

Retrieving interlayer transfer pathways in van der Waals heterostructures via photon-assisted tunneling

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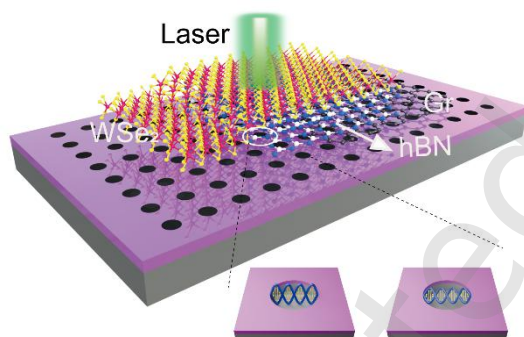
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This work integrates an optical microcavity with two-dimensional heterostructures, leveraging the cavity resonance as a switch to toggle the dominant mechanism from energy transfer to charge transfer. The cavity interacts with dipolar charge-transfer-associated configurations and introduces hybridized intermediate states that reconstruct carrier relaxation pathways, while enhanced local electromagnetic fields promote photon-assisted interfacial transport across the hBN barrier.

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ABSTRACT: Charge and energy transfer are fundamental processes in van der Waals heterostructures (vdWHs), and controlling the transfer pathway is crucial for applications spanning from photodetection to photoluminescence. However, deterministic modulation of energy transfer (ET) and charge transfer (CT) is limited by static band alignments and physical barrier thicknesses. Here, we integrate an optical microcavity with two-dimensional (2D) heterostructures, leveraging the cavity resonance as a switch to convert the dominant mechanism from energy transfer to charge transfer, an unexpected transition that is tunable via laser parameters and cavity geometry. We reveal that the cavity-modified local electromagnetic environment thermodynamically stabilizes the CT pathways via dipole-selective coupling. Furthermore, cavity-mediated exciton-photon hybridization generates intermediate states that reconstruct the non-equilibrium relaxation landscape, while the cavity-enhanced local field induces photon-assisted tunneling to overcome intrinsic barrier limitations. Our work provides unprecedented flexibility for manipulating carrier dynamics in tunable optoelectronics, opening new avenues for ultrafast photodetectors, low-power photovoltaics, tunable switches, and various interdisciplinary applications.

KEYWORDS: van der Waals heterostructures, charge transfer, energy transfer, optical microcavity, photon-assisted tunneling

Introduction

VdWHs are artificial quantum systems constructed by integrating different 2D layers with atomic-level precision [1]. The electronic band structure, exciton characteristics, and many-body effects can be effectively engineered through material selection, stacking order control, and interlayer twist-angle adjustment [2-4]. As an essential process, interfacial carrier dynamics [5], including CT and ET at the van der Waals heterojunction interface, determine the performance of devices across biosensing, energy storage, valleytronics, photocatalysis, and quantum optoelectronics [6-8]. For example, by combining the strong light-matter interaction and spin-valley coupling properties of transition metal dichalcogenides (TMDs) [9] with the high carrier mobility of graphene (Gr) [10], photodetectors based on Gr/TMD heterostructures exhibit high photoresponsivity and photogain [11, 12]. The macroscopic performance of these diverse devices is fundamentally governed by microscopic interfacial charge/energy transfer processes. Whether facilitating rapid carrier extraction in

phototransistors, mediating exciton recombination in light emitters, or maintaining spin/valley polarization during channel injection, the core functionality is dictated by the intricate competition between interlayer CT and ET.

Despite these advantages, the efficiencies of CT and ET remain fundamentally limited by static physical properties in device architectures, such as interlayer distance, energy band alignment, and the localized dielectric environment [13]. Concurrently, complex exciton states induced by quantum confinement and intrinsic doping further limit the uniformity and efficiency of these transfer mechanisms [14]. The integration of optical microcavities presents a powerful strategy to circumvent these static bottlenecks. Building upon the well-established, highly efficient light-matter coupling between TMDs excitons and cavity modes, incorporating a microcavity not only mitigates deleterious substrate effects to boost exciton emission [15, 16] but also enables control of interlayer interactions such as Förster resonance energy transfer (FRET) to facilitate efficient long-range energy funneling [17, 18]. Consequently, integrating 2D materials with optical microcavities establishes a

promising platform for actively engineering macroscopic device performance via the coupling between excitation states and photonic density of states.

Here, we demonstrate a cavity-enabled $\text{WSe}_2/\text{hBN}/\text{Gr}$ heterostructure that realizes a mechanistic transition from energy transfer to charge transfer. This architecture establishes coupling between the excitons in the heterostructure and cavity photon modes, thereby confining the optical field to enhance the local density of optical states (LDOS), as evidenced by angle-resolved reflectance spectroscopy. Fundamentally, the cavity-modified local electromagnetic environment thermodynamically stabilizes the CT pathways via dipole-selective coupling. Concurrently, cavity-mediated exciton-photon hybridization generates intermediate states that reconstruct the non-equilibrium relaxation landscape. According to the Tien-Gordon model, the enhanced electromagnetic field facilitates a photon-

assisted tunneling process, thereby providing a unique control lever to deterministically switch the interfacial dynamics from energy transfer to ultrafast charge transport. The extent of the mode transition can be modulated by adjusting temperature, laser power, and excitation wavelength. At the same time, theoretical calculations demonstrate that optical field control is capable of selectively altering the characteristic charge transfer behavior in $\text{WSe}_2/\text{hBN}/\text{Gr}$ heterostructure through the design of the optical cavity. By decoupling transport pathways from the static constraints of the physical crystal lattice, our approach lays the groundwork for next-generation, dynamically reconfigurable optoelectronic functionalities and device architectures.

Results and discussion

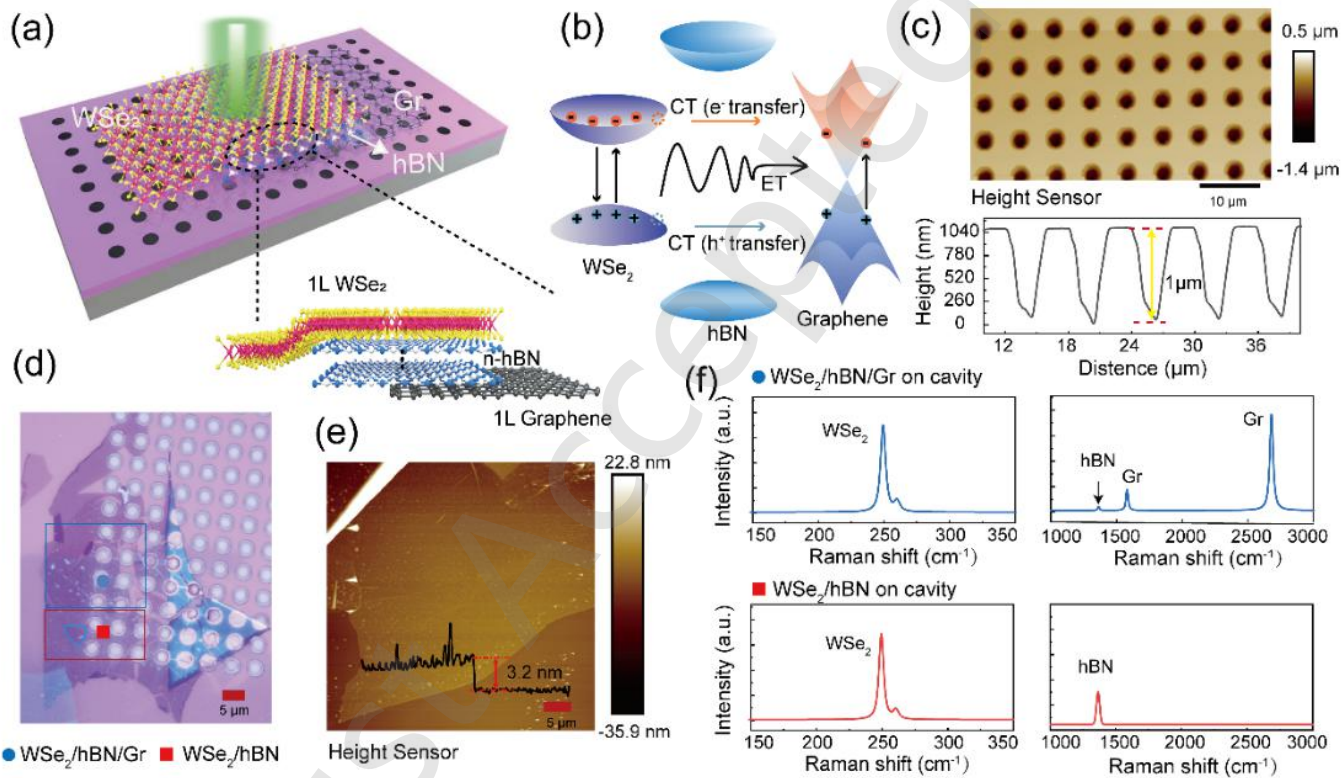


Figure 1. Structure and properties of $\text{WSe}_2/\text{hBN}/\text{Gr}$ heterojunction on cavity. (a) Schematic heterostructure on the SiO_2/Si substrate with a micron-scale holes array. The dashed box shows the atomic layer arrangement order; (b) Schematic illustration of transfer mechanism in band alignments of $\text{WSe}_2/\text{hBN}/\text{Gr}$ heterojunction; (c) AFM height image of the cavity; (d) Micrograph of incommensurately stacked heterostructure on cavity. The scale bar corresponds to 5 μm ; (e) AFM height image of hBN ; (f) Raman spectra of the constituent materials of heterostructure on cavity.

The two-dimensional layers used in this work were obtained by mechanical exfoliation from bulk crystals [19]. Figure 1a shows the cavity system composed of the heterostructures and micron-scale holes. Monolayer monolayer Gr , few-layer hBN , and WSe_2 were exfoliated and sequentially stacked onto a SiO_2/Si substrate with patterned cavities, forming two distinct regions: $\text{WSe}_2/\text{hBN}/\text{Gr}$ and WSe_2/hBN . Figure 1b illustrates the corresponding band alignment and the mechanisms of ET and CT. On the basis of band alignment considerations, ET is generally expected to play a major role in the interfacial dynamics of type-I vdWHs and Gr/TMD systems [20]; however, numerous studies have demonstrated that CT can also contribute significantly to these dynamics [21]. Extensive studies employing transient

spectroscopy have confirmed strong interlayer coupling and ultrafast transfer processes in monolayer TMD/Gr heterostructures [22,23]. All heterostructures investigated in this work employ monolayer Gr , which was selected because its low density of states near the Dirac point and low quantum capacitance make the interfacial carrier transfer highly sensitive to small charge redistribution, enabling Raman spectroscopy to sensitively probe cavity-induced doping [24,25]. Although increasing the Gr layer number can provide additional dissipative channels and modify dielectric screening, thereby influencing the balance between ET and CT, additional layers also alter the acceptor electronic structure and carrier-relaxation pathways [26]. Owing to its low electron affinity and wide bandgap, hBN serves as a

selective tunneling barrier for carriers and excitons [27,28]. While an ultrathin hBN layer facilitates the quantum tunneling of carriers between WSe₂ and graphene, increasing the hBN thickness effectively turns it into a robust insulating barrier, thereby suppressing direct interlayer charge transfer. The thickness of the hBN layer was measured to be approximately 3.2 nm using Atomic Force Microscopy (AFM), as shown in the height profile in Figure 1e. The microcavity array was fabricated using Inductively Coupled Plasma (ICP)

etching, resulting in cavities with a diameter of 3 μm and a depth of 1 μm [16]. An AFM image of the representative microcavity array is shown in Figure 1c. Figure 1d shows the optical microscopy image of the microcavity device. The successful transfer of the materials onto the optical cavity is verified by the Raman spectra (Figure 1f) acquired from the WSe₂/hBN/Gr (blue box) and WSe₂/hBN (red box) regions, where the signature peaks of graphene, WSe₂, and hBN are clearly resolved.

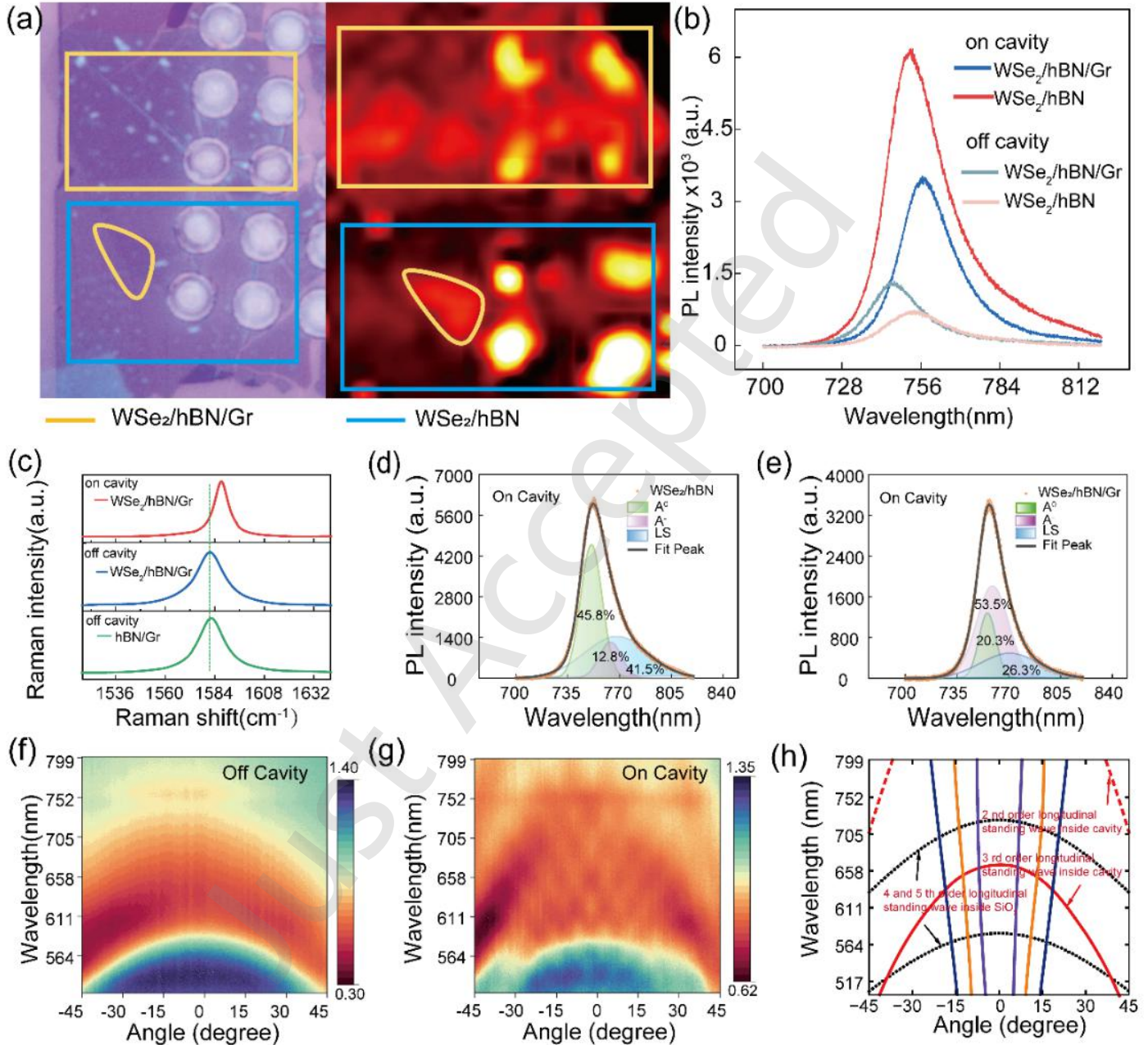


Figure 2. Heterostructure PL and Raman spectroscopy characterizations. (a) Optical microscope image and photoluminescence mapping of heterostructure. Yellow and blue lines indicate WSe₂/hBN/Gr and WSe₂/hBN, respectively; (b) The PL spectra of WSe₂/hBN/Gr and WSe₂/hBN were measured on the cavity and SiO₂/Si substrate respectively; (c) Room-temperature Raman spectra of graphene G-mode as measured in different regions, including at hBN/Gr, and WSe₂/hBN/Gr with and without cavity. (d,e) The total PL peaks of WSe₂/hBN and WSe₂/hBN/Gr on cavity are deconvoluted using Gaussian functions to extract exciton (A), trion (A⁻), and LS emissions; (f,g) Angle-resolved reflectance spectra of WSe₂/hBN/Gr on cavity and off cavity; (h) The theoretical predictions curve of WSe₂/hBN/Gr on cavity.

The optical microscope image and corresponding photoluminescence (PL) map of the microcavity system are depicted in Figure 2a. The device architecture incorporates both WSe₂/hBN/Gr (highlighted by the yellow frame) and WSe₂/hBN (blue frame) regions, enabling in-situ comparison

of their optical properties. Steady-state Raman and PL characterizations were conducted using a 532 nm (2.33 eV) solid-state laser as the excitation source. As revealed in Figure 2b, in the regions without the microcavity (off-cavity), the PL intensity of the WSe₂/hBN/Gr heterostructure is

consistently stronger than that of the WSe_2/hBN reference structure. Theoretical models predict that given the strong optical dipole moments inherent to both TMDs and graphene, FRET governs the interlayer dynamics in their vdW heterostructures, provided that a sufficiently large potential barrier exists to suppress electron wavefunction overlap [29,30]. To inhibit charge transfer pathways, a ~ 3 nm hBN insulating layer was strategically introduced between the WSe_2 and graphene layers. Since charge tunneling decays exponentially with distance while dipole-coupled energy transfer operates over longer ranges, the fact that no PL quenching was observed in the $\text{WSe}_2/\text{hBN}/\text{Gr}$ stack confirms the effective suppression of direct charge transfer [28,31]. In the heterostructure region on the cavity, the pronounced quenching of the $\text{WSe}_2/\text{hBN}/\text{Gr}$ PL indicates that a highly efficient non-radiative decay channel outcompetes the intrinsic radiative recombination. This strong PL quenching can originate from two distinct interfacial processes: CT and ET. In the CT mechanism, the ultrafast physical separation of photoexcited electrons and holes across the interface drastically diminishes their spatial wavefunction overlap, thereby suppressing radiative emission [32-34]. Conversely, in the ET mechanism, the exciton in WSe_2 non-radiatively transfers its energy to graphene via dipole-dipole interactions [35]. Since graphene acts as an efficient energy sink due to its zero bandgap and ultrafast thermalization, the transferred energy is rapidly dissipated as heat.

It is also noteworthy that the PL intensity of all on-cavity regions is enhanced relative to the off-cavity regions, with a maximum enhancement factor of approximately 10 (Figure 2b). This common enhancement establishes that the cavity modifies the local excitation and emission environment of both WSe_2/hBN and $\text{WSe}_2/\text{hBN}/\text{Gr}$ regions; therefore, the absolute PL enhancement itself is not used as evidence for charge transfer. On the SiO_2/Si substrate, the $\text{WSe}_2/\text{hBN}/\text{Gr}$ heterostructure (light blue line) exhibits a blueshift of 7 nm in its PL spectrum relative to the WSe_2/hBN control region (light red line). This blueshift indicates that the emission is dominated by neutral excitons, suggesting that the formation of charged excitons (trions) is suppressed in this heterostructure configuration [36]. In contrast, in the presence of the cavity, the $\text{WSe}_2/\text{hBN}/\text{Gr}$ region (blue line) displays a 4 nm redshift compared to the WSe_2/hBN control (red line). Because a peak shift alone cannot uniquely identify the transfer pathway, the carrier-related interpretation is based on the correlated Raman response of graphene, the exciton-trion spectral weights, and the PL decay dynamics discussed below.

In-situ Raman spectroscopy serves as a powerful, non-destructive probe of the doping level in graphene. We measured the peak positions (ω) and full widths at half maximum (FWHM) of the Raman G and 2D peaks for both the hBN/Gr and $\text{WSe}_2/\text{hBN}/\text{Gr}$ regions, as shown in Figure 2c. The G peak (ω_G) of the hBN/Gr structure on SiO_2/Si is centered at ~ 1582.3 cm^{-1} with a FWHM of 15.5 cm^{-1} . For the $\text{WSe}_2/\text{hBN}/\text{Gr}$ heterostructure in the off-cavity region, no significant shifts or broadening of the G peak were observed. This result is consistent with a scenario in which the insulating hBN layer acts as a tunneling barrier, inhibiting net charge transfer. In this case, photoexcited excitons in the WSe_2 decay non-radiatively through dipole-dipole coupling, leading to energy transfer to the graphene. Conversely, the G peak of graphene within the on-cavity $\text{WSe}_2/\text{hBN}/\text{Gr}$ region exhibits a substantial blueshift of approximately 4.9 cm^{-1} and narrows to a FWHM of 8.3 cm^{-1} . These spectral modifications are well-established signatures of carrier

doping. Coupled with the 2D peak analysis, they signify hole doping in the graphene (Figure S1), a behavior evidently modulated by the cavity [24]. Consequently, we conclude that the interfacial dynamics within the cavity-suspended heterostructure region are governed by a charge transfer mechanism. To eliminate strain as a possible origin for these shifts, we monitored the strain-sensitive E_{2g} mode of WSe_2 . A comparison between on-cavity and off-cavity regions revealed a negligible change in the E_{2g} peak position (Figure S2), thereby confirming that strain effects are not responsible for the observed modifications in the graphene Raman spectrum. To further exclude a cavity-induced Raman modification unrelated to interlayer carrier redistribution, we examined a control sample with a thin hBN spacer, for which appreciable charge transfer occurs both on and off the cavity (Figure S3a). The graphene G mode in this control shows no significant cavity-dependent frequency shift (Figure S3b), consistent with persistent interlayer charge transfer in both regions (Figure S3c).

Figure 2d-e displays the deconvoluted PL spectra of the heterostructure for the samples transferred onto the optical cavity. The individual contributions from the neutral exciton (A^0), the trion (A^-), and localized states (LS) were extracted using Gaussian fitting [37,38]. The A^0 peak originates from the direct bandgap emission at the K point of monolayer WSe_2 , while the A^- peak is associated with the recombination of charged excitons. For the on-cavity $\text{WSe}_2/\text{hBN}/\text{Gr}$ heterostructure, the A^-/A^0 intensity ratio increases relative to its graphene-free counterpart. Based on the principle of charge conservation, this anomalous electron accumulation in WSe_2 correlates with the concurrent in-situ Raman signatures of the graphene layer. As discussed earlier, the pronounced blueshift and linewidth narrowing of the graphene G and 2D modes uniquely verify hole (p-type) doping, which matches with the residual electrons left in the WSe_2 layer [39]. This redistribution between neutral and charged excitons, alongside the emergence and enhancement of trion emission, provides direct evidence of cavity-assisted charge transfer. Suspending the heterostructure on the cavity helps suppress substrate-induced doping that often facilitates defect-assisted non-radiative recombination. Consequently, for graphene-free areas, the weakened substrate screening on the cavity results in a significant reduction of trions relative to the off-cavity case (Figure S4) [16].

Angle-resolved reflectance spectroscopy was employed to evaluate the coupling strength between the microcavity modes and the excitonic resonances in $\text{WSe}_2/\text{hBN}/\text{Gr}$ heterostructure system. Figures 2f-2g display the spectra for the $\text{WSe}_2/\text{hBN}/\text{Gr}$ heterostructure in the absence and presence of the microcavity, respectively. Discernible cavity modes were not observed in the bare aperture spectrum (Figure S5). Without the cavity, a flat excitonic band is observed around 750 nm, exhibiting a single, angle-independent spectral feature characteristic of excitons in TMDs. In contrast, we observe a series of periodic, strongly dispersive optical modes with the microcavity. These modes exhibit strong angular dispersion and are attributed to Fabry-Pérot resonances arising from the heterostructure-cavity integration. Notably, these dispersive modes spatially overlap with and spectrally cross the exciton energy level. The spectra reveal clear crossing points between the photonic and excitonic dispersion relations, indicating that the microcavity introduces a modified photonic density of states (DOS). Based on the theoretical prediction dispersion curves as show in Figure 2h, the longitudinal photonic

branches can be resolved as both the WSe₂/hBN/Gr layer and the Si substrate behave like mirrors so as to support the longitudinal standing waves inside the proposed cylindrical cavity (Figure S6). The calculated transverse photonic branches are shown as purple (1st), orange (2nd), and blue (3rd) solid lines, with the exciton wavelength denoted by the pink shaded region. The clear crossing behaviors around the exciton energy state is observed in the dispersion curve, revealing the coupling, which is consistent with the measurements shown in Figure 2g.

The overlap of dispersive cavity modes and exciton energy level creates “hot spots” in momentum space, leading to enhanced local fields ($|E|^2$) and increased exciton generation rates, thereby maximizing the light-matter interaction. According to the Tien-Gordon model [40], microcavity resonances or plasmonic effects generate a highly amplified localized electromagnetic field that applies a strong periodic alternating current (AC) bias to the tunnel junction, facilitating photon-assisted tunneling [41,42]. Therefore, upon strong excitation by a resonant optical field, the interlayer transport dynamics at interfaces of WSe₂/hBN/Gr heterostructure with an inherent potential barrier are fundamentally altered. While hBN normally presents a static rectangular barrier, the cavity-enhanced local field induces an oscillation in the effective barrier height. This intense field modifies the excitonic wavefunction of WSe₂, which is no longer described by a single energy level E , but instead splits into a series of virtual states separated by the photon energy quanta $\hbar\omega$:

$$E_n = E_0 + n\hbar\omega \quad (n = 1, 2, \dots)$$

where E_n is the energy of the n -th Floquet sideband, E_0 represents the original unperturbed excitonic energy level of WSe₂, and n is the integer index denoting the number of photons exchanged with the cavity field. Although the hBN barrier blocks ground-state charge transport, electrons in these sidebands can gain sufficient energy to overcome the barrier or align with empty states in the graphene layer.

Driven by the resonant local field, charges absorb photon energy to tunnel across the interface [43]. Notably, even without the strong coupling regime (anti-crossing), the periodic cavity modes effectively modulate the photonic density of states at the exciton energy [44,45]. This resonant pumping mechanism induces significant non-equilibrium carrier dynamics.

Furthermore, this transition is bolstered by cavity-modified many-body interactions [46]. Without cavity confinement, the hBN barrier leads to stochastic and uncorrelated electron evolution in the adjacent layers. Coupling the system to a common vacuum electromagnetic field, however, enables cavity-photon exchange to phase-lock the donor and acceptor states. This cavity-mediated long-range interaction hybridizes isolated states into photon-dressed coherent many-body states, establishing long-range electron-electron correlations that drastically enhance interlayer electronic coupling. In contrast to neutral excitons, CT states possess a substantial electric dipole moment μ , the magnitude of which scales with the interlayer separation. Within the framework of cavity quantum electrodynamics (QED), the system-cavity coupling strength H_{int} is proportional to the dipole moment $H_{int} \propto -\mu E_{cav}$, where E_{cav} denotes the local electric field of the resonant cavity mode. Consequently, the coupling of CT states to the cavity field is significantly stronger than that of neutral excitons [47]. Moreover, recent exciton-based nonadiabatic molecular dynamics studies combining LR-TDDFT with the Pauli-Fierz cavity-QED framework have shown that cavity-induced exciton-photon hybridization can generate polaritonic intermediate states with interlayer character in TMD heterostructures [48]. These cavity-modified states reconstruct the excitonic relaxation landscape by introducing additional transfer pathways and reducing the constraints imposed by energy and momentum mismatches, thereby facilitating energy dissipation and interfacial charge-transfer processes [46].

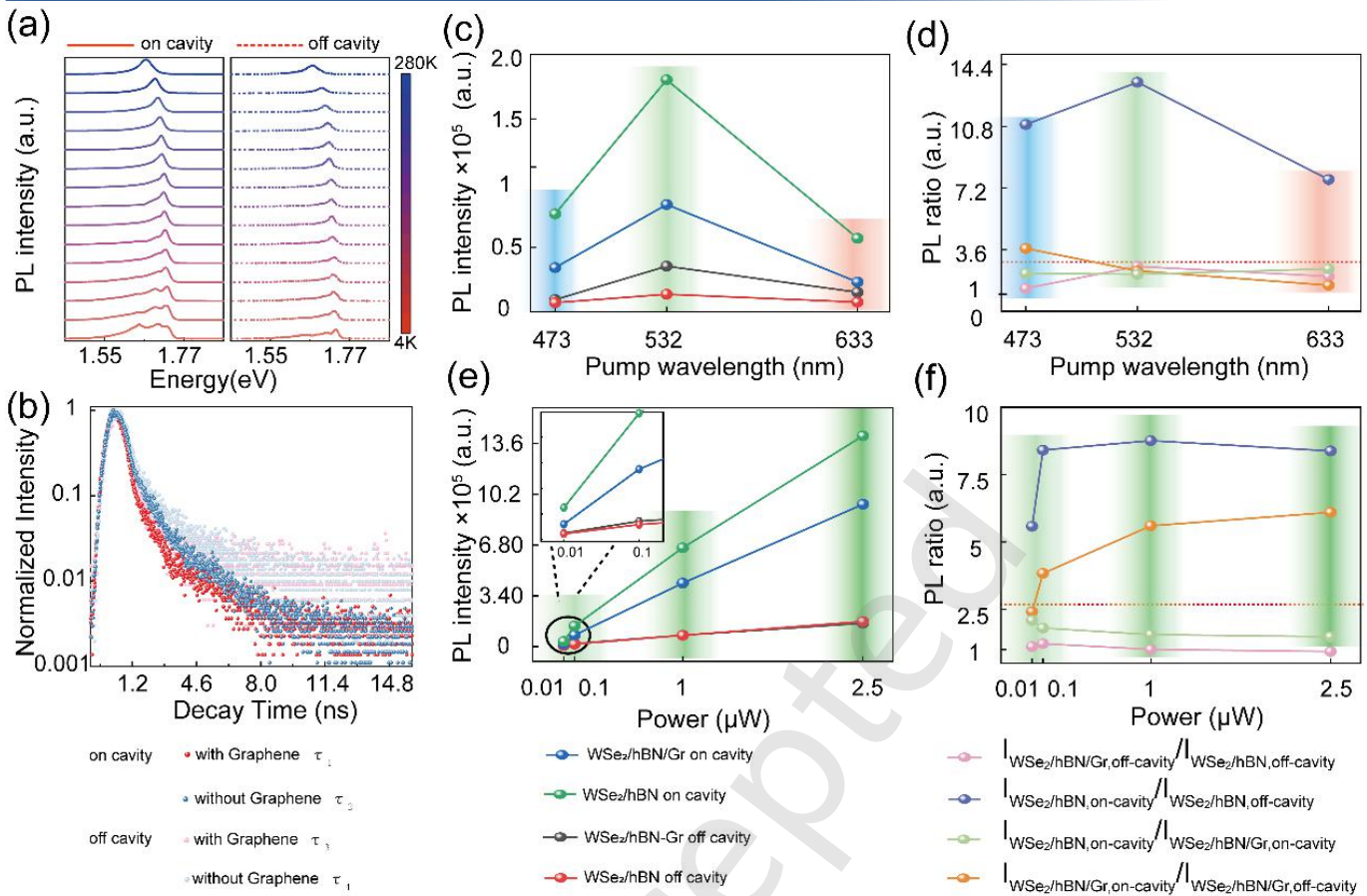


Figure 3. PL evolution and dynamics. (a) Temperature-dependent photoluminescence spectra of WSe₂/hBN/Gr measured on the cavity and off cavity regions; (b) Normalized time-resolved PL spectra of WSe₂/hBN/Gr and WSe₂/hBN were measured on the cavity and SiO₂/Si substrate respectively; (c) Integrated PL intensity from different regions under excitation at 473, 532, and 633 nm; (d) Integrated PL peak intensity ratio between distinct regions as a function of excitation wavelength (473 nm, 532 nm, and 633 nm); (e) Power-dependent integrated PL intensity from the same regions under 532 nm laser excitation; (f) Excitation power-dependent integrated PL peak intensity ratio for the corresponding regions, measured under 532 nm excitation.

Temperature-dependent PL spectroscopy was employed to probe the underlying carrier transfer dynamics. Figure 3a compares the temperature-dependent PL evolution from the WSe₂/hBN/Gr heterostructure with and without the cavity. At temperatures below 80 K, the spectra are dominated by exciton and trion emission. However, trion emission is thermally suppressed with increasing temperature from 80 to 280 K, leading to PL dominated by neutral excitons. The overall PL intensity decreases monotonically with rising temperature. This behavior is attributed to the thermal activation of localized excitons into delocalized states, where they are subsequently captured by competing non-radiative channels or recombine as free excitons [49]. Figure 3b presents the time-resolved photoluminescence (TRPL) measurements obtained from four distinct regions, both on- and off-cavity. The decay curves were fitted using a bi-exponential function ($\tau = \frac{A_a\tau_a^2 + A_b\tau_b^2}{A_a\tau_a + A_b\tau_b}$), yielding average lifetimes of $\tau_1 = 537$ ps, $\tau_2 = 705$ ps, $\tau_3 = 692$ ps, and $\tau_4 = 750$ ps. The reduction of τ_2 relative to τ_4 indicates an increased DOS induced by the cavity, which accelerates the intrinsic radiative decay rate. In the absence of the cavity, the lifetime is quenched attributed to energy transfer from WSe₂ to the graphene layer, consistent with dipole-dipole interactions across the barrier. Notably, the on-cavity heterostructure region exhibits the shortest lifetime (τ_1), signaling the

presence of a highly efficient decay channel. According to the principle of charge conservation, the vertical separation of photocarriers injects holes into graphene, manifested by the pronounced Raman G-mode upshift and 2D-mode evolution, while leaving residual electrons in the WSe₂ layer. This localized electron accumulation dynamically shifts the exciton-trion equilibrium, directly leading to the trion-dominated spectral weights observed in the low-temperature PL spectra. The opening of this rapid cavity-assisted charge transfer pathway efficiently depletes the exciton population, thereby giving rise to the minimum lifetime of 537 ps. This observation supports the hypothesis of cavity-enhanced charge transfer, where the photon-assisted tunneling rate exceeds the energy transfer rate, effectively depleting the exciton population.

Figure 3c-f illustrate the modulation of the PL emission and the transfer mechanisms of the heterostructure achieved by varying the laser wavelength and excitation power. The measurements were conducted on WSe₂/hBN/Gr heterostructures and WSe₂/hBN control regions, comparing samples fabricated on the optical cavity with those on a bare SiO₂/Si substrate. To quantitatively characterize the cavity-induced photonic enhancement effect and the competitive evolution between the interfacial ET and CT pathways, we define the PL ratio as the quotient of the integrated PL intensities from two designated regions under identical

optical collection conditions and integration times. Each data point represents the average integrated intensity measured from four distinct locations of the same stacking configuration. The purple and orange curves represent the cavity enhancement factors ($I_{\text{WSe}_2/\text{hBN, on-cavity}}/I_{\text{WSe}_2/\text{hBN, off-cavity}}$, $I_{\text{WSe}_2/\text{hBN/Gr, on-cavity}}/I_{\text{WSe}_2/\text{hBN/Gr, off-cavity}}$) for the WSe₂/hBN and WSe₂/hBN/Gr regions, respectively. The green curve is defined as $I_{\text{WSe}_2/\text{hBN, on-cavity}} / I_{\text{WSe}_2/\text{hBN/Gr, on-cavity}}$, which compares the relative PL contrast between the reference region and the heterostructure under cavity coupling; whereas the pink curve is defined as $I_{\text{WSe}_2/\text{hBN/Gr, off-cavity}} / I_{\text{WSe}_2/\text{hBN, off-cavity}}$, which evaluates the relative PL contrast between the heterostructure and the reference region under off-cavity conditions. Consequently, a PL ratio = 1 signifies that the two designated regions for a given curve exhibit identical integrated PL intensities. Under otherwise identical conditions, three excitation wavelengths—473 nm, 532 nm, and 633 nm were employed. First, we examined the effect of excitation wavelength under identical conditions (Fig. 3c). The absolute PL intensity from all regions was maximized under 532 nm excitation, exhibiting a significantly stronger signal compared to both 473 nm and 633 nm excitation. As depicted in Figure 3e, the integrated PL intensity from all regions increased monotonically with rising excitation power, although the rate of increase varies. Notably, the on-cavity regions show a significantly stronger power dependence than their off-cavity counterparts. The coupling between the optical cavity and the excitation laser determines the local electric field distribution and intensity. This modified field environment, in turn, modulates the exciton dynamics, either facilitating or impeding their transport and recombination pathways. Consequently, the PL from the heterostructure exhibits a distinct dependence on both the excitation wavelength and the laser power.

PL ratios are used to quantify the cavity enhancement effect and to distinguish the competing energy and charge transfer mechanisms. Figure 3(d) reveals that the cavity enhancement is wavelength-dependent (orange and purple lines), achieving a maximum PL enhancement factor of approximately 14 (purple line). The transfer mechanism is further assessed: in the on-cavity region (green line), the ratio of PL intensity between WSe₂/hBN and WSe₂/hBN/Gr exceeds unity, indicating that the PL of the heterostructure is quenched, which is characteristic of charge transfer. Conversely, in the off-cavity region, the ratio of WSe₂/hBN/Gr to WSe₂/hBN is greater than 1, signifying PL enhancement, consistent with non-radiative energy transfer.

The consistent on-cavity quenching and off-cavity enhancement observed across all investigated wavelengths indicate that the dominant transfer pathway is largely independent of excitation energy within this spectral range.

Furthermore, the nonlinear modulation of the aforementioned interfacial transfer mechanisms was examined by varying the excitation power. The power-dependent behavior of these ratios is detailed in Figure 3f. The PL emission ratio with and without the cavity (purple and orange lines) increases with laser power and eventually saturates, implying that the cavity enhancement effect can be tuned by power but has an upper limit. For the off-cavity samples, the PL intensity ratio between the WSe₂/hBN/Gr and WSe₂/hBN regions initially increases with excitation power and then decreases to below unity, as shown in Figure 3f. The heterostructure exhibits enhanced PL at low excitation power, which progressively weakens and reverses into quenching compared with the WSe₂/hBN reference. In the low-excitation-power regime, the gradual increase in laser intensity leads to a higher population of bound excitons. This substantial exciton density inherently enhances the collective dipole-dipole interactions, which in turn strongly stimulates and governs the non-radiative ET mechanism [50]. Under high-power excitation, emission in both regions approaches saturation. This nonlinear emission behavior is primarily governed by the onset of strong many-body interactions at elevated carrier densities [51]. The hBN layer isolates WSe₂ from graphene, causing the heterostructure to behave similarly to an isolated TMD at high exciton densities. This high density facilitates exciton-exciton annihilation (EEA) within the WSe₂ layer, leading to PL quenching [51,52]. Conversely, in the presence of the optical cavity, the PL intensity ratio of the WSe₂/hBN reference to the WSe₂/hBN/Gr heterostructure is greater than 1 (green line), indicating that the heterostructure emission is quenched. However, increasing excitation power reduces this ratio, consistent with a progressive suppression of charge transfer. Under high-power conditions, a higher density of electrons and holes is generated in the conduction and valence bands of WSe₂, leading to a rapid increase in the charge density injected into the Gr. The low density of states near the Dirac point of Gr allows ultrafast charge transfer to rapidly fill the available states around the Fermi level [53]. The ensuing state filling and Coulomb interactions suppress further charge transfer from WSe₂, thereby attenuating PL quenching.

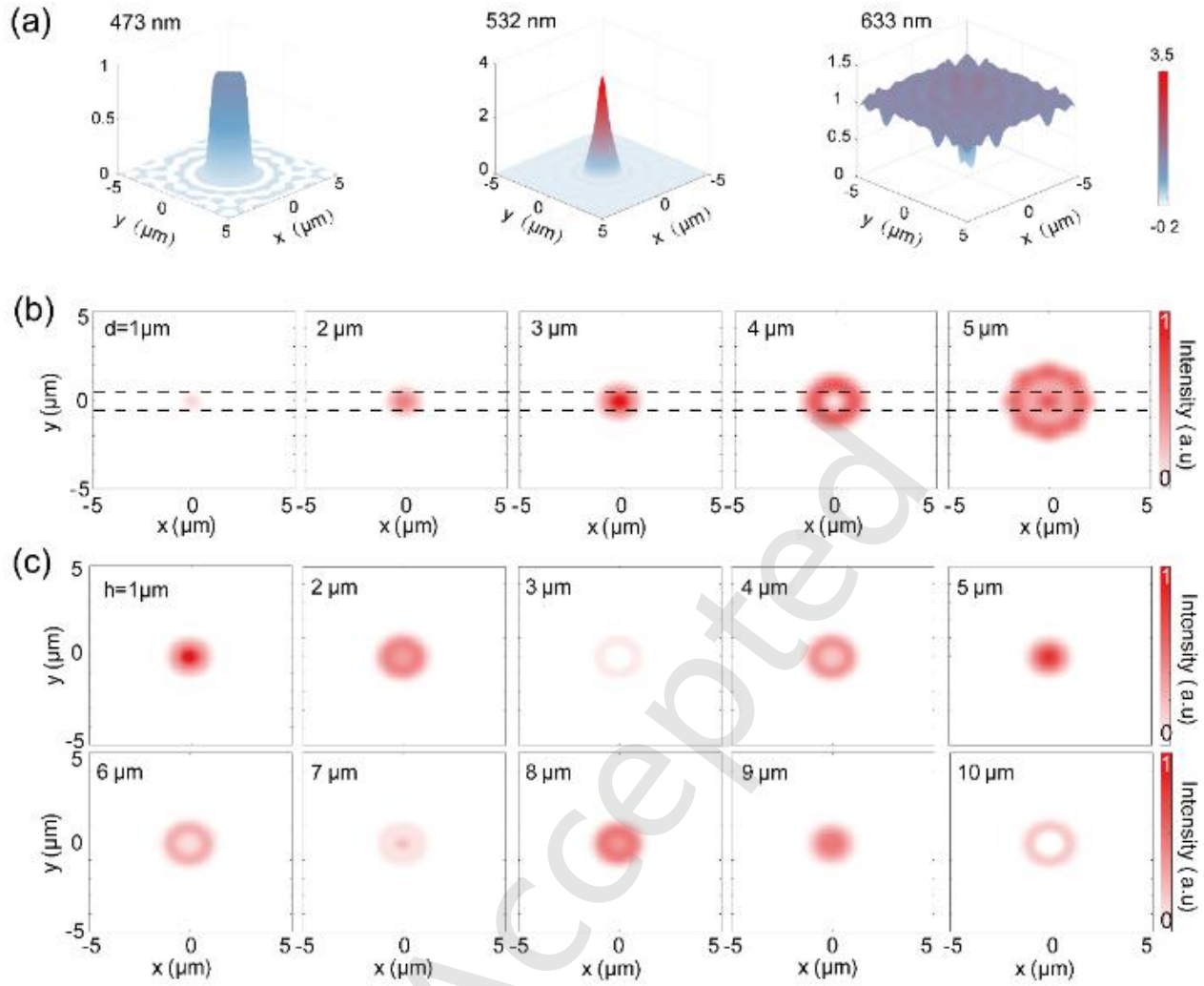


Figure 4. Optical field intensity distribution of heterostructure on cavity. (a) Local field intensity calculated for excitation wavelengths of 473nm, 532nm, and 633 nm; (b), (c) Calculated optical field intensity as a function of the microcavity aperture diameter (b) and depth (c).

The optical field profiles of the heterostructure were calculated using the finite-difference time-domain (FDTD) method. In the simulation model, cylindrical air holes with tunable depth h and diameter d were defined within a SiO_2/Si platform. The air-filled cavity forms a hybrid optical resonator. The interplay between the cavity geometry and the incident light gives rise to rich wavelength- and geometry-dependent field distributions. Simulated electric profiles at visible wavelengths of 473 nm, 532 nm, and 633 nm reveal pronounced spectral dependence of the localized field distribution in Figure 4a. At 473 nm, the field exhibits a ring-shaped intensity profile. At 532 nm, the structure supports a Gaussian-like focal spot. In contrast, at 633 nm, the resonance condition is no longer satisfied, leading to a disordered interference pattern with reduced field localization. The dependence of the optical field on the hole diameter d is shown in Figure 4b. As d increases from 1 μm to 5 μm , the cavity supports additional radial and azimuthal eigenmodes. Consequently, the central intensity maximum gradually broadens and the overall field distribution expands outward, forming a transition from a tightly confined Gaussian spot to a hollow ring-like pattern. This indicates that the lateral confinement weakens with increasing d , allowing higher-order modes with larger effective mode

volumes to be excited, consistent with reduced spatial confinement. In our simulations, we found that the 532 nm laser excitation yields the most highly concentrated optical field and the optimal enhancement effect. Furthermore, the optical field distribution varies distinctly under different excitation wavelengths. This simulated trend is in agreement with our experimental observations, where the wavelength-dependent cavity enhancements were quantitatively evaluated and compared using the PL ratio (as shown in Figure 3c,3d).

The variation with hole depth h demonstrates the axial resonance behavior of the microcavity (Figure 4c). Increasing h modifies the optical round-trip phase accumulated within the cavity, thereby tuning the resonance order and coupling efficiency. When the depth h and the effective refractive index n_{eff} satisfy the cavity resonance condition:

$$2n_{\text{eff}}h \approx m\lambda, m \in \mathbb{Z}^+$$

constructive interference enhances the standing-wave field inside the cavity. As h increases, successive integer orders m is satisfied, producing a periodic modulation of the localized intensity—manifested as the “strong-weak-strong” alternation observed across the subplots. For certain depths, the cavity supports hybridized modes exhibiting both axial

and radial nodes, giving rise to double-ring or annular field distributions. Theoretical calculations suggest that tuning the localized optical field via cavity design offers a potential method to modulate the charge transfer dynamics in WSe₂/hBN/Gr heterostructures. By engineering the photonic density of states, the optical cavity presents a mechanism to influence the fundamental competition between CT and ET, effectively bypassing the bottlenecks imposed by static band alignments.

Conclusions

In this work, an optical microcavity was constructed by integrating a WSe₂/hBN/Gr heterostructure with micro-scale voids etched into a SiO₂/Si substrate. Beyond enhancing optical absorption and photoluminescence, the cavity served as an external handle to actively regulate interfacial carrier dynamics through modifications of the photonic density of states and local electric field distribution. Angle-resolved reflectance spectroscopy revealed clear mode-exciton crossings, consistent with a Tien-Gordon photon-assisted tunneling scenario in which enhanced local fields and increased photonic density of states enable carriers to overcome intrinsic energy barriers and transition from energy transfer to charge transfer-dominated dynamics. Cavity-QED interactions and nonadiabatic dynamics studies suggest that exciton-photon hybridization can reconstruct nonequilibrium relaxation landscapes by introducing additional intermediate pathways with interlayer character. In addition, the large dipole moment of CT-associated configurations may enable stronger coupling to the cavity field, making charge-transfer pathways more competitive, while enhanced local electromagnetic fields may further facilitate photon-assisted interfacial transport. Moreover, the extent of this transition can be tuned through temperature, excitation wavelength, laser power, and cavity geometry. Our results offer a robust strategy for optically controlled, programmable optoelectronic devices, underscoring the integration of optical microcavities with vdWs heterostructures as a promising avenue for realizing light-manipulated quantum transport and next-generation cavity-driven electronics.

Experimental Section

Device Fabrication: Arrays of micro-scale voids were fabricated on a SiO₂/Si substrate using ICP etching. Two-dimensional material flakes were mechanically exfoliated from bulk crystals (HQ Graphene) using the adhesive tape method. Subsequently, the flakes were transferred onto the pre-patterned voids via a dry transfer technique to form the suspended microcavity structures. Finally, the device was annealed in a high-vacuum chamber (1.2×10^{-5} mbar) at 200 °C for 4 hours to remove residues and improve interfacial contact.

Optical Characterization: Steady-state PL and Raman spectra were collected using a confocal Raman spectrometer (Horiba Evolution 800) equipped with continuous-wave (CW) lasers. For Raman measurements, a 532 nm excitation laser was employed with an 1800 lines mm⁻¹ grating to ensure high spectral resolution. For PL measurements, excitation wavelengths of 473 nm, 532 nm, and 633 nm were used in conjunction with a 600 lines mm⁻¹ grating.

Optical Field Simulation: The simulated structure consists of cylindrical air apertures with tunable diameter d and depth h etched into a SiO₂ on Si platform, with the WSe₂, hBN, and graphene layers included using their measured

thicknesses. Three field monitors were placed at the top, middle, and bottom of the stack (spanning the WSe₂/graphene interface, the hBN layer, and the substrate region respectively) to record the full set of electric and magnetic field components ($E_x, E_y, E_z, H_x, H_y, H_z$) used to extract the local field intensity distributions shown in Figure 4a to 4c. The source configuration consisted of a broadband plane-wave excitation, with simulations run separately at 473, 532, and 633 nm to match the experimental excitation wavelengths, with incidence direction and polarization set consistently with the optical measurements. The optical response was calculated using a rigorous coupled-wave analysis formalism, which inherently imposes periodic boundary conditions in the lateral directions and is therefore well suited to modeling the periodic aperture array geometry under consideration. Convergence was verified by progressively increasing the retained diffraction-order truncation until the extracted field intensity profiles remained stable, with finer sampling applied locally around the aperture and layer interfaces to resolve the sharp index contrast.

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Data availability

All data needed to support the conclusions in the paper are presented in the manuscript and/or the Electronic Supplementary Material. Additional data related to this paper may be requested from the corresponding author upon request.

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Declaration of competing interest

All the contributing authors report no conflict of interests in this work.

Author contribution statement

Ting Wang: Conceptualization (lead); Methodology; Data curation (lead); Writing-review & editing (lead). Tianxiang Zhao: Investigation, Software, Validation, Data curation. Zhipeng Qi: Investigation, Software, Writing; Xiao Liu, Haonan Zhao & Jinchun Li: Device Fabrication, Software. Junyong Wang: Writing-review & editing (supporting). Xiaoran Wang & Hao Zhou: Conceptualization (supporting); Data curation (supporting); Hongliang Li: Software, Supervision, Writing & editing. Kai Zhang: Project administration (lead); Supervision (lead). Xuechao Yu: Project administration (lead); Supervision (lead); Conceptualization (lead); Writing-review & editing (lead). All the authors have approved the final manuscript.

Use of AI statement

None.

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Electronic Supplementary Material

Retrieving interlayer transfer pathways in van der Waals heterostructures via photon-assisted tunneling

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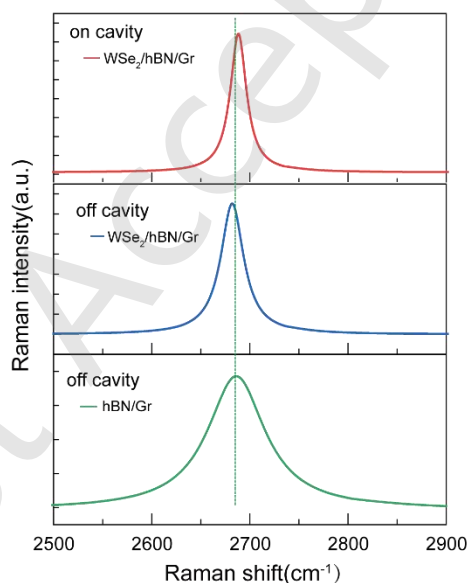


Figure S1 Room-temperature Raman spectra of graphene 2D-mode as measured in different regions, including at h-BN/Gr, and WSe₂/hBN/Gr with and without cavity.

Probing the heterostructure region, we observe that the graphene 2D peak over the cavity is blueshifted compared to the hBN/graphene domain, serving as a clear indicator of hole doping.^[1]

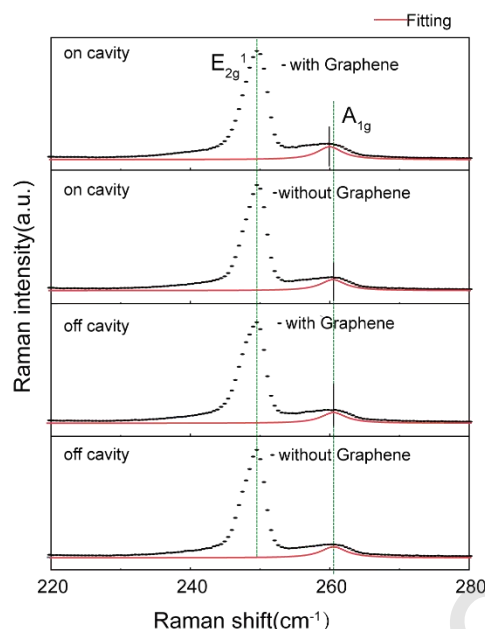


Figure S2 Room-temperature Raman spectra of WSe₂ E_{2g}¹ and A_{1g} as measured in different regions, including at hBN/Gr, and WSe₂/hBN/Gr with and without cavity.

To evaluate the strain and doping conditions of the material over the cavity, we analyzed the Raman peak positions of WSe₂ across different regions. The in-plane E_{2g}¹ mode, which is highly responsive to in-plane strain,^[2] exhibited no discernible shift whether on or off the cavity, indicating the absence of induced strain. Conversely, the out-of-plane A_{1g} mode is exceptionally sensitive to variations in the local charge environment. By deconvoluting this mode using Lorentzian fitting, we observed that on the Si/SiO₂ substrate, the A_{1g} mode of the WSe₂/hBN/Gr heterostructure showed no shift relative to the hBN/Gr region. However, when suspended on the cavity, the A_{1g} mode of the heterostructure underwent a pronounced redshift,^[3] unambiguously demonstrating additional electron injection into the WSe₂ layer.

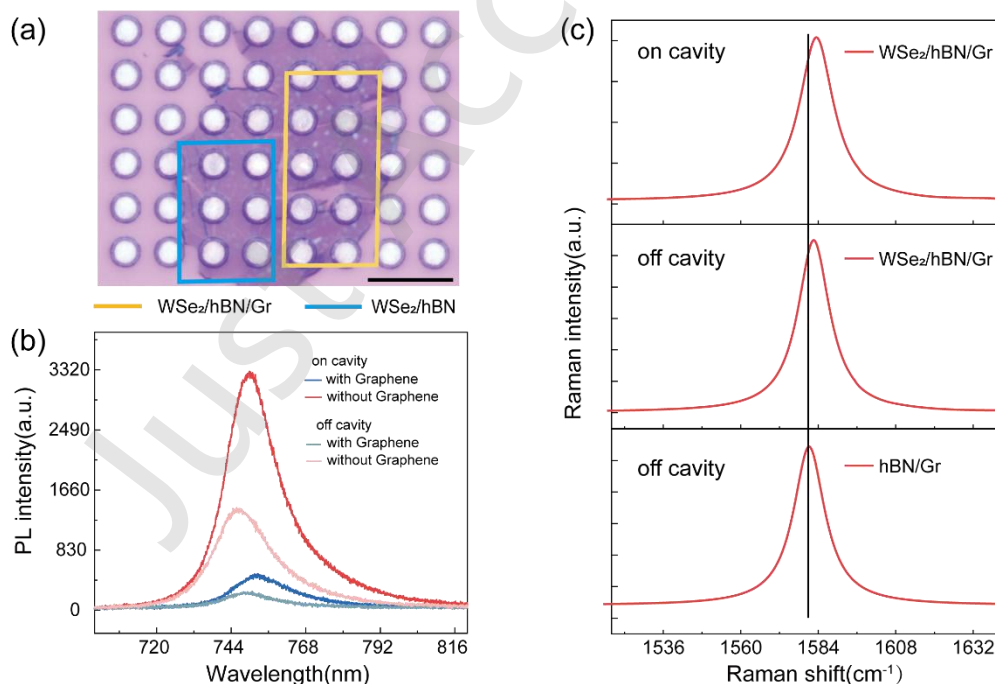


Figure S3 Heterostructure PL and Raman spectroscopy characterizations. (a) Optical microscope image of heterostructure. Yellow and blue lines indicate WSe₂/hBN/Gr and WSe₂/hBN, respectively; (b) Photoluminescence mapping of heterostructure. Yellow and blue lines indicate WSe₂/hBN/Gr and WSe₂/hBN, respectively; (c) Room-temperature Raman spectra of graphene G-mode as measured in different regions, including at h-BN/Gr, and WSe₂/hBN/Gr with and without cavity.

We further probed the system by intercalating a ~1 nm hBN spacer between the WSe₂ and graphene layers, followed by PL and Raman spectroscopic characterizations. The PL emission from the WSe₂/hBN/graphene region is severely quenched compared to the WSe₂/hBN domain, a phenomenon observed consistently across both the cavity-suspended and SiO₂/Si-supported areas. Furthermore, Raman measurements of the graphene G band reveal a discernible spectral shift in both the cavity and substrate regions compared to pristine hBN/graphene. This systematic shift, serving as a direct signature of carrier

doping, leads us to conclude that a charge transfer mechanism governs the interactions across the entire sample footprint.

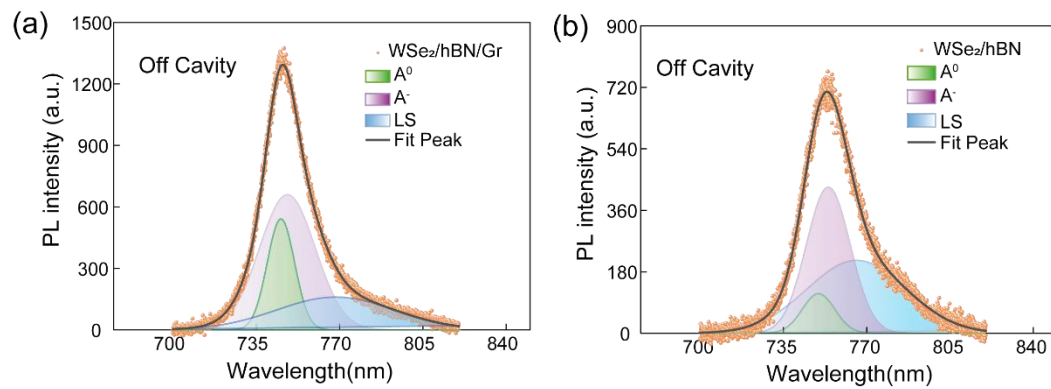


Figure S4 The total PL peaks of WSe₂/hBN/Gr and WSe₂/hBN on SiO₂/Si are deconvoluted using Gaussian functions to extract exciton (A), trion (A⁻), and LS emissions.

Gaussian deconvolution of the off-cavity PL spectra reveals that, without the optical cavity, the hBN spacer strongly suppresses interlayer exciton emission, rendering interlayer charge transfer negligible. This spatial isolation renders interlayer charge transfer negligible, thereby drastically reducing the exciton-to-trion conversion probability and yielding a significantly higher exciton-to-trion ratio in the WSe₂/hBN/graphene region compared to the graphene-free configuration.

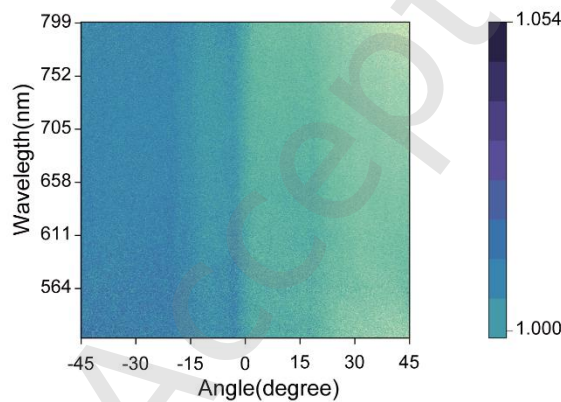


Figure S5 Angle-resolved reflectance spectra of bare cavity.

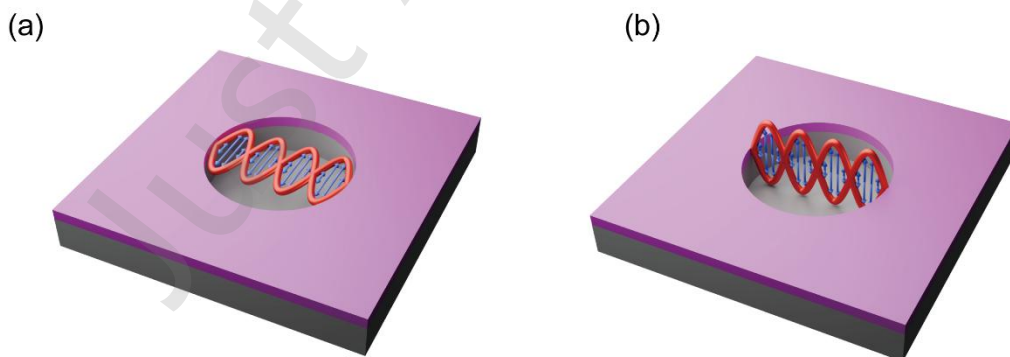


Figure S6 Transverse and longitudinal standing waves within the microcavity aperture.

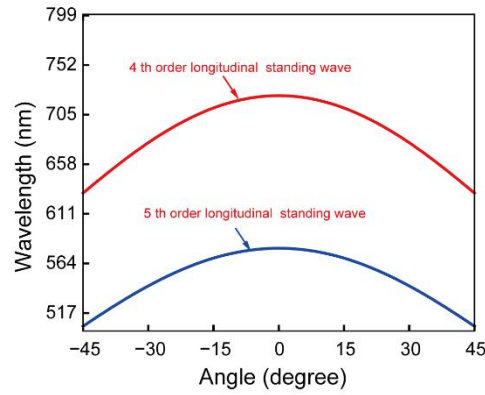


Figure S7 The theoretical predictions curve of WSe₂/hBN/Gr on SiO₂/Si.

To evaluate the coupling in the arrayed air holes with WSe₂/hBN/Gr layer on top of them, the corresponding photonic as well as exciton energy states were numerically calculated, as shown in Figure S6. Since these air holes are located far away from each other, they can be regarded as the individual cylindrical cavities, enabling the light-matter interactions by supplying strongly localized optical modes that follow the standing wave equation:

$$\omega^2 = \left(\frac{c}{n_{\text{eff}}} \right)^2 \left[\left(\frac{p\pi}{L} \right)^2 + \left(\frac{m\pi}{a} \right)^2 \right], (m, p = 0, 1, 2, 3, \dots)$$

where n_{eff} denotes the effective refractive indices of confined optical modes. $k_{\parallel} = p\pi/L$ symbolizes the longitudinal standing wave that is perpendicular to the chip, while $k_{\perp} = m\pi/a$ symbolizes the transverse standing wave that is parallel to the chip. And ω is the angular frequency of the confined optical mode.

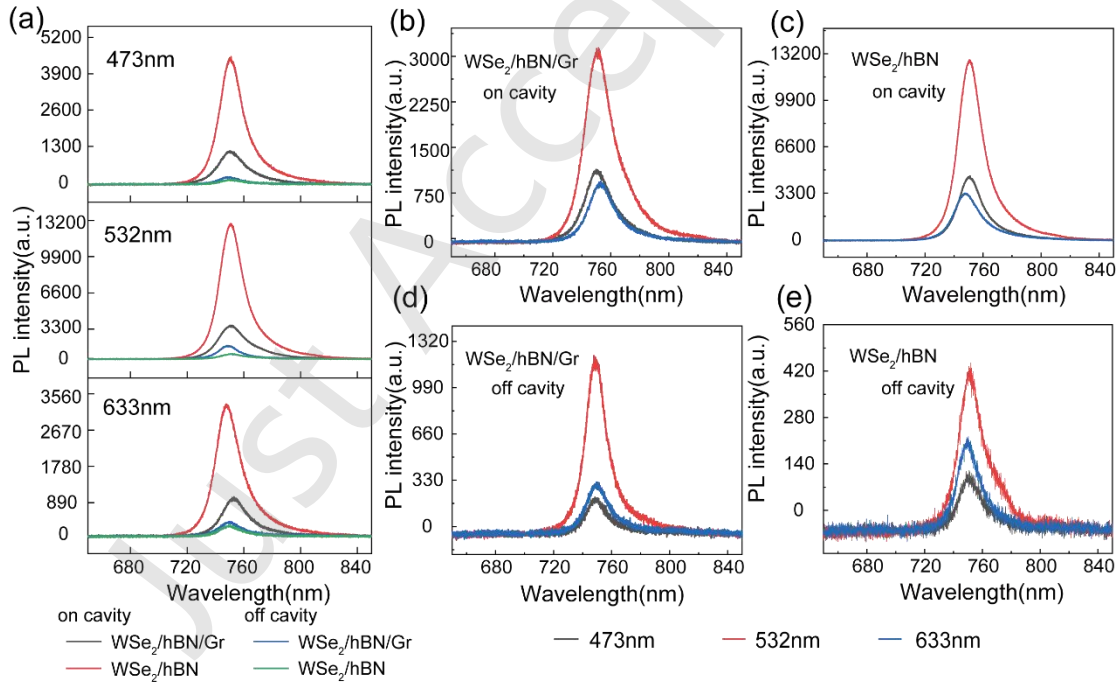


Figure S8 (a) Integrated PL intensity from different regions under excitation at 473, 532, and 633 nm; (b)-(e) Integrated PL intensity from the same regions under excitation at 473, 532, and 633 nm.

Optical excitation with varying laser wavelengths was employed to explore the modulation of the transfer mechanisms. As shown in Figure S8(a), the relative PL intensity ordering among different regions remains perfectly consistent, despite a 2- to 5-fold variation in absolute intensity upon changing the excitation source. This robust trend implies that varying the wavelength solely adjusts the initial carrier injection density without transitioning the underlying recombination dynamics. For a given spatial region, as detailed in Figures S8(b)–(e), the 532 nm excitation consistently produces stronger emission than both the 473 nm and 633 nm lasers.^[4] Distinctively, we observe that over the microcavity, the emission intensity excited at 473 nm exceeds that at 633 nm, directly contradicting the trend measured in the off-cavity regions. This spatial discrepancy clearly demonstrates that reshaping the local density of optical states can selectively enhance the absorption cross-section at specific excitation wavelengths, offering direct proof of cavity-modulated optical pumping.^[5]

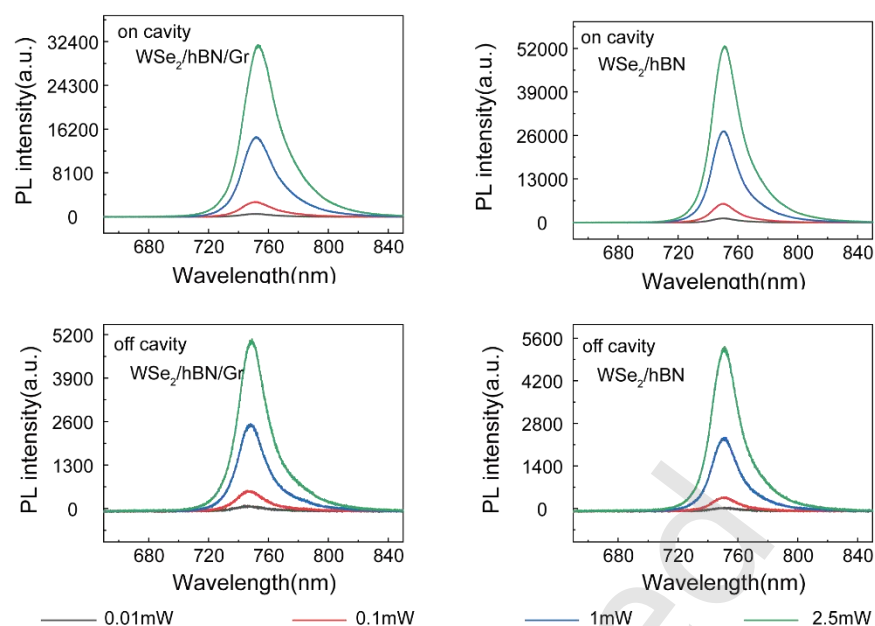


Figure S9 Power-dependent integrated PL intensity from the same regions under 532 nm laser excitation; Under varying optical excitation across distinct spatial domains, the PL intensity exhibits a monotonic enhancement with increasing incident laser power.

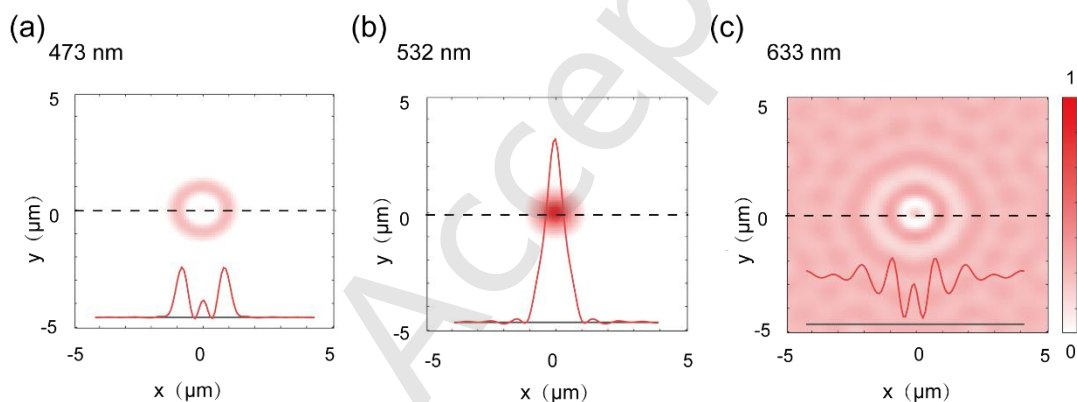


Figure S10 (a) Local field intensity profiles calculated for excitation wavelengths of 473 nm, 532 nm, and 633 nm;

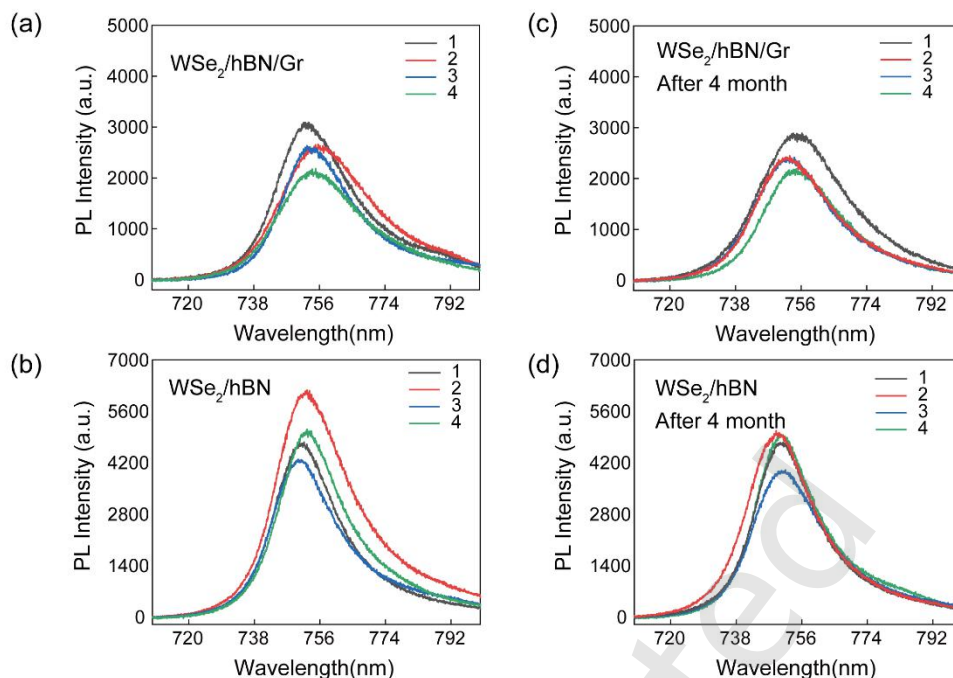


Figure S11 (a)-(b) PL spectra measured three months ago; (c)-(d) the recently measured PL spectra.

We remeasured the device which kept in a nitrogen-filled glovebox for approximately three months under identical excitation and collection conditions. We selected four distinct spots on both the on-cavity WSe_2/hBN and $\text{WSe}_2/\text{hBN}/\text{Gr}$ regions. In the newly added Supplementary Figure S11, (a) and (b) display the PL spectra measured four months ago, whereas (c) and (d) present the recently measured PL spectra from the same regions. By integrating the emission peak areas and calculating the average values, we observed that the overall PL intensity of the device decreased by $\sim 11\%$ after four months.

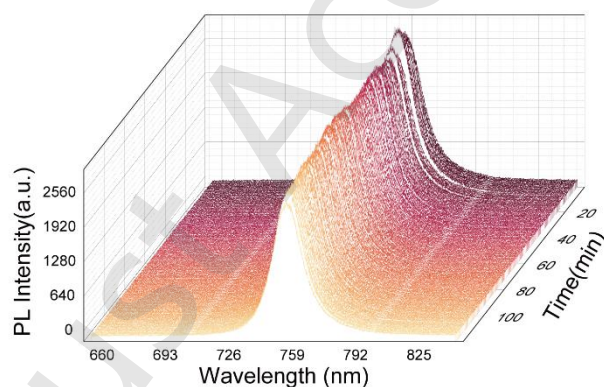


Figure S12 Long-term photostability of the cavity-integrated $\text{WSe}_2/\text{hBN}/\text{Gr}$ heterostructure.

2D contour map of the time-dependent PL spectra acquired from the on-cavity heterostructure region under continuous laser excitation at room temperature. The spectra were recorded at 1-minute intervals over a duration of 120 minutes. The invariant PL peak position and stable emission intensity explicitly demonstrate the exceptional operational robustness of the device against photodegradation and localized thermal effects.