

LETTER

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Nonlinear polaritons in a monolayer semiconductor coupled to optical bound states in the continuum

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Abstract

Optical bound states in the continuum (BICs) provide a way to engineer very narrow resonances in photonic crystals. The extended interaction time in these systems is particularly promising for the enhancement of nonlinear optical processes and the development of the next generation of active optical devices. However, the achievable interaction strength is limited by the purely photonic character of optical BICs. Here, we mix the optical BIC in a photonic crystal slab with excitons in the atomically thin semiconductor MoSe₂ to form nonlinear exciton-polaritons with a Rabi splitting of 27 meV, exhibiting large interaction-induced spectral blueshifts. The asymptotic BIC-like suppression of polariton radiation into the far field toward the BIC wavevector, in combination with effective reduction of the excitonic disorder through motional narrowing, results in small polariton linewidths below 3 meV. Together with a strongly wavevector-dependent *Q*-factor, this provides for the enhancement and control of polariton–polariton interactions and the resulting nonlinear optical effects, paving the way toward tuneable BIC-based polaritonic devices for sensing, lasing, and nonlinear optics.

Introduction

Optical bound states in the continuum (BICs), supported by photonic crystal structures of certain geometries, have received much attention recently as a novel approach to generating extremely spectrally narrow resonant responses^{1,2}. Since BICs are uncoupled from the radiation continuum through symmetry protection³ or resonance trapping⁴, they can be robust to perturbations of photonic crystal geometric parameters⁵. This robustness enables a broad range of practical applications,

including recently demonstrated spectral filtering⁶, chemical and biological sensing^{7,8}, and lasing⁴.

Providing an efficient light-trapping mechanism, optical BICs are particularly attractive for enhancing nonlinear optical effects^{9,10}, with recent theoretical proposals discussing enhanced bistability¹¹ and Kerr-type focusing nonlinearity¹². However, for the practical realization of these proposals, a significantly stronger material nonlinear susceptibility than that generally available in dielectric-based photonic crystals is needed.

An attractive approach to the enhancement of the effective nonlinearity is to use exciton-polaritons—hybrid quasiparticles that inherit both the coherent properties of the photonic modes and the interaction strength of the excitons^{13–15}. Hybrid nanophotonic systems incorporating atomically thin transition metal dichalcogenides (TMDs) have emerged as a particularly promising

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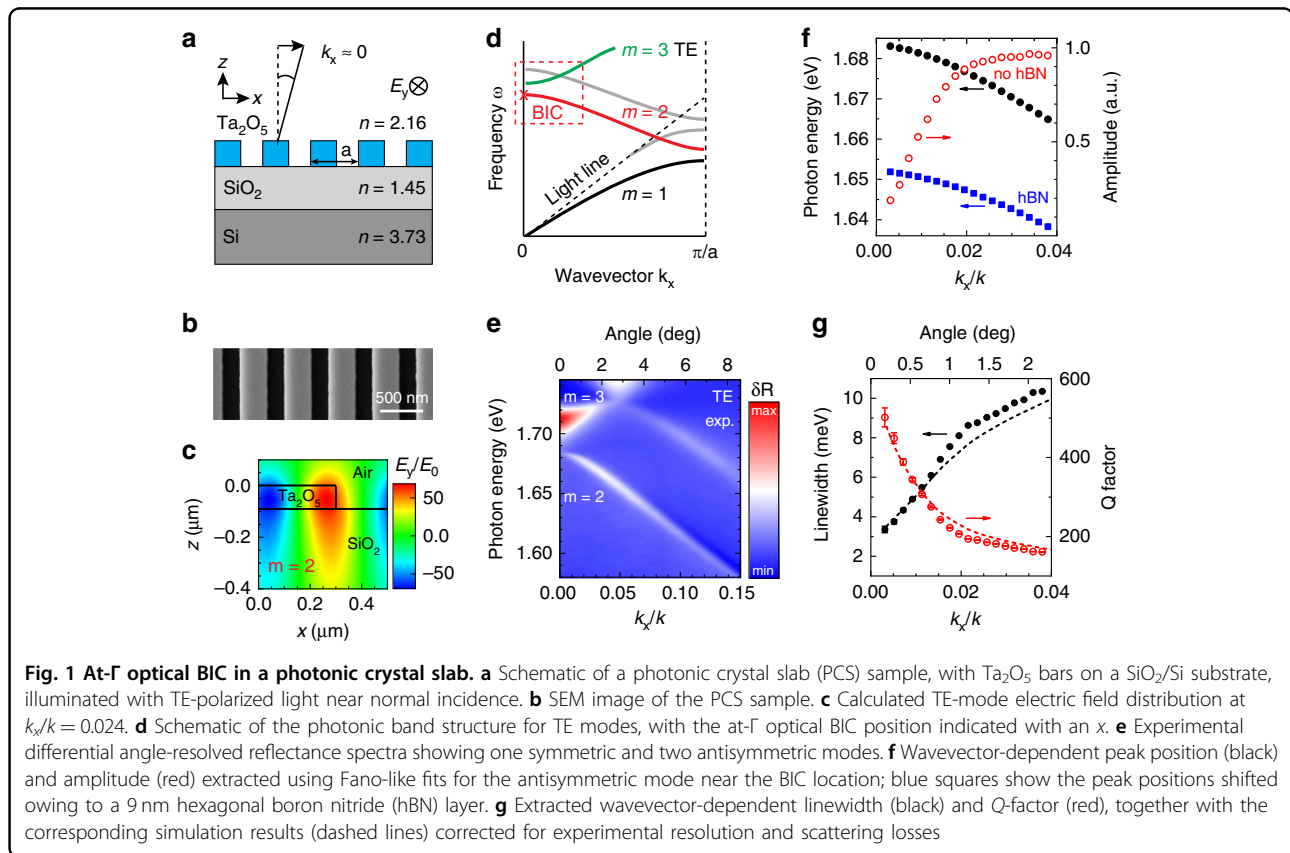
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platform owing to their ease of fabrication and the possibility of room-temperature operation^{16–18}. In addition to conventional microcavity-based designs, TMD exciton-polaritons have been observed in plasmonic lattices¹⁹, photonic crystal slabs (PCSs)^{20,21}, and other nanophotonic structures²².

Coupling TMD excitons to optical BICs in photonic crystals will not only boost the potentially achievable nonlinearities but also provide control on the resonant BIC properties through the excitonic fraction in the polariton, as has been proposed theoretically²³.

Here, we experimentally demonstrate and investigate nonlinear polaritons formed via the strong coupling of excitons in monolayer (1 L) MoSe₂ and optical BICs in a 1D PCS, with Rabi splitting of >27 meV and BIC-like radiation suppression in the surface-normal direction. Despite the large ~9 meV inhomogeneous broadening of the MoSe₂ excitonic line, we achieve a small polariton linewidth below 3 meV, corresponding to a very well resolved splitting-to-linewidth ratio of ~9 and Q-factors up to 900. Using the strongly wavevector-dependent Q-factor of the photonic crystal dispersion, we show a controllable reduction in the polariton linewidth by a factor of 5–10 when approaching the BIC. The narrow polariton lines allow us to accurately measure the polariton–polariton interaction strength through power-

dependent spectral blueshifts in the resonant reflectance experiment, corresponding to an exciton–exciton interaction strength of $g_X \sim 1.0 \mu\text{eV} \mu\text{m}^2$. This polariton nonlinearity is comparable with the values measured in III–V materials^{24,25} and significantly larger than those previously observed in TMD monolayer-based systems²², paving the way toward quantum applications of exciton-polaritons in atomically thin semiconductors.

In the experiment, we fabricate a 1D PCS sample consisting of 90 nm thick Ta₂O₅ bars on a SiO₂/Si (1 μm/500 μm) substrate, as schematically shown in Fig. 1a, with a scanning electron microscopy image shown in Fig. 1b. The PCS geometry (see “Methods”) is designed for large refractive index modulation to open a photonic band gap and support an optical BIC close to the exciton resonance in monolayer MoSe₂. As illustrated in the photonic band structure shown in Fig. 1d, the BIC is expected to form on the lower-energy $m = 2$ TE mode (red) at the Γ point in the crystal momentum space²³, with characteristic confinement and antisymmetric spatial distribution of the optical field (c) with respect to the mirror symmetry plane of the PCS cell (see Supplementary Fig. S6).

We measure the PCS band structure via angle-resolved reflectance spectroscopy (see “Methods” and Supplementary Fig. S1). Figure 1e shows the experimental differential reflectance spectra for varying angle θ , where the

signal from the un-patterned $\text{Ta}_2\text{O}_5/\text{SiO}_2/\text{Si}$ substrate is subtracted for clarity: $\delta R(\theta, \omega) = R_{\text{PCS}}(\theta, \omega) - R_{\text{Sub}}(\theta, \omega)$. Three modes, a broad symmetric one ($m=3$) and two narrower antisymmetric ones ($m=2$), are clearly observed in the figure, in agreement with the theory (d, red dashed box). We fit the lower-energy antisymmetric mode peak in the reflectance spectra using a Fano-like line shape $F(\omega) \propto (q\gamma/2 + \omega - \omega_0)^2/(\gamma^2/4 + (\omega - \omega_0)^2)$ with resonance frequency ω_0 , linewidth γ , and asymmetry parameter q , which arises due to interference with the broad symmetric mode and an even broader Fabry–Perot response of the layered substrate (see Supplementary Fig. S2).

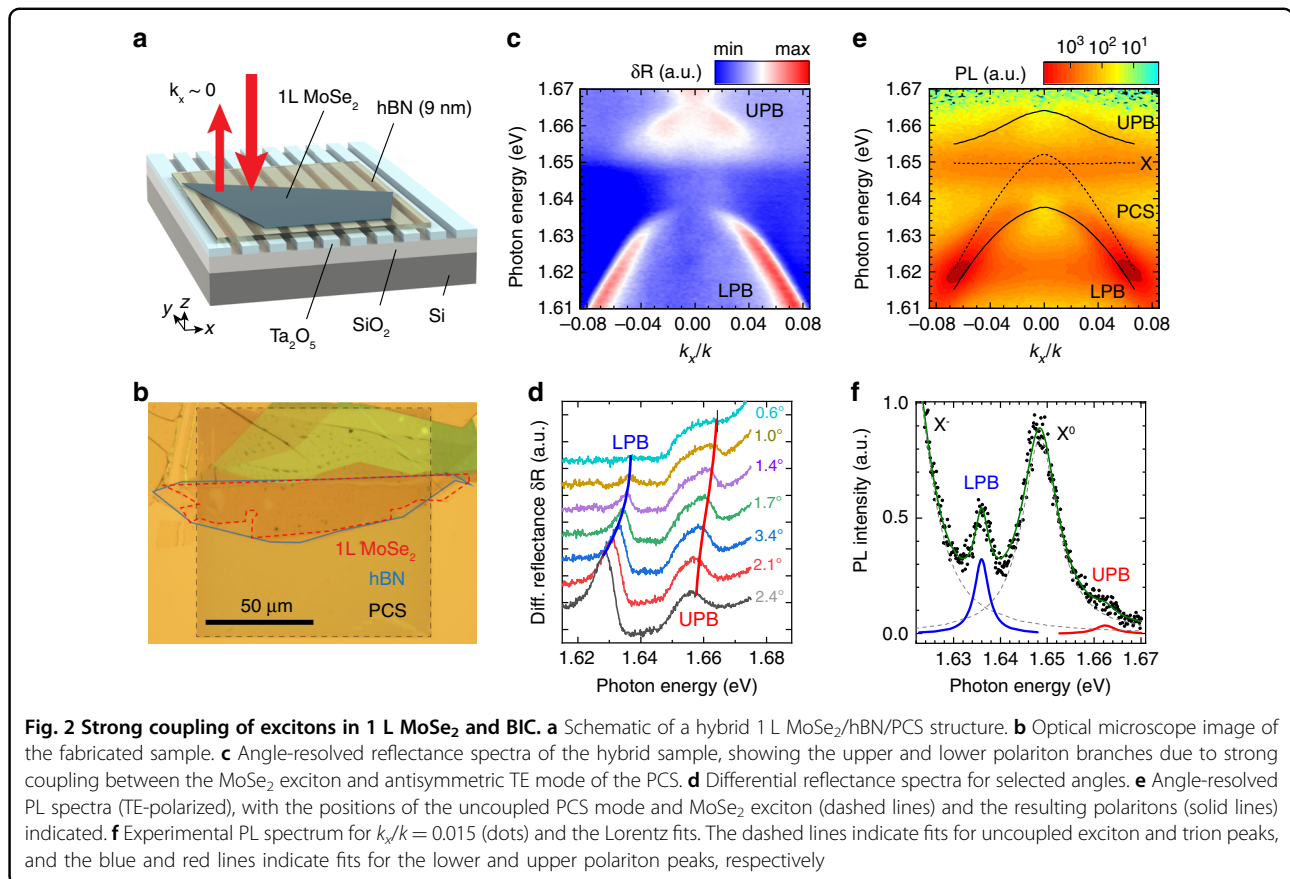
The extracted Fano fit parameters are plotted in Fig. 1f, g as functions of the ratio $k_x/k = \sin\theta$, where k_x is the in-plane wavevector component, k is the free-space wavevector magnitude, and θ is the angle with respect to the surface normal. Toward the Γ point ($k_x \rightarrow 0$), the reflectivity associated with the mode sharply decreases (f, red), while the spectral line narrows (g, black circles), resulting in a sharply increasing Q -factor (g, red open circles), defined as $Q = \omega_0/\gamma$. This behavior is a characteristic of an at- Γ optical BIC²⁶, where the interference of optical waves outgoing in opposite directions leads to effective light trapping in the near field and vanishing far-field radiation.

This is in contrast to the case studied recently^{20,21}, where only radiating PCS modes with smaller and largely angle-independent Q -factors were considered for strong coupling to excitons in 2D semiconductors.

The theoretically predicted radiative Q -factor of the BIC diverges toward infinity at $k_x = 0$. In practice, the measured Q -factor is limited predominantly by nonradiative losses^{5,26}. The two major contributions in our case are (1) intrinsic absorption in Ta_2O_5 and (2) surface-roughness-induced symmetry breaking and scattering²⁷, limiting Q to $\sim 10^3$. Additional resonance broadening comes from leaky losses in the Si due to near-field penetration through the SiO_2 layer²⁷ and finite sample size effects^{3,28}.

We simulate the PCS dispersion and associated Q -factors with the Fourier modal method, taking into account these four loss mechanisms (see Supplementary Note 1 and Fig. S3). As shown in Fig. 1g (dashed lines), good agreement with the experiment can be achieved by considering the scattering losses through an additional imaginary part of the Ta_2O_5 refractive index $\delta n \sim 0.002i$ (Fig. 1g, dashed lines).

We then create polaritons by coupling the observed BIC to excitons in the monolayer MoSe_2 in a vertically stacked structure consisting of 1 L MoSe_2 , multilayer hexagonal boron nitride (hBN), and a PCS, as illustrated in Fig. 2a.



To maximize the Q -factor of the resulting polariton modes, we use large-area multilayer hBN and monolayer MoSe₂ flakes of $\sim 100\ \mu\text{m}$ in size, covering ~ 200 periods of the PCS, as demonstrated in the optical microscope image in (b). The hBN spacer plays a threefold role: it avoids MoSe₂ flakes “sagging” into the PCS grooves, reduces the influence of the Ta₂O₅ surface roughness, and provides tunability of the BIC frequency through the hBN thickness. In our case, a 9 nm thick hBN spacer shifts the PCS mode and spectral position of the BIC by $\sim 30\ \text{meV}$ to bring it close to resonance with the neutral exciton in the 1 L MoSe₂ at 7 K, with $\hbar\omega_X = 1.65\ \text{eV}$ (Fig. 1f, blue squares).

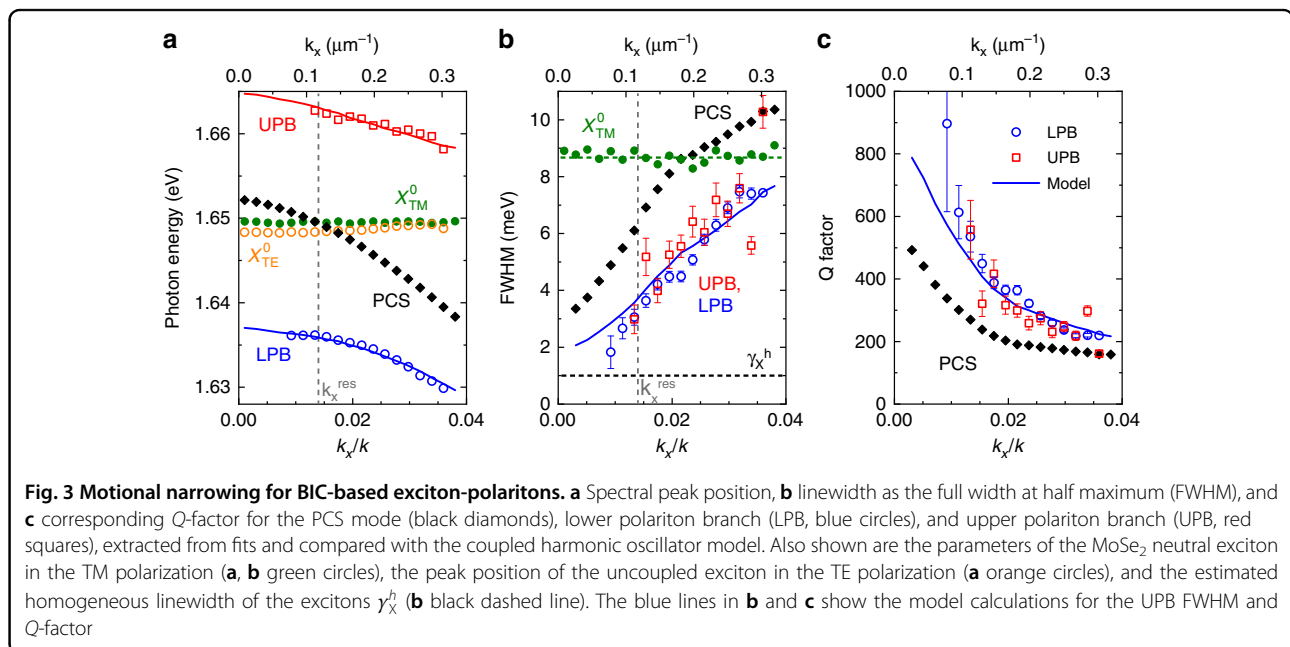
We study the polaritons experimentally via angle-resolved reflectivity and photoluminescence (PL) measurements at 7 K (see “Methods”), with the results of TE-polarized detection shown in Fig. 2c–f. In comparison with Fig. 1e, the lower-energy antisymmetric PCS mode observed in reflectivity (Fig. 2c, d) is now redshifted by $\sim 30\ \text{meV}$ owing to the presence of hBN/MoSe₂ and split into upper and lower polariton branches (UPB and LPB, respectively) owing to strong coupling with the neutral exciton in the 1 L MoSe₂ centered at $\hbar\omega_X = 1.65\ \text{eV}$ (see also Supplementary Fig. S4). Both the LPB and UPB retain BIC-like behavior near the Γ point, exhibiting several distinctive properties.

First, at the Γ point, both the LPB and UPB are “dark,” as radiation into the far field becomes symmetry forbidden, effectively extending the interaction time for potential enhancement of the optical nonlinearities. Second, close to the Γ point, polaritons possess a negative effective mass and associated negative group velocity inherited from the PCS

mode, providing a potential platform for studying TMD-based polariton self-focusing and soliton formation, similar to what has been discussed theoretically and studied experimentally in other polariton systems^{14,29}. Third, the strong variation in the PCS mode linewidth in the vicinity of the BIC results in a wavevector-dependent Q -factor of both the LPB and UPB, enabling control of the polariton linewidth with the angle. These strongly modulated Q -factors, when combined with polariton–polariton interactions, can lead to novel phenomena such as the emergence of the so-called “weak lasing” state of matter³⁰ or the spontaneous formation of superfluid polariton currents³¹.

Further details of the optical response are revealed by TE-polarized angle-resolved PL spectra (e), showing emission from both polariton branches and the uncoupled neutral exciton (X^0). The latter is increasingly enhanced toward small wavevectors and exhibits a slight redshift of $\sim 1\ \text{meV}$, which we attribute to weak coupling to the higher frequency and broader $m = 3$ symmetric mode (see Supplementary Note 2 and Fig. S5). Charged exciton (trion, X^-) emission is also observed at $\hbar\omega_T = 1.62\ \text{meV}$ independent of k_x , implying a weak coupling.

We analyze the wavevector-dependent behavior of the LPB, UPB, uncoupled neutral exciton, and trion by fitting the PL spectra for each k_x with four Lorentzian functions $L_i(\omega) \propto ((\omega - \omega_i)^2 + (\gamma_i/2)^2)^{-1}$, as shown in Fig. 2f, and extracting the spectral position ω_i and linewidth γ_i as the full width at half maximum (FWHM) for each peak. The extracted parameters are plotted as functions of the in-plane wavevector in Fig. 3, with spectral positions (a) for the UPB (red symbols), LPB (blue symbols), and uncoupled neutral exciton (orange symbols), corresponding



values of the FWHM (b), and calculated Q -factors (c). The parameters for the uncoupled excitons extracted from the TM-polarized PL are plotted as green dots.

We then fit the extracted spectral positions of the UPB (ω_+) and LPB (ω_-) with a coupled oscillator model³², using the spectral position and homogeneous linewidth for the uncoupled neutral exciton $\tilde{\omega}_X = \omega_X + i\gamma_X$ and for the PCS/hBN mode $\tilde{\omega}_C(k_x) = \delta\omega_C + \omega_C(k_x) + i\gamma_C(k_x)$:

$$\omega_{\pm} = \text{Re} \left[\frac{\tilde{\omega}_C + \tilde{\omega}_X}{2} \pm \frac{1}{2} \sqrt{\hbar^2 \Omega_R^2 + (\tilde{\omega}_C - \tilde{\omega}_X)^2} \right],$$

$$\hbar \Omega_R = 2 \sqrt{\kappa^2 - \frac{(\gamma_C - \gamma_X)}{4}}.$$

Here, Ω_R is the Rabi splitting between the UPB and LPB, and the two fit parameters are the coupling strength κ and additional spectral shift $\delta\omega_C$ of the PCS mode²⁰ due to the presence of 1 L MoSe₂. The fit curves for the spectral positions of the UPB and LPB are shown in Fig. 3a by the red and blue solid lines, respectively. The uncoupled PCS/hBN mode, indicated by the black squares, comes into resonance with the uncoupled neutral exciton at $k_x^{\text{res}}/k \simeq \pm 0.014$, corresponding to an angle of $\theta^{\text{res}} \simeq \pm 0.8^\circ$. From the fits in Fig. 3a, we extract a coupling strength of $\kappa = 13.9$ meV, which corresponds to a Rabi splitting of $\Omega_R = 27.4$ meV and splitting-to-linewidth ratio of ~ 9 , exceeding the values recently reported for a WSe₂/PCS system²⁰ and theoretical estimates for strong coupling to an optical BIC²³. Because Ω_R is larger than the sum of the exciton (~ 9 meV) and PCS mode linewidth (~ 3 – 11 meV), the hybrid MoSe₂/hBN/PCS system is unambiguously in the strong coupling regime.

Quantitatively, the polariton linewidth $\gamma_{\pm}$ is expected to vary between that of the exciton (γ_X) and PCS modes (γ_C) depending on the excitonic fraction in the polariton. However, our experimentally observed values of the polariton linewidth (Fig. 3b, open symbols) close to resonance $k_x = k_x^{\text{res}}$ are significantly smaller than both γ_X (green) and γ_C (black). We attribute this to polariton motional narrowing, similar to the effects studied previously for quantum wells in microcavities^{33–37}.

Here, the large polariton mode size (tens of μm) together with the large Rabi splitting lead to effective averaging over excitonic disorder³⁸ in the 1 L MoSe₂ over a broad (nm– μm) range of length scales. As a result, the excitonic contribution to the polariton FWHM close to resonance is given by only the homogeneous exciton linewidth γ_X^h ³⁹, while away from resonance, where the polariton frequency overlaps with the exciton peak, it changes toward the inhomogeneous linewidth γ_X^{inh} due to an increasing interaction with disorder and associated scattering with higher momenta excitonic states as well as absorption^{36,40}. We use a phenomenological model that

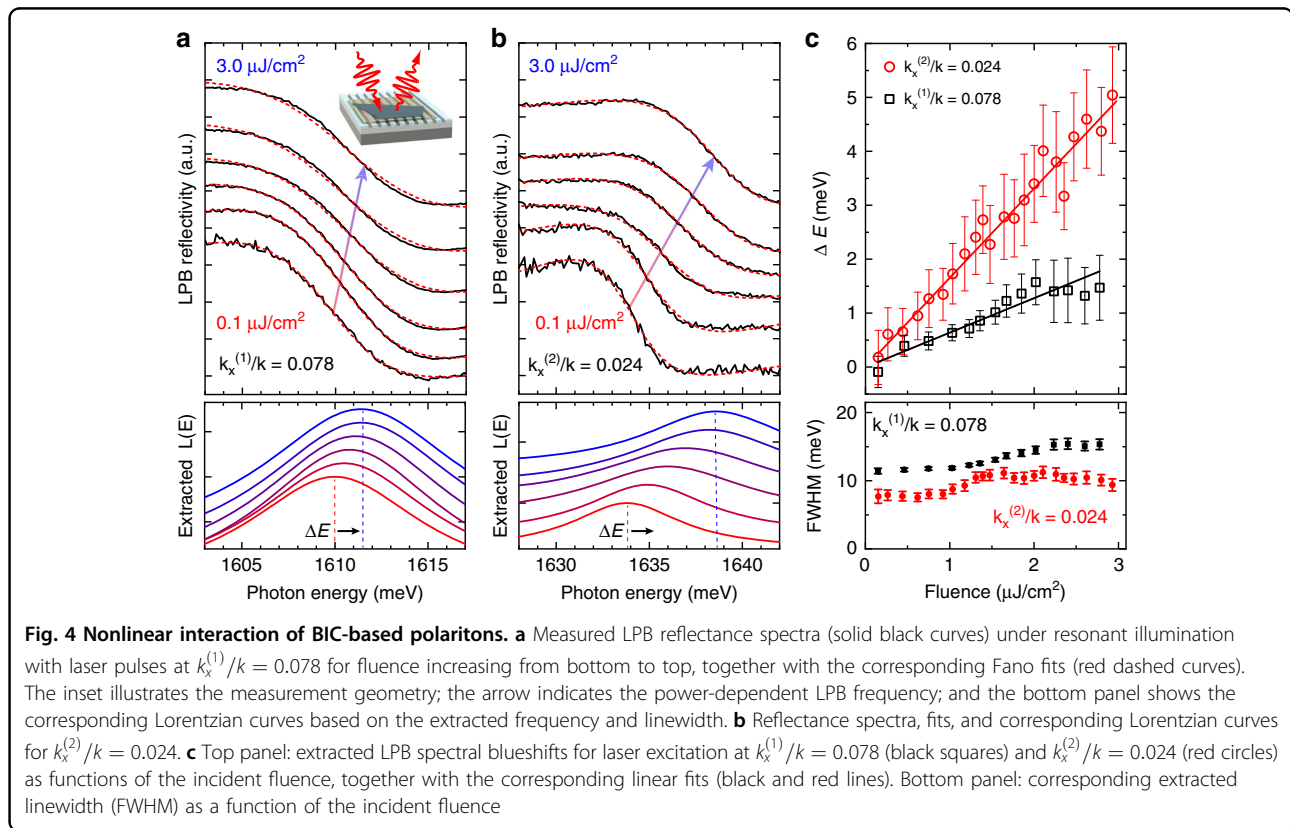
accounts for homogeneous and inhomogeneous contributions to the polariton linewidth (see Supplementary Note 3). The model shows good agreement with the experimental data (Fig. 3b, blue curve) for a homogeneous linewidth of $\gamma_X^h \sim 1$ meV (b, black dashed line), which is within the range of recently reported values^{41–44} for the low-temperature radiative decay rate of excitons in monolayer MoSe₂.

As a result of excitonic and photonic disorder averaging, the Q -factors achieved for polaritons around the Γ point in our structure are ~ 2 times higher than those for the bare PCS mode (Fig. 3c), reaching $Q \sim 900$. These higher Q -factors offer the potential for further improvement of the polariton linewidth through the fabrication of macroscopic photonic crystal samples³ with improved surface quality and the use of large TMD flakes grown by chemical vapor deposition. In addition, the strongly k -dependent Q -factor of the PCS mode in the vicinity of the BIC enables precise control of the polariton linewidth and corresponding Q -factors via angle or temperature tuning (see Supplementary Note 4 and Figs. S7 and S8).

Mixing photonic modes with excitons in our hybrid MoSe₂/hBN/PCS structures leads to a dramatic enhancement of the associated optical nonlinearities. We probe the underlying polariton–polariton interaction due to the excitonic contribution by measuring the pump-dependent frequency shifts of the polariton peaks in the reflectivity spectra. The polariton modes are excited resonantly in both the frequency and wavevector domains by ~ 130 fs laser pulses (Fig. 4a, inset), with the incident fluence varying from $0.1 \mu\text{J}/\text{cm}^2$ to $3.0 \mu\text{J}/\text{cm}^2$ (see “Methods”).

Figures 4a, b show the measured pump-dependent reflectivity spectra of the LPB resonance (solid black curves) for selected values of the incident fluence, increasing from bottom to top, and for two different x -components of the wavevector: $k_x^{(1)}/k = 0.078$ (a) and $k_x^{(2)}/k = 0.024$ (b). Due to the exciton–exciton interaction, which increases with the density of the created quasiparticles⁴⁵, the lower-energy polariton resonance in the reflectivity spectra is continuously shifted with the fluence toward higher energies, as clearly seen from the Lorentzian curves $L(E)$ based on the resonance frequency and linewidth extracted from the fits (Fig. 4a, b, bottom panels). We observe larger blueshifts (b) for wavevectors closer to the anticrossing condition $k_x^{\text{res}}/k = 0.014$, as expected for stronger polariton–polariton interactions associated with the increasing exciton fraction in the polariton.

The top panel in Fig. 4c shows the blueshift values for $k_x^{(1)}/k = 0.078$ (black squares) and $k_x^{(2)}/k = 0.024$ (red circles) extracted from the Fano line shape fitting (a, b, red dashed curves) for varying fluence, together with the corresponding linear fits (black and red lines). Calculating the polariton density n_p for each fluence (see



Supplementary Note 5), we obtain the polariton–polariton interaction strength $g_p = dE_p/dn_p$ of $g_p(k_x^{(1)}) \sim 0.04 \mu\text{eV} \mu\text{m}^2$ and $g_p(k_x^{(2)}) \sim 0.16 \mu\text{eV} \mu\text{m}^2$. Furthermore, from the $g_p(k_x) \propto g_X |X(k_x)|^4$ dependence on the Hopfield coefficient $X(k_x)$, which describes the exciton fraction in the polariton, we estimate the exciton–exciton interaction strength in our measurement to be $g_X = 1.0 \pm 0.4 \mu\text{eV} \mu\text{m}^2$. This value is on the same order as the theoretical estimate $g_X \sim 1.6 \mu\text{eV} \mu\text{m}^2$, as well as the value $g_X \sim 1.4 \mu\text{eV} \mu\text{m}^2$ we extract from a direct measurement of the pump-dependent excitonic blueshifts in TM polarization (see Supplementary Note 6 and Fig. S9).

While it is difficult to directly compare our g_X values with those observed in III–V materials due to the few orders of magnitude difference in the reported numbers, our interaction strength is not much lower than the values of $g_X \simeq 10 \mu\text{eV} \mu\text{m}^2$ recently extracted from careful measurements in GaAs-based polaritonic systems²⁵. On the other hand, our nonlinearities are considerably larger than those reported previously for WS₂-based polaritons²², where the estimation of the exciton–exciton interaction strength could possibly be uncertain due to efficient local heating at 300 K and the use of high excitation densities leading to higher-order interaction effects and associated redshifts.

The exciton densities in our experiment ($\leq 10^{12} \text{ cm}^{-2}$) are far below the Mott transition density ($\sim 10^{14} \text{ cm}^{-2}$),

and the observed polariton nonlinearity is mostly due to the exciton–exciton interaction, with phase space filling effects likely playing only a minor role. As seen from the fluence-dependent linewidth plots in the bottom panel of Fig. 4c, the increased interaction at higher densities also leads to a faster polariton decay, manifested as power-dependent broadening. In addition, we note that the observed nonlinearities are fast at least on a 100 fs scale, providing future opportunities for developing polariton-based ultrafast modulators and switches.

In summary, we present the first experimental demonstration and investigation of optical BIC-based polaritonic excitations. The formation of BIC-like polaritons in a hybrid system of a monolayer semiconductor interfaced with a PCS, with suppressed radiation into the far field and line narrowing due to effective disorder averaging, extends the polariton–polariton interaction time, which enhances the nonlinear optical response. In the future, these “dark” states can be accessed through near fields using guided modes excited by grating coupling or by nonlinear frequency conversion. With the strength of the underlying exciton–exciton interaction $g_X \sim 1.0 \mu\text{eV} \mu\text{m}^2$, our polaritons exhibit strong exciton-fraction-dependent optical nonlinearities that are fast on a 100 fs time scale. In addition, the planar geometry of our structure allows straightforward fabrication of the electrical contacts for electrostatic control of the polaritons and associated

interactions, while the use of atomically thin semiconductors in principle allows room-temperature operation. Thus, the formation of BIC-based polaritons can enable not only significantly enhanced but also controllable and fast nonlinear optical responses in photonic crystal systems due to the strong excitonic interaction in monolayer semiconductors and can open a new way to develop active and nonlinear all-optical on-chip devices.

Methods

Sample fabrication

Ta₂O₅ layers of 90 nm thickness were deposited on commercial SiO₂/Si substrates via e-beam assisted ion-beam sputtering. PCSs were fabricated by patterning the Ta₂O₅ layers via a combination of electron-beam lithography and plasma etching to yield the following geometric parameters: pitch $p = 500$ nm, groove width $w = 220$ nm, and depth $d = 90$ nm, as characterized by scanning electron and atomic force microscopy measurements. Large-area high-quality flakes of multilayer hBN and monolayer MoSe₂ were mechanically exfoliated from commercial bulk crystals (HQ Graphene) and stacked vertically onto the photonic crystal sample surface via dry transfer to form a hybrid 1 L MoSe₂/hBN/PCS structure.

Optical measurements

Angle-resolved reflectance spectroscopy was performed in a back-focal-plane setup with a slit spectrometer coupled to a liquid-nitrogen-cooled imaging CCD camera (Princeton Instruments SP2500+PyLoN), using white light from a halogen lamp for illumination (see Supplementary Fig. S1). For pump-dependent reflectivity measurements, the sample was excited by 130 fs pulses from a wavelength-tunable Ti:sapphire oscillator (Spectra-Physics, Tsunami, 80 MHz repetition rate) with wavevector control via laser beam positioning within the back focal plane of the objective. A single-slit optical chopper with a duty cycle of 0.001 was used in the laser beam to avoid sample heating. Angle-resolved PL measurements were performed in the same setup with off-resonant excitation by monochromatic light from a HeNe laser with a wavelength $\lambda_{exc} = 632.8$ nm. The sample was mounted in an ultra-low-vibration closed-cycle helium cryostat (Advanced Research Systems) and maintained at a controllable temperature in the range of 7–300 K. The cryostat was mounted onto a precise xyz stage for sample positioning. Spatial filtering in the detection channel was used to selectively measure signals from the 1 L MoSe₂/hBN/PCS sample area.

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Data availability

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

Conflict of interest

The authors declare that they have no conflict of interest.

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