

# Strongly Nonlinear Nanocavity Exciton Polaritons in Gate-Tunable Monolayer Semiconductors

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Achieving optical nonlinearities at ultralow light intensities in solid-state platforms is essential for advancing nonlinear and quantum photonic technologies. A promising approach involves coupling excitons to photons in optical cavities to create exciton polaritons, where effective photon-photon interactions are mediated by the intrinsic excitonic nonlinearity. However, realizing strong polariton nonlinearities within a scalable architecture remains a significant experimental challenge. Here, we demonstrate highly nonlinear two-dimensional exciton polaritons by coupling a charge-tunable MoSe<sub>2</sub> monolayer to a planar photonic crystal nanocavity. The pronounced excitonic resonance of the monolayer, combined with its seamless integration with the planar nanocavity, facilitates robust exciton-photon hybridization. Remarkably, the strong mode confinement of the nanocavity substantially enhances polariton-polariton interactions, enabling all-optical switching of the cavity spectrum with excitation energies as low as  $\sim 4$  fJ—several orders of magnitude below previously reported thresholds in 2D exciton-polariton systems. Pump-probe spectroscopy reveals that this switching operates on an ultrafast timescale of a few picoseconds. Our Letter establishes a robust platform for nonlinear 2D polaritonics with broad applications in integrated photonic technologies, including all-optical neuromorphic computing and quantum photonic information processing.

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The coherent coupling between excitonic resonances in solid-state materials and optical fields in a cavity gives rise to hybrid light-matter quasiparticles known as exciton polaritons. Exhibiting dual characteristics of light and matter, these quasiparticles underpin a wide range of intriguing classical and quantum phenomena, including polariton lasing [1], Bose-Einstein condensation [2], and superfluidity [3]. Among their many remarkable properties, exciton polaritons are particularly appealing for their pronounced nonlinearity mediated by their underlying excitonic constituents. This nonlinear response far exceeds that of conventional nonlinear optical materials [4], providing a promising pathway toward all-optical computing and photonic quantum information processing. Historically, representative exciton-polariton systems coupling III-V quantum

wells [5,6], quantum dots [7,8], or organic semiconductors [9] to optical microcavities have enabled the observation of nonlinear optical phenomena at extremely low light intensities, even down to the single-photon level [7,8]. However, their widespread deployment is hindered by intrinsic material trade-offs. For example, III-V exciton-polariton systems are restricted to cryogenic temperatures and suffer from surface-induced degradation during nanofabrication, while organic systems often lack material stability and process compatibility [10]. These limitations prevent the realization of dense, scalable polaritonic circuits.

Recently, the emergence of two-dimensional (2D) transition metal dichalcogenides (TMDs) has established a new frontier for exploring nonlinear exciton polaritons in the atomically thin limit [11]. The sharp exciton resonances of 2D TMD materials—arising from strong quantum confinement and reduced dielectric screening in 2D materials—combined with their facile integration with photonic platforms, enable the realization of robust 2D nonlinear exciton polaritons [12–18]. In such hybrid light-matter systems, achieving strong light confinement in all three dimensions is essential for enhancing both exciton-photon and effective polariton-polariton interactions. For instance, when a uniform 2D TMD flake is integrated with a planar optical cavity (assuming the flake size is comparable to the lateral

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dimensions of the cavity mode), vertical optical field confinement plays a key role in strengthening exciton-photon coupling and thereby increasing the Rabi splitting  $\Omega$ . Meanwhile, effective polariton-polariton interactions arise from either short-range exciton-exciton interactions [19,20] or excitonic state depletion (also known as the phase-space filling effect) [12–14]. Tight lateral optical confinement enhances both mechanisms by increasing the spatial overlap of excitons and reducing the number of available exciton states within the cavity, thereby substantially boosting polariton nonlinearity [19,20]. However, most prior work on 2D exciton polaritons has relied on extended optical cavities, such as distributed Bragg reflectors (DBRs), where optical fields spread over one or more dimensions. This results in large mode volumes—typically from  $\sim 1 \mu\text{m}^3$  to  $> 10 \mu\text{m}^3$  [5,19,20]—and consequently diminished exciton-photon and polariton-polariton interactions. This weak light confinement remains a key barrier to reducing the polariton nonlinear threshold. Conversely, while plasmonic structures can offer deep subwavelength confinement, they inherently suffer from high optical losses due to metal absorption [21]. These losses significantly broaden the polariton linewidths, suppress polariton accumulation, and ultimately hinder the realization of nonlinear polaritonic effects.

Another major challenge in integrating 2D TMD materials with optical cavities is controlling the charge doping of the 2D semiconductors. Unintentional doping, often introduced during exfoliation and transfer, can significantly impact their excitonic properties. While electrostatic gating offers a means to tune the charge concentration, most gate-tunable devices rely on metallic gates such as graphite or metals, which either introduce substantial optical losses [22] or require extended cavity geometries [12,23]. As a result, developing electrostatic tuning mechanisms for 2D TMD-cavity systems that preserve their optical performance remains a significant experimental hurdle.

Here, we demonstrate robust and highly nonlinear 2D exciton polaritons by integrating a gate-tunable  $\text{MoSe}_2$  monolayer with a silicon nitride photonic crystal (PhC) nanocavity. Notably, our use of a 2D heterostructure enables precise control over the doping level in the  $\text{MoSe}_2$  monolayer, allowing for strong excitonic responses without perturbing the nanocavity resonance. Meanwhile, the nanocavity features an ultracompact mode volume of  $\sim 0.05 \mu\text{m}^3$ , which is several orders of magnitude smaller than that of conventional DBR cavities. The enhanced light confinement in all three dimensions not only boosts the exciton-photon coupling strength, but also facilitates strong polariton nonlinearity. Remarkably, we achieve all-optical switching of the cavity spectrum with excitation energies as low as  $\sim 4 \text{ fJ}$ , establishing a new benchmark for switching energy in 2D exciton-polariton systems. Pump-probe measurements further confirm that the spectral switching occurs on an ultrafast timescale of only a few picoseconds,

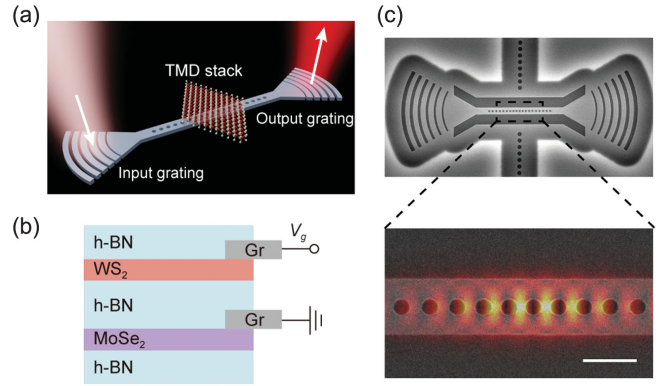


FIG. 1. 2D nanocavity exciton polaritons. (a) Schematic of the coupled TMD-PhC nanocavity. (b) Schematic of the gate-tunable TMD stack. (c) Scanning electron microscope image of the suspended  $\text{Si}_3\text{N}_4$  nanobeam cavity, with the inset showing the simulated cavity mode profile. The dark area is suspended from the  $\text{SiO}_2$  substrate. Scale bar, 500 nm.

limited by the exciton lifetime. Our Letter represents a significant step toward realizing robust and scalable nonlinear polaritonics on an integrated photonic platform for both classical and quantum applications.

The TMD-PhC nanocavity is schematically shown in Fig. 1(a). The TMD stack consists of  $\text{MoSe}_2$  and  $\text{WS}_2$  monolayers separated by a  $\sim 10 \text{ nm}$  thick hexagonal boron nitride (hBN) layer [Fig. 1(b)], forming a parallel-plate capacitor. Our Letter focuses on excitons in the  $\text{MoSe}_2$  monolayer, with the  $\text{WS}_2$  monolayer serving as a transparent top gate that minimizes optical absorption near the  $\text{MoSe}_2$  exciton energy. Both TMD monolayers are in contact with graphite electrodes for electrostatic gating, and the entire stack is encapsulated with  $\sim 5 \text{ nm}$  thick top and bottom hBN layers. To achieve strong light confinement in all three dimensions, we fabricate a photonic crystal nanobeam cavity on a silicon nitride-on-insulator wafer with a  $145 \text{ nm}$  thick silicon nitride layer ( $n = 2.17$ ). Figure 1(c) shows a scanning electron microscope image of the resulting device. The structure is suspended from the  $\text{SiO}_2$  substrate (visible as the dark region) to enhance refractive index contrast and optical confinement. To facilitate gas release during the dry transfer process, an array of circular holes is patterned perpendicular to the nanobeam cavity. To probe the cavity resonance, light is coupled into and out of the cavity using a pair of grating couplers at the two ends. In the absence of the TMD stack, the bare cavity exhibits an optical resonance at  $1.736 \text{ eV}$  with a full-width-half-maximum linewidth of  $\gamma_{\text{cav}} = 1.9 \text{ meV}$ . The cavity linewidth is determined by radiative coupling to the waveguides on both sides ( $\gamma_{\text{cav,r}} \sim 0.5 \text{ meV}$ ) and nonradiative optical losses due to scattering from fabrication imperfections and material absorption ( $\gamma_{\text{cav,nr}} \sim 1.4 \text{ meV}$ ). The TMD stack is transferred onto the photonic chip with prepatterned gold electrodes using a standard dry transfer

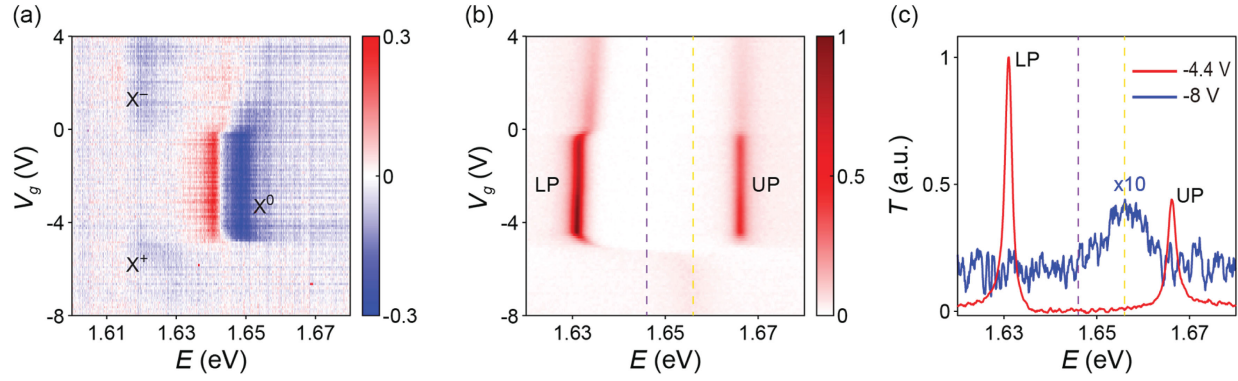


FIG. 2. Linear characterization of the coupled TMD-PhC nanocavity. (a) Gate-dependent reflectance spectrum ( $dR/R$ ) features excitons ( $X^0$ ) in the charge-neutral regime and trions ( $X^\pm$ ) in the doped regime. The plot is normalized to the reflectance spectrum measured at  $V_g = -10$  V, where no excitonic features are observed. (b) Gate-dependent cavity transmission spectrum measured through grating couplers, featuring lower polaritons (LP) and upper polaritons (UP). (c) Transmission spectra measured in the charge-neutral (red) and  $p$ -doped regime (blue). In panels (b) and (c), the purple and yellow dashed lines mark the exciton and cavity resonances, respectively.

technique. The presence of the TMD stack redshifts the cavity resonance due to modified dielectric environment, resulting in a cavity-exciton detuning  $\delta_{c-X} \sim 10$  meV at cryogenic temperatures. Meanwhile, it is also estimated to increase the cavity-waveguide coupling to  $\gamma_{\text{cav,r}} \sim 1$  meV, owing to reduced photonic band gap confinement (see Supplemental Material [24] for details). Notably, we intentionally design the cavity-waveguide coupling strength to match the non-radiative-limited exciton linewidth  $\gamma_{X,\text{nr}} \sim 1$  meV [25,26], ensuring narrow polariton linewidths in the strong coupling regime while maintaining robust optical access through the waveguides.

The coupled TMD-PhC device is measured at 4 K. We first characterize the linear optical properties of MoSe<sub>2</sub> excitons via gate-dependent reflectance measurements in a confocal microscope setup. A broadband light source is tightly focused to a  $\sim 1$   $\mu\text{m}$  spot at the cavity center using a high-NA objective (NA = 0.75), and the reflected light is collected. The reflectance spectra in Fig. 2(a) reveal rich excitonic features that vary significantly with the applied gate voltage [27,28]. Specifically, in the charge-neutral regime ( $-4.5$  V  $< V_g < 0$  V), the optical response is dominated by a strong neutral exciton resonance ( $X^0$ ) at 1.646 eV, with a linewidth of  $\gamma_X = 5.4$  meV. As  $V_g$  is tuned above 0 V or below  $-4.5$  V, the MoSe<sub>2</sub> monolayer becomes  $n$ - or  $p$ -doped, respectively, leading to the suppression of the neutral exciton resonance and the emergence of weak trion resonances ( $X^\pm$ ) at 1.616 eV. We observe a sharp transition between the charge-neutral and doped regimes, which we attribute to Schottky barriers at the TMD-graphite interfaces that hinder continuous doping control in the monolayer [29]. Additionally, we observe an asymmetric gate response between the  $n$ - and  $p$ -doped regimes: the  $X^-$  feature remains nearly constant for  $V_g > 0$  V, whereas the  $X^+$  feature varies strongly for  $V_g < -4.5$  V. This asymmetry likely arises from reduced gating efficiency in the  $n$ -doped regime, which we

attribute to the quantum capacitance of the WS<sub>2</sub> monolayer [30].

Next, we investigate the linear optical properties of cavity exciton polaritons arising from the strong hybridization between MoSe<sub>2</sub> neutral excitons and cavity photons. For this, light is coupled into the waveguide and the transmitted light is collected through grating couplers. The measured gate-dependent cavity transmission spectra are shown in Fig. 2(b). In the charge-neutral regime, where the neutral exciton oscillator strength is maximal, the spectra exhibit two prominent peaks at 1.666 and 1.631 eV, corresponding to upper (UP) and lower (LP) polariton states, respectively. The measured linewidths are 2.3 meV for the UP and 1.8 meV for the LP. Using a coupled oscillator model, we extract the exciton-photon coupling strength  $g = 16.8$  meV. This strong exciton-photon coupling reflects the tight vertical optical confinement enabled by the high index contrast of the nanocavity. In contrast, as the system is tuned into the  $p$ -doped regime ( $V_g < -4.5$  V), the two polariton peaks merge into a single peak at 1.656 eV, effectively recovering the bare nanocavity resonance but with a significantly broader linewidth. This strong-to-weak coupling transition reflects a reduction in the exciton oscillator strength with increasing doping level, consistent with the previous reflection measurement [Fig. 2(a)]. The measured broad cavity linewidth of 9.9 meV and the suppressed optical transmission in the  $p$ -doped regime together suggest increased optical absorption at high doping levels [31] [Fig. 2(c)]. A similar reduction in exciton-photon coupling is observed for  $n$ -doping ( $V_g > 0$  V), but the system remains in the strong coupling regime due to the residual exciton oscillator strength.

We then investigate the nonlinear responses of excitons and cavity polaritons using femtosecond laser excitation (pulse duration  $\sim 50$  fs, spectral width  $\sim 60$  meV, repetition rate 3 kHz). To characterize the neutral exciton nonlinearity, we fix the pump photon energy at 1.63 eV and

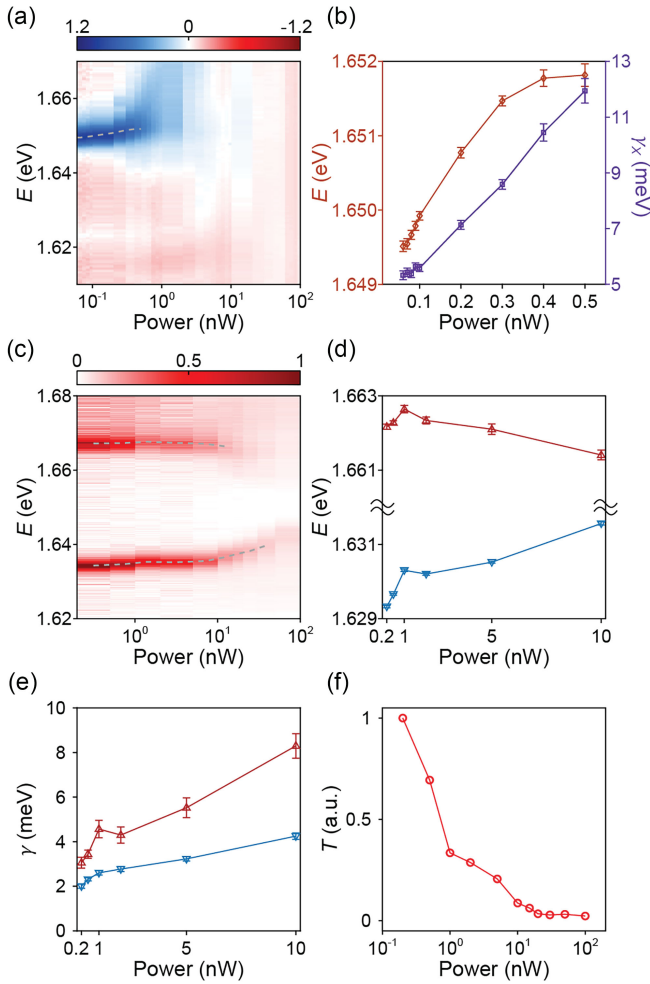


FIG. 3. Nonlinear cavity exciton polaritons. (a) Exciton reflectance spectra as a function of excitation power, measured at  $V_g = -3.9$  V. The gray dashed line guides the eye for the evolution of the exciton resonance. (b) Extracted exciton energy (orange) and linewidth (purple). (c) Cavity transmission spectra as a function of excitation power, measured at  $V_g = -3$  V. (d), (e) Extracted energies and linewidths for the UP (red) and LP (blue), respectively. (f) Normalized cavity transmission at the LP resonance ( $E_{LP} = 1.629$  eV) as a function of excitation power.

measure the reflectance spectra as a function of optical power [Fig. 3(a)]. The extracted exciton parameters [Fig. 3(b)] show that increasing the excitation power from 0.1 to 0.4 nW induces a blueshift of  $\sim 2$  meV and accompanying linewidth broadening. We attribute the blueshift to energy renormalization driven by exciton-exciton interactions [13,14], while the linewidth broadening results from excitation-induced dephasing (EID) due to scattering with the incoherent exciton reservoir (i.e., the population of excitons that have lost phase coherence with the laser field). This EID effect has been widely observed in conventional 3D and quasi-2D semiconductors [32,33], and more recently in 2D TMD monolayers [34].

To investigate how this exciton nonlinearity affects the polariton response, we measure the cavity transmission

spectra as a function of input optical power (pump photon energy at 1.647 eV). As shown in Fig. 3(c), under low-power excitation, both LP and UP peaks remain clearly observable, consistent with the linear spectra in Fig. 2(b). Their energies are slightly redshifted by a few meV relative to the linear measurement due to the slow temporal drift of the cavity resonance (see Supplemental Material [24] for details). The extracted polariton parameters [Figs. 3(d) and 3(e)] reveal that as the excitation power is increased from 0.2 to 1 nW (measured before the input grating coupler), the LP and UP energies exhibit blueshifts of 1 and 0.5 meV, respectively. However, at even higher excitation powers, the LP continues to blueshift whereas the UP redshifts. In contrast to these diverging energy shifts, the linewidths of both polariton modes broaden across the entire power range, resulting in damped polariton resonances and a suppression of optical transmission. We note that the cavity polaritons exhibit a less pronounced power dependence compared to bare excitons. This reduced sensitivity arises from excitation-induced broadening of the polariton linewidths, which diminishes the waveguide-cavity coupling efficiency with increasing excitation power. Nevertheless, our device operates as a highly efficient all-optical switch. As shown in Fig. 3(f), the normalized cavity transmission at the LP resonance ( $E_{LP} = 1.629$  eV) drops by 50% at excitation power less than 1 nW. Given that the spectral width of the pulsed laser (60 meV) is significantly broader than the polariton linewidths, as a result, less than 7% of the pulse energy contributes to polariton excitation. Accounting for both this spectral mismatch and the grating coupler efficiency yields a nonlinear switching energy as low as 4 fJ.

The observed asymmetric polariton energy shifts originate from the interplay of two concurrent mechanisms, which can be qualitatively described by the coupled oscillator model. Specifically, the eigenvalues of the coupled TMD-PhC nanocavity are given by

$$E_{\pm} = (E_X + E_{\text{cav}})/2 - i(\gamma_X + \gamma_{\text{cav}})/4 \pm \sqrt{g^2 + \frac{1}{4} \left[ \delta_{\text{c-X}} - i \frac{(\gamma_{\text{cav}} - \gamma_X)}{2} \right]^2}.$$

Note that  $\gamma_X$  used in the coupled oscillator model differs from the bare material value due to the strong suppression of radiative decay in nanophotonic structures [35–38]. The Rabi splitting  $\Omega$  between UP and LP can be expressed as

$$\Omega = \text{Re} \left\{ 2 \sqrt{g^2 + \frac{1}{4} \left[ \delta_{\text{c-X}} - i \frac{(\gamma_{\text{cav}} - \gamma_X)}{2} \right]^2} \right\}.$$

Increasing pump power blueshifts the exciton energy  $E_X$ , pushing both LP and UP to higher energies. Simultaneously, the significant broadening of the exciton linewidth  $\gamma_X$  reduces the effective Rabi splitting  $\Omega$ .

Crucially, this contraction of the Rabi splitting reinforces the blueshift for the LP while counteracting it for the UP. At high pump powers, the splitting reduction dominates the excitonic blueshift for the UP, resulting in the net redshift observed in Fig. 3(c). A more rigorous theoretical analysis of the nonlinear polariton properties requires accounting for the incoherent couplings between the two polariton states and the exciton reservoir, going beyond the minimal coupled oscillator model [32,33].

To demonstrate the ultrafast timescale of the polariton nonlinearity, we employ pump-probe spectroscopy to measure the transient cavity transmission. A pump beam is focused onto the cavity center at normal incidence from free space. Crucially, the pump beam does not excite cavity polaritons directly, as the polariton modes exhibit negligible radiative coupling to free space. Instead, it generates an exciton population, which modulates the cavity polaritons via exciton-exciton interactions. Meanwhile, a probe beam (derived from the same laser source but significantly attenuated) is coupled into the cavity through the grating coupler to monitor the cavity transmission spectra as a function of the time delay  $\tau$ . The resulting transient transmission spectra are shown in Fig. 4. Under low pump power [1 nW, Fig. 4(a)], the LP begins to blueshift at positive delay, reaching a maximum shift of 1.3 meV at  $\tau \sim 133$  fs, before gradually relaxing back to its equilibrium energy within  $\sim 3$  ps. In contrast, the UP mode shows

no significant energy shift, consistent with the previous power-dependent cavity spectra measurement [Fig. 3(c)]. This transient transmission suppression becomes more pronounced at higher pump power [5 nW, Fig. 4(b)], where the cavity transmission is quenched within  $\sim 200$  fs. In addition to the fast dynamics, we also observe a reduction in transmission for both LP and UP at much longer positive delays, with full recovery to equilibrium occurring on the timescale of hundreds of picoseconds (see Supplemental Material [24] for details). The slow recovery is likely due to the excitation of impurity-trapped exciton states, which exhibit relatively long lifetimes on the order of several hundred picoseconds [25]. Further investigation is needed to clarify the origin of these long-lived excitations and to explore potential mitigation strategies, such as the use of high-quality bulk crystals with reduced defect densities.

Finally, we outline the potential strategies to further reduce the current switching energy of  $\sim 4$  fJ (corresponding to  $N \sim 10^4$  intracavity polaritons). A reduction of two to three orders of magnitude is achievable through synergistic improvements in both materials and photonic device design. On the materials side, leveraging excitonic states with reduced density of states—such as trions in TMD monolayers [12,13] or moiré excitons [14] in TMD heterostructures—can yield nonlinear excitonic responses one to two orders of magnitude stronger than those of the neutral excitons used in this Letter. These engineered excitonic states can be seamlessly integrated into our hybrid TMD-PhC platform and are expected to significantly enhance polariton nonlinearity. On the photonic side, employing high-index-contrast, low-loss systems such as the recently developed InGaP-on-insulator platform [39] ( $n_{\text{InGaP}} = 3.4$ ) would provide tighter mode confinement and longer polariton lifetimes [38], thereby reducing the switching energy by at least an order of magnitude. Together, these improvements could lower the nonlinear threshold by over three orders of magnitude, paving the way toward few- ( $N < 10$ ) or even single-polariton nonlinearities.

In summary, we demonstrate strongly nonlinear 2D exciton polaritons by coupling a gate-tunable TMD monolayer to a planar photonic crystal nanocavity, achieving ultrafast all-optical switching with femtojoule energies. This represents a drastic reduction compared to the picojoule-range thresholds of state-of-the-art 2D exciton-polariton systems [40], bringing our platform within striking distance of the quantum nonlinear regime realized in III-V polaritons. Our findings establish a robust and scalable platform for nonlinear 2D exciton polaritons with promising applications in both classical and quantum photonics. In particular, this system could accelerate the development of all-optical neural networks for artificial intelligence, where computation occurs entirely in the optical domain—offering unprecedented speed and energy efficiency beyond the reach of electronic architectures. Furthermore, with a clear pathway to the single-polariton

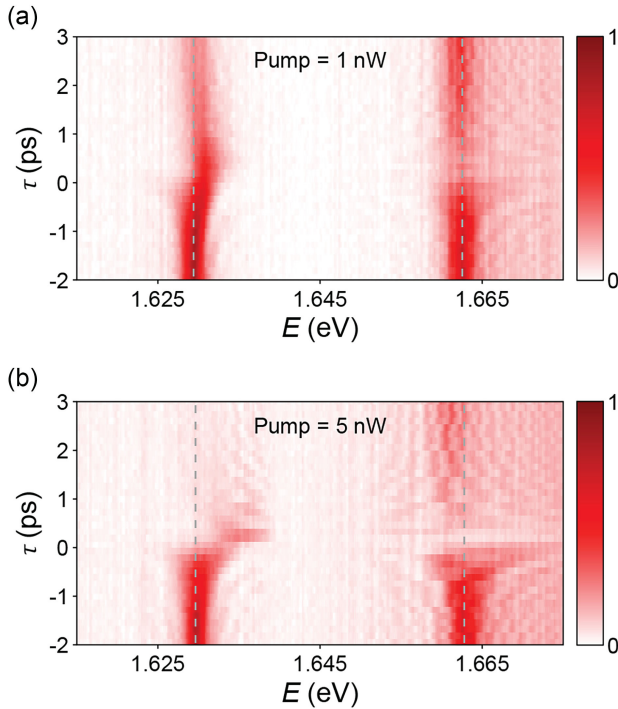


FIG. 4. Pump-probe spectroscopy of nonlinear cavity polaritons at (a) 1 nW and (b) 5 nW excitation powers. The gray dashed lines represent the equilibrium energies of the LP and the UP states.

limit, our platform is poised to unlock scalable quantum photonic information processing and the exploration of strongly correlated states of light [41].

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L. H. and B. Z. conceived the idea. Z. W. fabricated the device. Z. W. and L. H. performed the experiment. All authors discussed the results. L. H. and B. Z. wrote the paper with contributions from all authors. B. Z. supervised the project.

**Data availability**—The data are not publicly available. The data are available from the authors upon reasonable request.

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