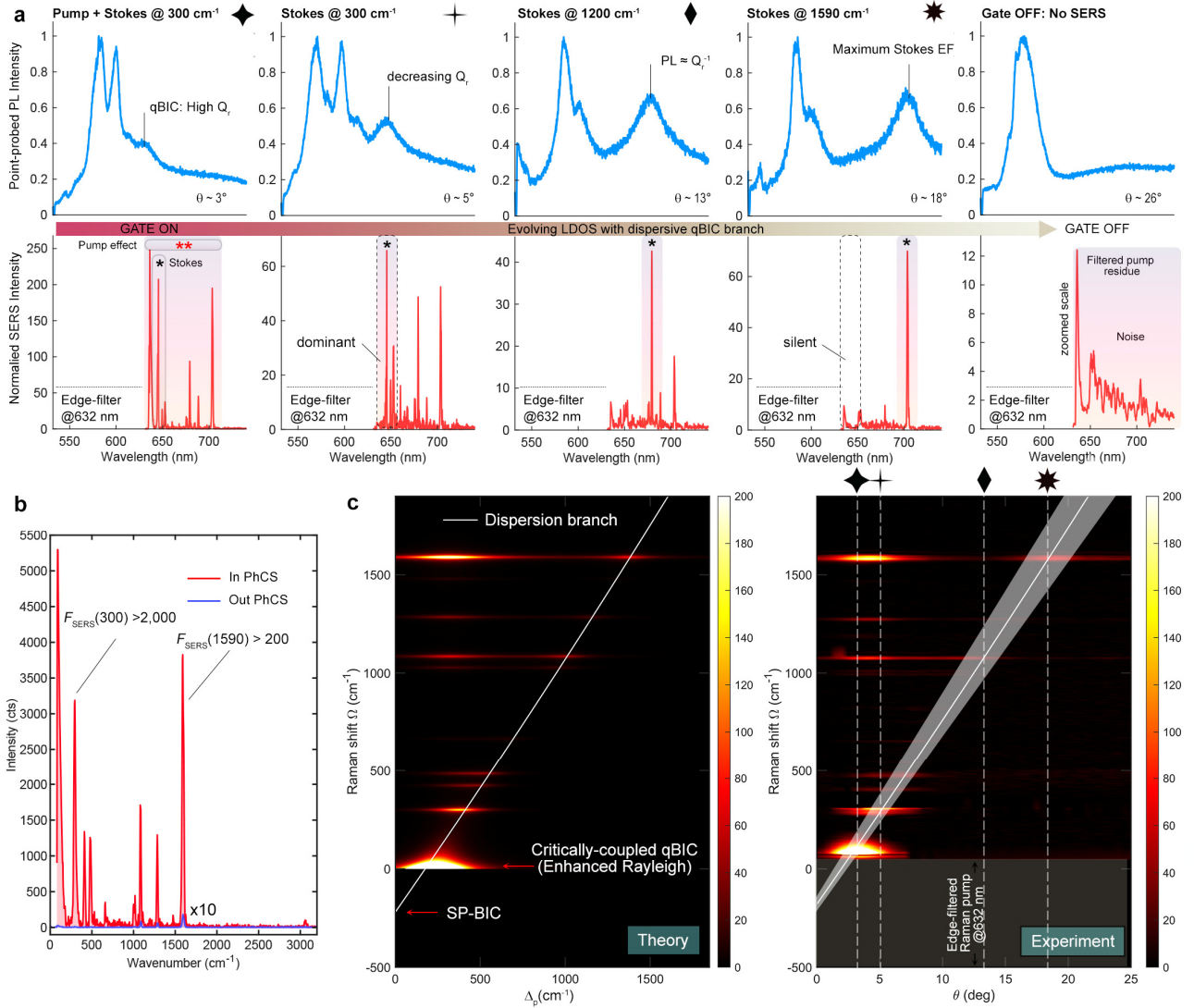
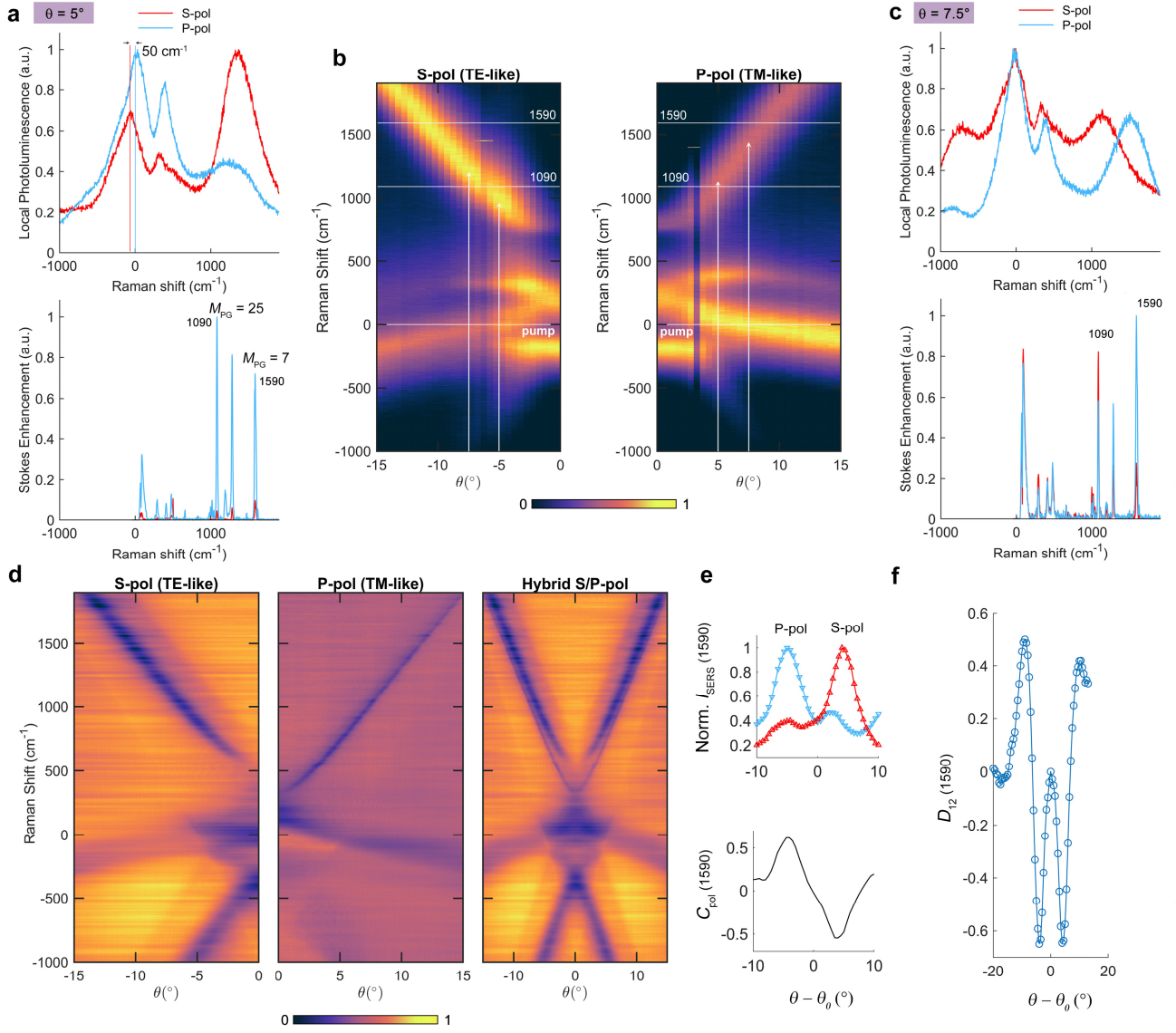


FIG. 4. Angle-dependent activation of SERS by pump-assisted qBIC gating. **a**, Top row: point-probed PL spectra measured at selected angles, showing the evolution of the dispersive qBIC branch and its angle-dependent radiative linewidth. At small angles, the mode exhibits high Q_r ; with increasing angle, Q_r decreases, the mode broadens, and the PL intensity increases approximately as Q_r^{-1} , reaching a maximum Stokes enhancement window near $\theta \sim 20^\circ$. At larger angles ($\theta \sim 26^\circ$), the pump-Stokes overlap is lost and the gate is effectively OFF. The symbols refer to the angle cuts associated with panel c. Bottom row: corresponding edge-filtered spectra acquired under 633 nm excitation, evidencing strong SERS only when both pump enhancement and Stokes-mode coupling are satisfied. The colored bar indicates the transition from the gate-on to the gate-off regime as the dispersive quasi-BIC branch evolves with angle, the characteristic sideband transition from dominant to silent and the matching of distinct sidebands with the PhCS branch peak. **b**, Representative Raman spectra collected inside and outside the PhCS, highlighting the strong enhancement of multiple Raman bands within the PhCS region. **c**, Left: theoretical Raman enhancement map as a function of Stokes detuning Δ_p and Raman shift Ω , showing the dispersive branch that mediates Stokes enhancement, the critically coupled qBIC condition near $\Omega \approx 0$ (enhanced Rayleigh channel), and the symmetry-protected BIC (SP-BIC) regime at negative detuning. Right: experimental

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SERS map versus angle θ , displaying the same angle-selective enhancement trend and confirming that strong Raman amplification occurs only where the pump condition and the dispersive Stokes branch overlap. Black symbols indicate the angular positions of the cross-sections shown in panel **a**.



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FIG. 5. Polarization-multiplexed and momentum-space control of SERS in multibranch PhCS. **a**, Local photoluminescence (PL) spectra at $\theta = 5^\circ$ under S- (red) and P-polarized (blue) excitation, together with the corresponding Stokes enhancement. The vertical line at 0 cm^{-1} denotes the pump laser at 633 nm. Pump-mode matching strongly affects the polarization-gating figure of merit M_{PG} , yielding selective enhancement of specific Raman bands (e.g., 1090 and 1590 cm^{-1}) with M_{PG} up to ~ 25 . **b**, Angle-resolved local fluorescence maps under s-polarized (TE-like) and p-polarized (TM-like) excitation. A hybrid behavior is observed, indicating near-field

hybridization between photonic quasi-BIC modes and plasmonic contributions. Vertical cuts highlight the angular positions used for spectral extraction. **c**, Local PL spectra at $\theta = 7.5^\circ$ showing polarization-dependent spectral reshaping and detuning of resonance peaks, leading to different Stokes enhancement pathways for *s* and *p* polarizations. Bottom: corresponding Stokes enhancement spectra. **d**, Experimental angle-resolved transmittance maps for *s*-pol (TE-like), *p*-pol (TM-like), and their hybrid (summed) response. The hybrid map reproduces the mixed features observed in the local fluorescence (panel **b**), confirming the coexistence and coupling of polarization channels. Despite this, the angular dispersion coefficients differ, leading to polarization-dependent detuning of resonances. **e**, Dispersive $I_{\text{SERS}}(\Omega_{\text{fixed}}, \theta, \sigma)$ and Stokes polarization contrast $C_{\text{pol}}(\Omega_{\text{fixed}}, \theta, \sigma)$ with $\Omega_{\text{fixed}} = 1590 \text{ cm}^{-1}$ as a function of angular detuning, highlighting the polarization-dependent modulation of Raman emission arising from distinct dispersion slopes and resonance conditions in *s* and *p* channels. **f**, LDOS-gradient Raman dichroism $D_{12}(1590)$ as a function of angular detuning $\theta - \theta_0$, reflecting the variation of Stokes peak intensity driven by the LDOS variation across the quasi-BIC resonance.

Materials and Methods

Synthesis and functionalization of AuNPs

Spherical AuNPs with an average diameter of $40 \pm 10 \text{ nm}$ were synthesized using a seed-mediated growth method adapted from the protocol reported in Ref.⁶⁷. Briefly, an aqueous trisodium citrate solution (2.2 mM, 150 mL) was heated to 100°C under vigorous stirring, followed by the rapid addition of HAuCl₄ (1 mL, 25 mM). After approximately 10 min, the formation of gold seeds, with an estimated diameter of $\sim 10 \text{ nm}$ and concentration of $\sim 3 \times 10^{12} \text{ NPs mL}^{-1}$, was indicated by a color change to deep red. The seed solution was then cooled to 90°C , and two sequential additions of HAuCl₄ (1 mL, 25 mM) were performed, allowing 30 min of reaction time after each addition to promote nanoparticle growth. After 1 h, 55 mL of the solution was removed, and the remaining fraction was diluted with Milli-Q water (53 mL) and trisodium citrate (2 mL, 60 mM) to obtain AuNPs with a target diameter of $40 \pm 10 \text{ nm}$.

The nanoparticle size distribution and colloidal stability were evaluated by dynamic light scattering using a Zetasizer Nano ZS system (Malvern Instruments, UK) equipped with a He–Ne laser operating at 633

670 nm, a scattering angle of 173° , and a measurement temperature of 25°C . The synthesized AuNPs were subsequently functionalized with BPT, used as the Raman reporter molecule. Functionalization was carried out by incubating the AuNP colloidal solution with BPT dissolved in ethanol at a final concentration of $5\ \mu\text{M}$, under gentle stirring at room temperature for 2 h. This process promotes molecular adsorption through thiol–gold bonding. Dynamic light scattering measurements performed
675 before and after functionalization showed no significant change in the nanoparticle size distribution, confirming that BPT adsorption did not induce substantial colloidal aggregation. The functionalized AuNP solution was stored in the dark at 4°C until use. UV–Vis absorption spectra were recorded in the 400–900 nm range using a Lambda 850+ spectrophotometer (PerkinElmer) to evaluate the plasmonic response and verify successful functionalization.

680 For assembly on the PhCSs, the AuNP suspension ($250\ \mu\text{g mL}^{-1}$) was diluted to a final concentration of $100\ \mu\text{g mL}^{-1}$. A $10\ \mu\text{L}$ droplet was deposited onto the PhCS surface and allowed to dry on a hot plate at 50°C for approximately 20 min before optical characterization. This procedure produced a sparse distribution of AuNPs on the patterned dielectric surface, including isolated nanoparticles and a minority fraction of dimers and small oligomeric aggregates, which provide the dominant plasmonic hot spots for
685 SERS under 633 nm excitation.

The SERS spectrum of BPT adsorbed on 40-nm AuNPs exhibited the characteristic vibrational fingerprint of the biphenyl moiety together with clear signatures of chemisorption through the thiol group. A strong band near $1,075\ \text{cm}^{-1}$ was assigned to the C–S stretching mode coupled to aromatic ring vibrations, confirming the formation of Au–S bonds. The most intense bands in the high-frequency
690 region, located around $1,585\text{--}1,600\ \text{cm}^{-1}$, were attributed to C=C stretching modes of the phenyl rings. Additional bands around $1,280\text{--}1,300\ \text{cm}^{-1}$ were assigned to inter-ring C–C stretching, while features in the $1,150\text{--}1,200\ \text{cm}^{-1}$ range were associated with C–H in-plane bending modes. Weaker contributions around $1,000\text{--}1,030\ \text{cm}^{-1}$ were assigned to ring-breathing vibrations, whereas bands around $1,475\text{--}1,485\ \text{cm}^{-1}$ were attributed to aromatic $\nu(\text{C}=\text{C})$ modes coupled to $\delta(\text{C}-\text{H})$ in-plane bending. Low-frequency
695 bands observed near 300 , 415 , and $480\ \text{cm}^{-1}$ were assigned to vibrational modes associated with thiolate chemisorption on gold, including Au–S stretching, C–S bending, and coupled aromatic ring deformation modes. These features, which are absent or much weaker in conventional Raman spectra, provide evidence of BPT adsorption as a surface-bound thiolate species and confirm the formation of stable Au–S interfacial bonding. Overall, the measured spectral profile was consistent with literature reports and

700 confirmed both successful functionalization of the plasmonic substrate and efficient SERS activity of BPT.

Estimated baseline SERS substrate EF

The SERS substrate enhancement factor (EF) of BPT on 40-nm AuNPs was estimated by assuming that BPT forms a self-assembled monolayer on the Au surface because of the strong affinity of the thiol group
705 for gold. The baseline EF was calculated as

$$EF = \frac{I_{\text{SERS}}^{\text{AuNP}}}{I_{\text{Raman}}} \frac{N_{\text{Raman}}}{N_{\text{SERS}}},$$

where $I_{\text{SERS}}^{\text{AuNP}}$ and I_{Raman} are the SERS and normal Raman intensities, respectively, and N_{SERS} and N_{Raman} are the corresponding numbers of molecules contributing to the detected signals. The number of molecules probed in the conventional Raman configuration was estimated as

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$$N_{\text{Raman}} = \frac{A_{\text{Laser}} h \rho_{\text{BPT}} N_{\text{A}}}{M},$$

where A_{Laser} is the laser spot area, calculated by considering a beam waist of approximately $3 \mu\text{m}$, h is the Rayleigh length, taken as approximately $10 \mu\text{m}$, $\rho_{\text{BPT}} = 1.1 \text{ g cm}^{-3}$ is the density of BPT, $N_{\text{A}} = 6.022 \times 10^{23} \text{ mol}^{-1}$ is Avogadro's number, and $M = 186.3 \text{ g mol}^{-1}$ is the molecular weight of BPT.

The number of molecules contributing to the SERS signal was estimated as

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$$N_{\text{SERS}} = \pi R^2 A_{\text{Laser}} \sigma_{\text{AuNP}} \sigma_{\text{BPT}},$$

where πR^2 is the projected area of a 40-nm AuNP, σ_{AuNP} is the surface density of AuNPs, and σ_{BPT} is the BPT surface coverage, taken as $4 \text{ molecules nm}^{-2}$. Reference SERS measurements were performed on AuNPs deposited onto glass coverslips. Using this procedure, the baseline SERS enhancement factor of BPT on the 40-nm AuNP substrate was estimated to be approximately $1.03 \times 10^5 \pm 30\%$.

720 SEM and dark-field analysis of AuNP surface coverage and clustering

The surface distribution, aggregation state, and optical response of the AuNPs deposited on the PhCS were analyzed by combining SEM and dark-field hyperspectral imaging (HSI). SEM images acquired over

representative regions of the patterned substrate were used to quantify the AuNP surface coverage and classify the deposited particles as isolated single nanoparticles or clustered configurations, including dimers, trimers, and small oligomeric aggregates. Bright AuNP features were segmented from the holey silicon nitride background and analyzed morphologically. The total (aggregated plus isolated) NP number density was approximately $10.6 \mu\text{m}^{-2}$. Among the detected AuNP population, approximately 74% corresponded to isolated single nanoparticles, whereas approximately 26% was associated with clustered AuNP configurations. This corresponds to an areal density of clustered AuNP objects of approximately 2.8 clusters μm^{-2} , i.e. nearly 3 clusters μm^{-2} .

This distribution is important for the interpretation of the SERS response. Although isolated AuNPs represent the majority of the deposited particles, their contribution to the measured SERS signal is expected to be marginal under 633 nm excitation, because individual 40-nm AuNPs provide comparatively weak local field confinement at this wavelength. By contrast, dimers, trimers, and small oligomers support stronger interparticle electromagnetic hot spots and therefore dominate the SERS-active population. Considering a laser spot diameter of approximately $6 \mu\text{m}$, corresponding to an illuminated area of about $28 \mu\text{m}^2$, each SERS sampling point probes on average approximately 79 clustered AuNP hot spots. Therefore, despite the random spatial distribution and random in-plane orientation of the clusters, the measured SERS signal is statistically averaged over a sufficiently large ensemble of hot spots and polarization orientations within each acquisition point. This ensemble averaging reduces the impact of individual dimer orientation or local morphological disorder and supports the interpretation of the measured angular and polarization trends as arising primarily from the quasi-BIC photonic environment rather than from a single accidental plasmonic configuration.

Dark-field hyperspectral imaging was then performed to investigate the optical scattering properties of the AuNPs on the PhCS. Measurements were carried out using an upright optical microscope (Evident BX43, Evident) equipped with a dark-field oil condenser and coupled to a hyperspectral imaging system (CytoViva). Samples were imaged under dark-field illumination using a $60\times$ objective, enabling the collection of scattered light from both isolated and clustered AuNPs on the PhCS. Hyperspectral image cubes (x, y, λ) were acquired over the visible–near-infrared spectral range defined by the system configuration, allowing the extraction of a full scattering spectrum from each sampled pixel. The system was equipped with a broadband illumination source, and acquisition parameters were kept constant for all measurements to ensure data comparability, with an integration time of 1 s, 100% illumination, and

696 acquired lines, for a total number of pixels $101 \times 101 = 10,201$ (scattering spectra) acquired in each area. The oil dark-field condenser enhanced the signal-to-noise ratio by selectively collecting scattered light while minimizing direct illumination.

Background spectra were acquired from nanoparticle-free regions of the substrate and subtracted from the sample spectra to correct for contributions from the glass slide and surrounding medium. Spectral data were processed and analyzed using the proprietary software provided by the manufacturer (CytoViva), enabling the extraction of spectral signatures and the determination of the localized surface plasmon resonance features of the AuNPs. Approximately 10,201 pixels were sampled over representative AuNP-decorated regions. The extracted spectra were grouped according to their dominant scattering maximum. Scattering features peaked near 532 nm were mainly assigned to isolated AuNPs, whereas red-shifted spectra peaked near 633 nm were attributed predominantly to dimers and oligomeric clusters. The scattering signal associated with the cluster-dominated spectral population peaked near 633 nm was approximately seven times larger than that associated with the 532-nm-peaked population. This confirms that clustered AuNPs provide the dominant optical contribution at the Raman excitation wavelength and supports the assignment of the measured SERS response primarily to dimeric and oligomeric AuNP hot spots embedded in the quasi-BIC photonic environment.

Theory of pump-gated and sideband-selective SERS

The Raman response of the hybrid plasmonic–photonic platform was modeled by separating the intrinsic molecular Raman spectrum from the optical enhancement channels associated with the plasmonic hot spots and with the dispersive quasi-BIC modes of the photonic-crystal slab. In spontaneous Raman scattering, the detected Stokes intensity at Raman shift Ω is proportional to the local pump intensity and to the optical local density of states (LDOS) available at the Stokes-shifted frequency. The Raman signal can therefore be written, to leading order, as

$$I_{\text{Raman}}(\Omega, \theta, \sigma) \propto S_{\text{mol}}(\Omega) |E_{\text{pump}}(\omega_L, \theta, \sigma)|^2 \rho_{\text{Stokes}}(\omega_L - \Omega, \theta, \sigma), \quad (1)$$

where $S_{\text{mol}}(\Omega)$ is the intrinsic molecular Raman spectrum, ω_L is the laser frequency, θ is the incidence angle, and σ denotes the input polarization. This factorization provides the basis for describing the hybrid system as a product of broadband plasmonic enhancement and narrowband photonic LDOS control.

The intrinsic Raman response of biphenyl thiol was reconstructed from the experimentally measured reference SERS spectrum on AuNPs outside the photonic crystal region and shown in **Extended Data Fig. 3**. The spectrum was modeled as a sum of Lorentzian vibrational lines,

$$S_{\text{mol}}(\Omega) = \sum_n A_n \frac{\Gamma_n^2}{(\Omega - \Omega_n)^2 + \Gamma_n^2}, \quad (2)$$

785 where Ω_n , Γ_n , and A_n are, respectively, the Raman shift, linewidth, and relative amplitude of the n -th vibrational mode. This reconstructed molecular spectrum was then used as the input response for calculating the photonic reshaping induced by the quasi-BIC modes.

The plasmonic contribution was described phenomenologically as a broadband Lorentzian enhancement
790 centered at the localized surface plasmon resonance of the AuNP ensemble,

$$G_{\text{pl}}(\lambda) = G_{\text{pl},0} \frac{\Delta\lambda_{\text{pl}}^2}{(\lambda - \lambda_{\text{pl}})^2 + \Delta\lambda_{\text{pl}}^2}, \quad (3)$$

where λ_{pl} and $\Delta\lambda_{\text{pl}}$ are the plasmonic resonance wavelength and linewidth, respectively. Since the electromagnetic SERS enhancement acts at both excitation and emission stages, the total plasmonic factor was taken as

$$795 \quad G_{\text{pl}}^{\text{tot}}(\Omega) = G_{\text{pl}}(\lambda_L)G_{\text{pl}}(\lambda_S), \quad (4)$$

where λ_L is the pump wavelength and λ_S is the Stokes wavelength corresponding to Raman shift Ω . Of course, wavelengths and angular frequencies are related by $\omega = 2\pi c/\lambda$, with $\omega_L = 2\pi c/\lambda_L$ for the pump and $\omega_S = 2\pi c/\lambda_S$ for the Stokes photon (similarly for λ_{pl}); the Raman shift is then $\Omega = \omega_L - \omega_S$, or
800 equivalently $\Omega/2\pi c = 1/\lambda_L - 1/\lambda_S$ in cm^{-1} . Thus, $\Omega = 0$ corresponds to the 633 nm pump line, while positive Raman shifts correspond to Stokes photons at longer wavelengths and lower optical frequencies. This term accounts for the broadband baseline enhancement provided by the AuNP hot spots. Assuming a weak angular dependence of the plasmonic contribution, the SERS spectrum of BPT outside the PhCS directly provides the product quantity $S_{\text{SERS,BPT}} = S_{\text{mol}}(\Omega) \times G_{\text{pl}}(\omega_L)G_{\text{pl}}(\omega_S)$, which is the baseline
805 SERS spectrum simulated in the angle-resolved SERS maps and used to probe the additional gating and LDOS angular dependencies introduced by the PhCS.

The photonic contribution was modeled using a minimal temporal coupled-mode description of the quasi-BIC modes. Each photonic channel (j), corresponding either to a pump-like or Stokes-like quasi-BIC branch, was represented by the susceptibility

$$\chi_j(\omega, \theta, \sigma) = \frac{1}{i[\omega - \omega_j(\theta, \sigma)] + \gamma_j/2}, \quad (5)$$

where $\omega_j(\theta, \sigma)$ is the polarization-dependent angular dispersion of the quasi-BIC mode and γ_j is its total linewidth. The photonic LDOS associated with this mode scales as $\rho_j(\omega, \theta, \sigma) \propto |\chi_j(\omega, \theta, \sigma)|^2$.

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The quasi-BIC pump gate was obtained by evaluating the relevant pump-mode susceptibility at the laser frequency. The corresponding pump enhancement factor was written as

$$G_{\text{pump}}^{\text{BIC}}(\theta, \sigma) = 1 + A_p \frac{\Gamma_p^2}{\Delta_p^2(\theta, \sigma) + \Gamma_p^2}, \quad (6)$$

820 where A_p is the pump-enhancement amplitude, Γ_p is the effective pump-mode linewidth, and

$$\Delta_p(\theta, \sigma) = \omega_L - \omega_p(\theta, \sigma) \quad (7)$$

is the detuning between the laser frequency and the pump quasi-BIC branch. This factor describes the opening or suppression of the Raman excitation channel through momentum- and polarization-dependent matching of the pump to the quasi-BIC resonance.

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The Stokes-sideband enhancement was described analogously by evaluating the photonic LDOS at the Stokes frequency $\omega_S = \omega_L - \Omega$. For a given vibrational shift Ω , the detuning from the Stokes quasi-BIC branch was defined as

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$$\Delta_S(\Omega, \theta, \sigma) = \omega_L - \Omega - \omega_S^{\text{BIC}}(\theta, \sigma), \quad (8)$$

leading to the enhancement factor

$$G_{\text{Stokes}}^{\text{BIC}}(\Omega, \theta, \sigma) = 1 + A_S \frac{\Gamma_S^2}{\Delta_S^2(\Omega, \theta, \sigma) + \Gamma_S^2}, \quad (9)$$

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where A_S and Γ_S are the amplitude and linewidth of the Stokes-mode enhancement. To account phenomenologically for reduced spectral and spatial overlap between high-frequency Raman sidebands and the photonic near-field mode, a roll-off factor was included,

$$R(\Omega) = \exp \left[- \left(\frac{\Omega}{\Omega_c} \right)^2 \right], \quad (10)$$

where Ω_c is the characteristic Raman-shift scale over which the overlap decreases. The effective Stokes enhancement was therefore taken as

$$\tilde{G}_{\text{Stokes}}^{\text{BIC}}(\Omega, \theta, \sigma) = 1 + R(\Omega) A_S \frac{\Gamma_S^2}{\Delta_S^2(\Omega, \theta, \sigma) + \Gamma_S^2}. \quad (11)$$

Combining the intrinsic molecular response with the plasmonic and photonic contributions, the calculated SERS intensity was written as

$$I_{\text{SERS}}(\Omega, \theta, \sigma) = \zeta S_{\text{mol}}(\Omega) G_{\text{pl}}(\lambda_L) G_{\text{pl}}(\lambda_S) G_{\text{pump}}^{\text{BIC}}(\theta, \sigma) \tilde{G}_{\text{Stokes}}^{\text{BIC}}(\Omega, \theta, \sigma), \quad (12)$$

where ζ is an overall proportionality constant that accounts for collection efficiency, molecular surface density, acquisition parameters, and normalization. This expression was used to reproduce the angle-dependent Raman maps and to identify the regimes of pump gating, Stokes-sideband selectivity, and double-resonant enhancement.

For the simulated Raman maps, the quasi-BIC dispersion $\omega_j(\theta, \sigma)$ ($j = \text{pump/Stokes}$) was obtained either from RCWA-calculated band diagrams or from Lorentzian fits to the experimentally measured local photoluminescence dispersion, and $\Delta_p(\theta, \sigma) = \omega_L - \omega_j(\theta, \sigma)$ fitted $c \sin^2 \theta + b \sin \theta + a$. The calculated dispersion was then converted into Raman-shift units referenced to the 633 nm excitation laser. The pump mode corresponds to spectral matching near $\Omega = 0$, whereas the Stokes enhancement occurs when the dispersive branch intersects the molecular Raman bands at finite Ω . This procedure allowed direct comparison between theoretical Raman enhancement maps and the experimentally acquired angle-resolved SERS maps.

Theory of polarization-dependent pump gating

To describe polarization switching, the pump-mode detuning was generalized to include the polarization-resolved photonic dispersion,

$$\Delta_p(\theta, \sigma) = \omega_L - \omega_p(\theta, \sigma), \quad (13)$$

870 with $\sigma = s, p$. The Raman intensity for a given polarization was therefore written as

$$I_\sigma(\Omega, \theta) = \zeta S_{\text{mol}}(\Omega) G_{\text{pl}}(\lambda_L) G_{\text{pl}}(\lambda_S) G_{\text{pump}}^{\text{BIC}}(\theta, \sigma) \tilde{G}_{\text{Stokes}}^{\text{BIC}}(\Omega, \theta, \sigma). \quad (14)$$

In the experimentally relevant regime where the dominant polarization dependence arises from the pump channel, the ratio between Raman signals measured under orthogonal input polarizations directly maps the relative pump-gating efficiency. The normalized polarization contrast was defined as

$$C_{\text{pol}}(\Omega, \theta) = \frac{I_s(\Omega, \theta) - I_p(\Omega, \theta)}{I_s(\Omega, \theta) + I_p(\Omega, \theta)}. \quad (15)$$

A sign change of C_{pol} as a function of angle indicates inversion of the preferred Raman generation channel, corresponding to momentum-dependent switching between the polarization-resolved quasi-BIC branches. When the Stokes-shift (Ω) dependent factor is comparable in the two channels, Eq. (15) shows that the measured polarization asymmetry predominantly maps the polarization dependence of pump-resonance matching. **Figure 5a** highlights a detuning of 50 cm^{-1} . This regime is best interpreted as a polarization-selective pump-gating effect. At a given incidence angle, one polarization can be nearly resonant with the pump mode (quasi-BIC or another band) and thus strongly enhances Raman generation, while the orthogonal polarization remains detuned and yields a suppressed signal. A useful figure of merit for the multiplexing strength is the polarization gating ratio

$$M_{\text{PG}}(\Omega, \theta) = \frac{I_s(\Omega, \theta)}{I_p(\Omega, \theta)}, \quad (16)$$

or its inverse when the P channel is dominant. Values $M_{\text{PG}} \gg 1$ or $M_{\text{PG}} \ll 1$ indicate strong polarization selectivity of Raman generation, while the sign change of C_{pol} across momentum identifies the inversion of the preferred pump channel. This behavior provides a direct route to angle-programmable Raman switching, polarization-encoded vibrational readout, and momentum-space routing of inelastically scattered light.

895 Theory of LDOS-gradient Raman contrast

To quantify sideband-selective Raman redistribution, we considered two Raman bands centered at shifts Ω_1 and Ω_2 , with detected intensities I_1 and I_2 . Their normalized contrast was defined as

$$D_{12}(\theta, \sigma) = \frac{I_1(\theta, \sigma) - I_2(\theta, \sigma)}{I_1(\theta, \sigma) + I_2(\theta, \sigma)}. \quad (17)$$

900 When the two sidebands are sufficiently close in Raman shift, the intrinsic molecular response and broadband plasmonic enhancement vary weakly between Ω_1 and Ω_2 . Under this condition, the contrast is governed primarily by the Stokes LDOS,

$$D_{12}(\theta, \sigma) \simeq \frac{\rho(\omega_1, \theta, \sigma) - \rho(\omega_2, \theta, \sigma)}{\rho(\omega_1, \theta, \sigma) + \rho(\omega_2, \theta, \sigma)}, \quad (18)$$

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where $\omega_i = \omega_L - \Omega_i$. Introducing the mean Stokes frequency $\bar{\omega} = \frac{\omega_1 + \omega_2}{2}$ and the sideband separation $\delta\omega = \omega_1 - \omega_2$, the LDOS can be expanded to first order as

$$\rho(\omega_{1,2}) \simeq \rho(\bar{\omega}) \pm \frac{\delta\omega}{2} \frac{\partial \rho}{\partial \omega} \bigg|_{\bar{\omega}}. \quad (19)$$

Substitution into the normalized contrast gives

$$910 \quad D_{12}(\theta, \sigma) \simeq \frac{\delta\omega}{2} \frac{\partial \ln \rho}{\partial \omega} \bigg|_{\bar{\omega}}. \quad (20)$$

Thus, the normalized difference between two neighboring Raman sidebands provides a self-referenced measurement of the spectral gradient of the photonic LDOS. Where the LDOS changes rapidly with frequency, this quantity acquires a dispersive line shape and changes sign across resonance. This observable was used to identify LDOS-gradient Raman dichroism and to distinguish sideband redistribution from common-mode fluctuations in pump power, molecular coverage, or broadband plasmonic enhancement.

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Numerical simulations

Electromagnetic simulations were performed using the rigorous coupled-wave analysis method implemented in Ansys Lumerical 2023. The simulated structure consisted of a silicon nitride photonic-crystal slab with square symmetry on a silica substrate. The optical dispersion was accurately measured through variable angle spectral ellipsometry and presented in Ref.³³. The nominal geometrical parameters were lattice spacing $a=365$ nm, slab thickness $h=180$ nm, and hole radius $r=117$ nm. The wavelength-

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dependent refractive index of silicon nitride was obtained from variable-angle spectroscopic ellipsometry, while the silica substrate was modeled using its measured dispersion. Up to 100 Fourier components were used, with an angular resolution of 0.01° for the calculated band diagrams and spectral maps.

To reproduce the hybrid plasmonic–photonic platform, a dimer composed of two 40-nm AuNPs was introduced in the photonic-crystal unit cell. Simulations were performed both without and with the AuNP dimer to evaluate the perturbation of the quasi-BIC dispersion, the opening of radiative coupling channels, and the redistribution of the near-field intensity. Electric-field intensity maps were calculated at the relevant quasi-BIC wavelengths to compare the bare dielectric mode profile with the hybridized plasmonic–photonic hot-spot configuration. TE-like and TM-like responses were obtained by changing the input polarization and incidence geometry. The simulated photonic dispersions were converted into Raman-shift units relative to the 633 nm excitation wavelength to enable direct comparison with the measured angle-resolved SERS maps.

Experimental angle-resolved SERS and local PL

Angle-resolved SERS and local photoluminescence measurements were performed using a modified WiTec Alpha-300 RAS confocal Raman microscope equipped with 532 nm and 633 nm continuous-wave excitation lasers. The 532 nm laser was used to probe local photoluminescence from the photonic-crystal slab and AuNP-modified regions, allowing reconstruction of the local quasi-BIC dispersion. The 633 nm laser was used for SERS excitation of biphenyl thiol molecules adsorbed on the AuNPs.

The sample was mounted on a motorized rotational stage to vary the incidence angle (θ) while maintaining the optical focus on the same region of the photonic crystal. The excitation beam was focused using a $5\times$ or $10\times$ microscope objective, depending on the desired spot size and angular acceptance. Polarization control was achieved using a half-wave plate, enabling selective excitation of the relevant TE-like and TM-like quasi-BIC channels. A white-light source was used for alignment and for comparison with far-field transmittance measurements.

The emitted signal was collected through the same objective, dispersed by a spectrometer, and detected using an EMCCD camera. Local PL spectra were acquired as a function of angle and polarization to map

955 the photonic LDOS sampled by the AuNP hot spots. SERS spectra were then acquired under 633 nm
excitation at the same or equivalent spatial positions and converted into Raman-shift units referenced to
the laser wavelength. Correlating the local PL dispersion with the angle-dependent Raman maps allowed
the observed SERS enhancement and spectral redistribution to be assigned to quasi-BIC-mediated pump
gating and Stokes LDOS shaping, rather than to uncontrolled spatial variations of the AuNP distribution.

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Evaluation of photonic contribution to EF

The Raman enhancement associated with the PhCS region was evaluated by comparing SERS spectra
acquired on AuNP-decorated regions inside the photonic crystal with reference spectra acquired on
AuNP-decorated regions outside the patterned area. Spectra were corrected for acquisition conditions,
965 integration time, laser power, and background. The global enhancement factor, given by the product of
the photonic contribution and baseline SERS substrate EF, $F_{\text{PhCS}}(\Omega, \theta) \times EF$, was then obtained from
the ratio between the integrated intensity of selected Raman bands measured under quasi-BIC-matched
conditions and the corresponding intensity measured on the reference plasmonic substrate outside the
PhCS. For angle-dependent measurements, enhancement maps were obtained by normalizing each
970 Raman spectrum to the reference AuNP response and tracking the intensity of selected vibrational bands
as a function of incidence angle and polarization. The maximum enhancement was identified under
double-resonant conditions, where the pump laser was matched to a quasi-BIC-assisted excitation
channel and the Stokes-shifted Raman band simultaneously overlapped a dispersive quasi-BIC LDOS
branch.

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In detail, spatial reproducibility of the PhCS-mediated Raman enhancement was evaluated by acquiring
SERS spectra from multiple independent positions on the AuNP-decorated PhCS and from reference
AuNP regions outside the patterned area under identical acquisition conditions. The PhCS enhancement
factor was calculated as $F_{\text{PhCS}}(\Omega, \theta) = I_{\text{SERS}}^{\text{PhCS}}(\Omega, \theta) / I_{\text{SERS}}^{\text{AuNP}}(\Omega)$, where $I_{\text{SERS}}^{\text{PhCS}}$ and $I_{\text{SERS}}^{\text{AuNP}}$ are the integrated
980 Raman intensities measured on and off the PhCS, respectively, for the same vibrational band and optical
configuration. Across repeated sampling positions ~ 30 , the maximum enhancement factor reached
 $F_{\text{PhCS}} = 250 \pm 35$, with a coefficient of variation of approximately 14%. This uncertainty reflects the
combined contribution of AuNP coverage fluctuations and local cluster-density variations. This
observation indicates that the effect is not caused by a single unusually bright plasmonic hot spot.

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The polarization-gating response was quantified by measuring the same Raman bands under orthogonal pump/analyzer polarization configurations. The polarization-gating ratio was defined according to Eq. (16). Integrated Raman intensities of a selected vibrational band in the high- and low-transmission polarization states, respectively, were compared. For the most strongly gated Raman bands, M_{PG} reached
990 25 ± 4 across repeated spatial positions. This confirms that the switching response is not determined by the orientation of individual AuNP dimers, but by the polarization-selective quasi-BIC LDOS sampled by an ensemble of randomly oriented hot spots.

Finally, the agreement between the measured angular positions of the SERS maxima and the predicted
995 quasi-BIC/Stokes-sideband matching angles was quantified according to the following routine. For each selected Raman band, the expected matching angle was extracted from the measured or simulated quasi-BIC dispersion by imposing spectral overlap between the dispersive photonic branch and the Stokes-shifted emission frequency. The detuning with angle was considered as the function $\Delta_p(\theta) = c\theta^2 + b\theta + a$ to account for the typical dispersion of the photonic branch (when sufficiently distant from
1000 interfering modes). The experimentally measured SERS maximum was then obtained by fitting the angular dependence of the integrated Raman intensity. The coefficients c , b and a were found by least square minimization of the residuals between model and experiment. The measured and predicted matching angles showed a Pearson correlation coefficient of $r = 0.94$, with a root-mean-square angular deviation of 0.6° . This demonstrates that the angular redistribution of the Raman signal follows the
1005 dispersive quasi-BIC resonance.

DATA AVAILABILITY

Data supporting the findings of this study are available from the corresponding author upon reasonable request.

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AUTHOR CONTRIBUTIONS

B.M., S.R., and G.Z. conceived the conceptual framework and designed the experiments. S.D., A.K.,
A.G. and A.M.S. fabricated photonic crystal slabs. B.M. and S.R. carried out experiments and sample
characterization. G.Z. and B.M. performed numerical simulations; G.Z. developed the theoretical model.
1025 B.M., T.N., S.R., G.Z. and F.D.O. verified results and cross-validated experimental and theoretical data.
B.M., S.R., and G.Z. wrote the original draft. T.N. and F.D.O. contributed to manuscript review and
editing. F.D.O. contributed funding support and access to computational resources. S.R. and G.Z.
coordinated and supervised the research, with contributions from F.D.O. All authors discussed the results
and contributed to the final manuscript preparation.

1030 **CONFLICT OF INTEREST**

The authors declare no conflict of interest.

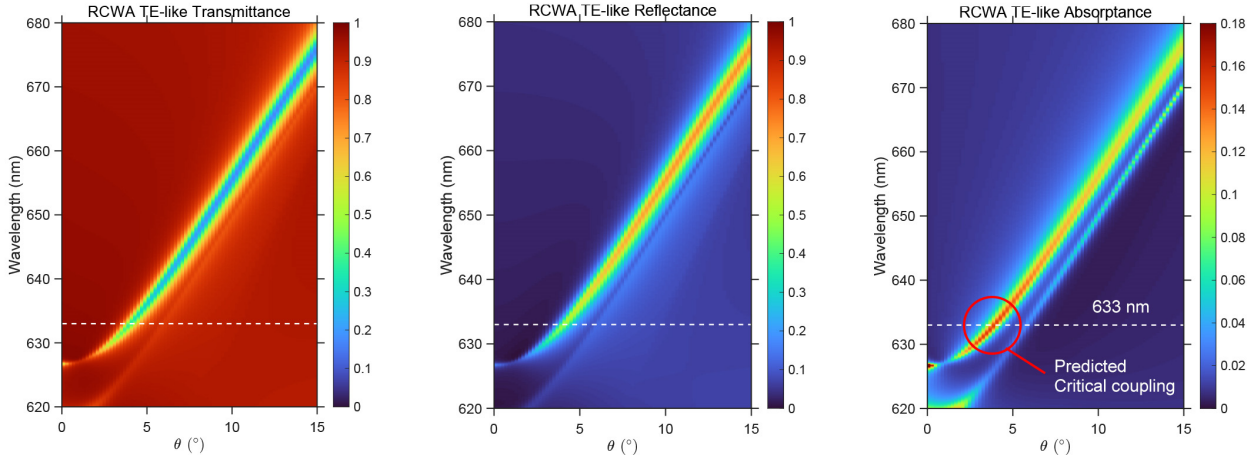
SUPPLEMENTARY INFORMATION *not applicable.*

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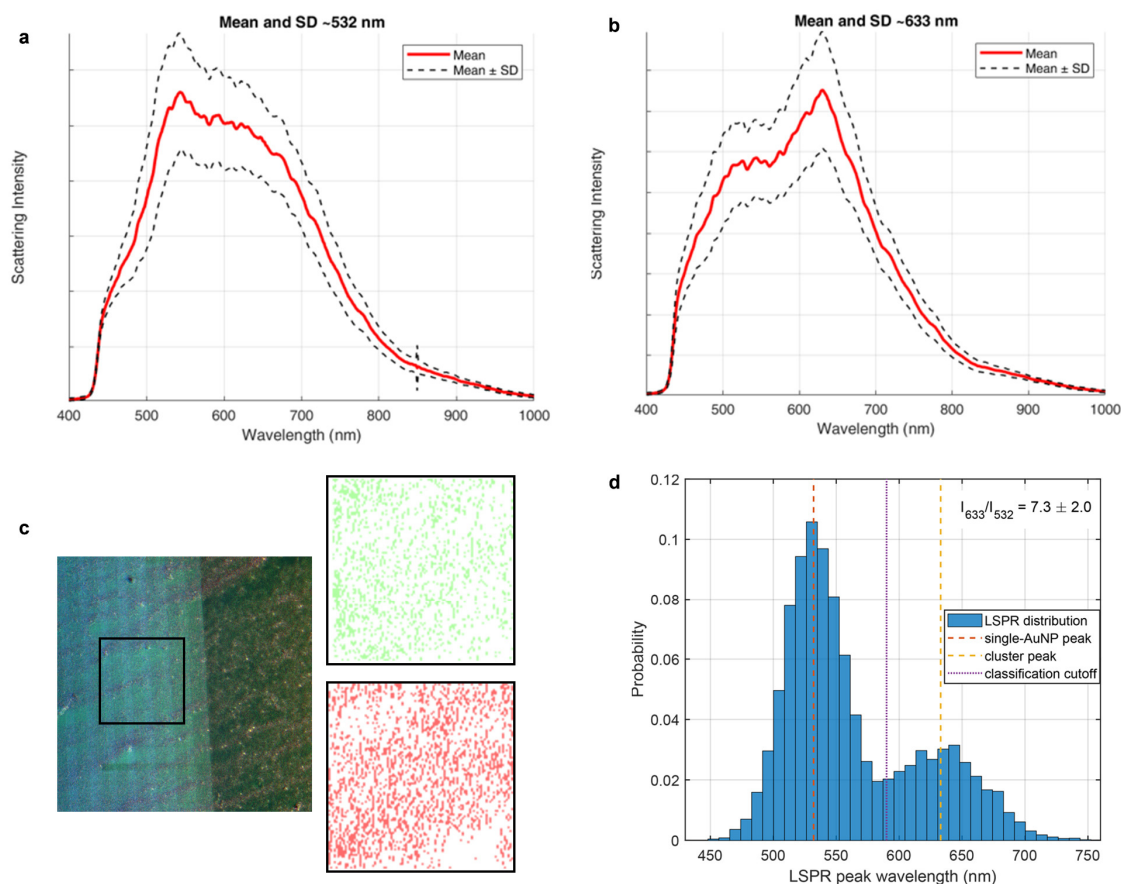
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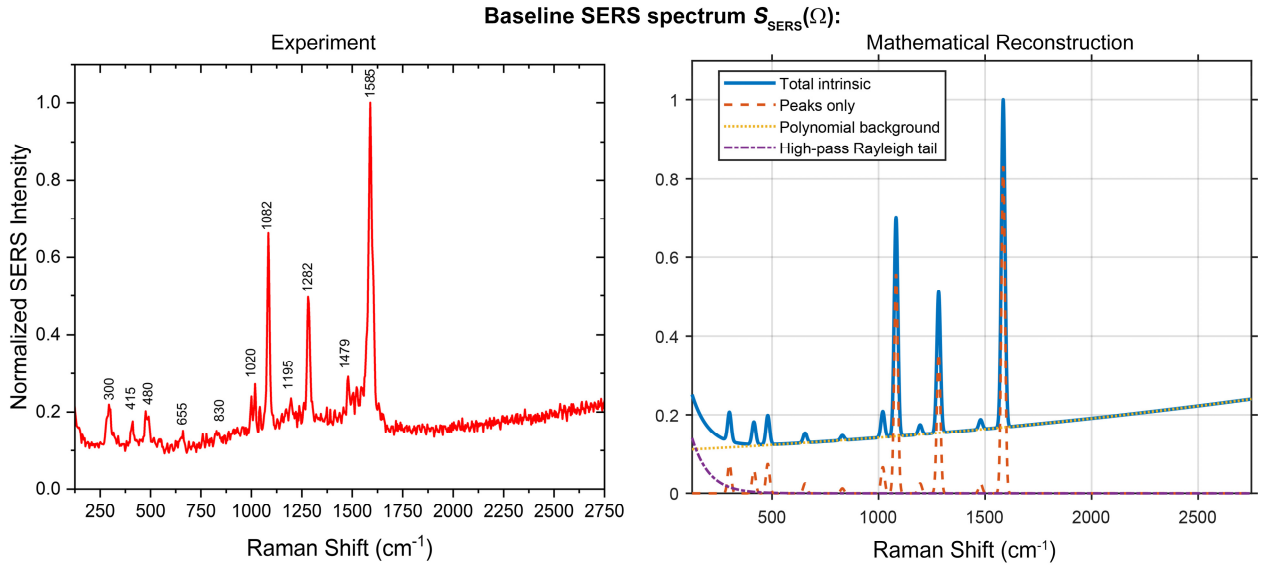
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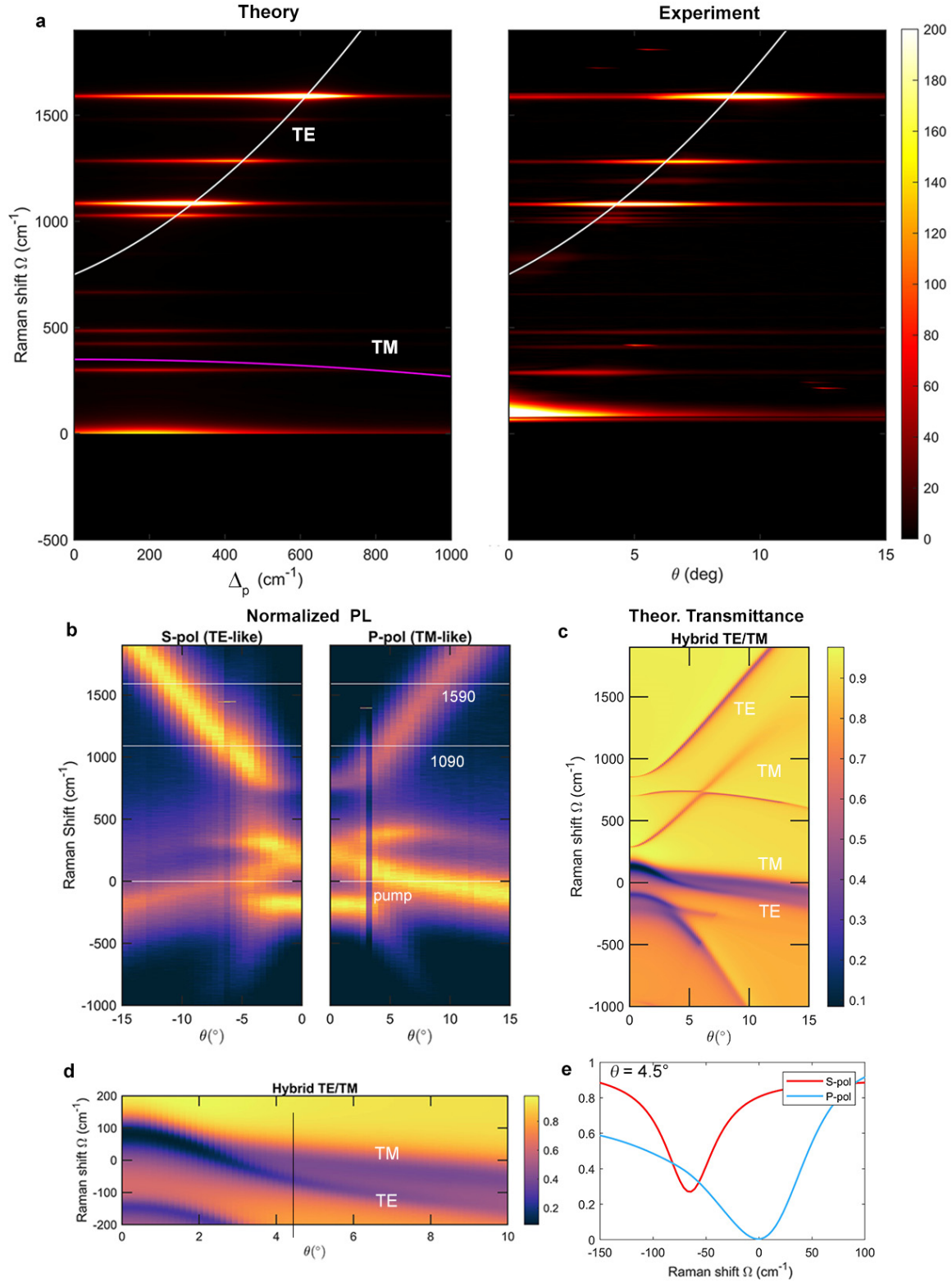
Extended Data Fig. 1 | RCWA prediction of the quasi-BIC pump-coupling condition. Calculated angle-resolved TE-like transmittance, reflectance and absorptance spectra of the PhCS obtained by RCWA, operating in the single branch regime. The dispersive qBIC branch crosses the fixed pump wavelength of 633 nm indicated by the horizontal dashed line. At small but finite incidence angle, symmetry-protected radiative leakage is opened extrinsically, allowing the pump to couple to the qBIC mode. The absorptance map identifies the predicted critical-coupling condition, where the radiative coupling rate approaches the intrinsic loss rate and pump injection into the hybrid plasmonic–photonic system is maximized (maximum absorption = maximum local field). This condition, occurring at approximately 3–4° for the simulated design, defines the angular window used for qBIC-mediated pump gating of the SERS response.



Extended Data Fig. 2 | Dark-field hyperspectral analysis of isolated and clustered AuNPs on the PhCS. a, b, Mean dark-field scattering spectra extracted from hyperspectral image pixels associated with AuNP populations displaying LSPR maxima near 532 nm (**a**) and 633 nm (**b**), respectively. Solid red curves indicate the mean scattering spectrum, while dashed black curves show the mean $\pm$ s.d. The 532-nm-peaked population is assigned mainly to isolated AuNPs, whereas the red-shifted 633-nm-peaked population is attributed predominantly to dimers and oligomeric AuNP clusters. **c,** Representative dark-field optical image of the AuNP-decorated PhCS region used for hyperspectral analysis. The boxed area indicates the region selected for pixel-wise spectral classification. The corresponding classification maps show pixels assigned to the 532-nm-peaked population (green) and to the 633-nm-peaked cluster-dominated population (red). **d,** Distribution of LSPR peak wavelengths extracted from the hyperspectral dataset. Vertical dashed lines mark the characteristic peak positions of isolated AuNPs and clustered AuNPs, while the dotted line indicates the wavelength cutoff used to separate the two spectral populations. The integrated scattering contribution of the 633-nm-peaked population is approximately $I_{633}/I_{532} = 7.3 \pm 2.0$ times larger than that of the 532-nm-peaked population, confirming that clustered AuNPs dominate the optical response at the Raman excitation wavelength.



Extended Data Fig. 3 | SERS spectrum and vibrational assignment of BPT on AuNPs. (Left) Normalized SERS spectrum of BPT adsorbed on 40-nm AuNPs, measured under 633-nm excitation. The labelled peaks correspond to the characteristic vibrational fingerprint of BPT chemisorbed on gold through thiol–gold bonding as detailed in the Methods section. The spectrum provides the reference vibrational bands used for the quasi-BIC-mediated SERS analysis. (Right) Numerically reconstructed SERS spectrum used for the model $S_{\text{SERS}}(\Omega)$ function.



Extended Data Fig. 4 | Momentum-space control of SERS in multibranch PhCS. **a**, Calculated and experimental SERS enhancement maps for the multibranch PhCS, showing the Raman response as a function of Raman shift and momentum detuning/angle. Unlike the simpler single-branch case, this platform exhibits multiple

1105 dispersive photonic branches that cooperatively shape the Raman enhancement landscape across momentum
space. Their combined action governs the redistribution of SERS intensity through selective pump-Stokes
matching, yielding a structured, strongly angle-dependent enhancement pattern. The close correspondence
between theory and experiment confirms the multimode photonic origin of the observed Raman shaping. **b**,
Experimental local photoluminescence maps under *s*-polarized (TE-like) and *p*-polarized (TM-like) excitation,
1110 compared with RCWA-calculated hybrid TE/TM transmittance map, reproducing the main spectral trends
observed experimentally and supporting the interpretation in terms of multiple quasi-BIC-derived photonic
channels in **c**. **d**, Enlarged view of the spectral region of interest, relevant to the experiment indicated in **Fig. 5a**,
highlighting the low-shift window where pump-mode matching dominates the Raman response. **e**, Polarization-
resolved spectral cut at $\theta = 4.5^\circ$ of panel **d**, showing that the pump resonances for *s*- and *p* polarization are offset
1115 by about 50 cm^{-1} . This offset directly demonstrates that the excitation pathway is polarization dependent, in
agreement with experiments shown in **Fig. 5a**.