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Imaging propagative exciton polaritons in atomically-thin WSe₂ waveguides

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Abstract

Exciton polariton (EP) is a half-light and half-matter quasiparticle promising for exploring both fundamental quantum phenomena as well as photonic applications. Van der Waals materials, such as transition-metal dichalcogenide (TMD), emerge as a new nanophotonics platform due to its support of long propagative EPs even at room temperature. However, real-space studies have been limited to bulk crystal waveguides with a thickness no less than 60 nm. Here we report the nano-optical imaging of the transverse-electric EPs in WSe₂ nanoflakes down to a few atomic layers, which can be turned on and off by tuning excitation laser polarization. Unlike previously-studied transverse-magnetic modes that exist only in bulk TMD waveguides, we found that the transverse-electric EPs could reside in ultrathin WSe₂ samples, owing to the alignment of the electric field with the in-plane dipole orientation of 2D excitons. Furthermore, we show that the EP wavelength and propagation length can be largely controlled by varying laser energy and sample thickness. These findings open opportunities to realize near-infrared polaritonic devices and circuits truly at the atomically-thin limit.

Main Text

Polaritons, which are hybrid light-matter quasiparticles, have been widely explored in two-dimensional (2D) and van der Waals materials in recent years [1-3]. For example, graphene was discovered to support electrically tunable Dirac plasmons with high confinement, long lifetime, broad spectral range, and a strong sensitivity to molecular vibrational modes [4-15]. Hexagonal boron nitride [16-19] and α -phase molybdenum trioxide [20,21] nanoflakes were found to support hyperbolic phonon polaritons. Hybrid plasmon-phonon polaritons have been observed in black phosphorus thin flakes upon ultrafast photoexcitation [22]. These spatially-confined nanophotonic modes provide a convenient way for manipulations of electromagnetic waves in the nanometer length scale.

Despite their unique properties, polaritons discussed above are mainly in the terahertz to the mid-IR region, far away from the near-IR to visible frequencies where fiber-based modern photonics technologies prevail. Recently, exciton polaritons (EPs), formed due to the strong coupling between photons and excitons [23-26], have been observed in group VIB transition-metal dichalcogenides (TMDs) [27-36]. These EPs residing the near-IR to visible regime are stable at room temperature due to the tightly bound excitons. They have also been demonstrated to have relatively long propagation lengths even at room temperature [35,36]. Such a combination is appealing for developing practical polaritonic devices. So far, propagative EPs have only been observed in bulk TMD waveguides, far away from the desirable 2D regime, where excitons have stronger binding energy and are more amenable to external controls (e.g. electrical gating and interface engineering).

In this rapid communication, we report the observation of EP transport in ultrathin WSe₂ waveguides down to a few atomic layers, imaging via the scattering-type scanning near-field optical microscope (s-SNOM) excited by a continuous-wave Ti:Al₂O₃ laser (see Supplemental Material [37]). Unlike common s-SNOM works where the incident laser is p polarized (namely electric field in the incident plane), EPs observed here are excited by an s-polarized laser beam (namely electric field perpendicular to the incident plane) [Fig. 1(b)]. To demonstrate that, we present in Fig. 1(c) polarization-dependent s-SNOM images of a thin WSe₂ flake with a thickness of about 9 nm (~ 13 atomic layers). In all the s-SNOM images presented in the paper, we plot the scattering near-field amplitude (s) that measures the electric-field amplitude underneath the tip. The sample edge is along the y direction, perpendicular to the incident laser beam. The laser energy is set to be at E=1.44 eV, lower than the A exciton energy ($E_A \approx 1.6$ eV). From the top to the bottom panels of Fig. 1(c), we tune the incident laser polarization from s to p step by step by rotating a half-wave plate. The key features in the top panels of Fig. 1(c) (close to the *s* polarization) are the bright fringes parallel to the sample edge (marked by the white dashed line). These fringes have the same period (ρ) in all top panels and are completely turned off when switching to the *p* polarization (bottom panel).

The observed bright fringes are generated due to the interference from photons collected from two paths [labeled as 'P1' and 'P2' in Fig. 1(b)], where 'P2' involves in-plane propagative modes [Fig. 1(b)] [35,36]. These tip-launched in-plane modes propagate along the sample and then get scattered into free-space photons at the sample edge, which interferes with photons directly scattered by the tip, thus forming interference fringes. Unlike thicker samples (≥ 60 nm) in previous studies [35,36], the 9-nm-thick WSe₂ sample studied here only show fringes when there is an *s*-polarized excitation field component [top panels of Fig. 1(c)]. In the case of pure *p*polarized excitation [bottom panel of Fig. 1(c)], no fringes are observed. This implies that the observed fringes are solely due to transverse electric (TE) modes, namely the electric field is along the *y* direction and perpendicular to the propagation direction of the mode.

Based on the origin of the fringe formation described above, we performed a quantitative analysis of the TE mode to extract the mode wavelength (λ_{ep}). By evaluating the phase difference between photon paths 'P1' and 'P2', the fringe period (ρ) of the measured fringes in the current experimental configuration [see Fig. 1(b)] has a simple relationship with λ_{ep} (see Supplemental Material [37]):

$$\lambda_0 / \rho \equiv k_\rho / k_0 \approx \lambda_0 / \lambda_{ep} - \cos \alpha, \tag{1}$$

where λ_0 is the excitation laser wavelength, $k_{\rho} = 2\pi/\rho$ is the inverse fringe period, and $\alpha \approx 30^{\circ}$ is the incident angle of the laser beam relative to the sample plane (*x*-*y* plane). The fringe period in

Fig. 1(c) is about 1.63 μ m, so the wavelength of these in-plane modes is about 617 nm at E = 1.44 eV according to Eq. (1).

To further confirm the TE nature of the observed modes, we performed finite-element simulations of a 9-nm-thick WSe₂ thin layer with COMSOL Multiphysics. In the modeling, we used y- and z-directional dipoles to simulate the s- and p-polarized tips, which can excite respectively the TE and TM modes in the device. In Figs. 1(d) and 1(e), we plot the y- or zdirectional electric field components (E_v and E_z) on the sample surface to reveal the TE and TM modes. The sample was set to be a highly-symmetric circular shape in the model for efficient computation. Unlike the *p*-polarized tip that can be treated approximately as a point dipole, the *s*polarized tip behaves like an extended dipole (see Supplemental Material [37]). In Fig. 1(d), one can see that the TE mode with a wavelength of about 620 nm (marked by a red arrow) propagating away from the dipole, which is consistent with the experimental result discussed above. In addition, we found that the TE mode pattern shows clear anisotropy: the TE mode favors propagating along the x direction over the y direction. This is expected for an extended inplane dipole, which excites approximately a plane-wave mode close to the source and only shows significant radial decay when it is far away [Fig. 1(d)]. In the case of p_z dipole excitation [Fig. 1(e)], there is no clear mode forming inside the 9-nm-thick WSe₂ sample, which is consistent with our experimental findings [Fig. 1(c)].

Following the polarization-dependence study, we wish to examine the polariton nature of the observed TE mode. In Fig. 2, we present s-SNOM images and line profiles taken with different excitation laser energy E. The line profiles [Fig. 2(b)] were extracted directly from the images in Fig. 2(a). Here the peaks in the line profiles correspond to the bright fringes in the data images. From both the images and profiles, one can see that fringe period ρ evolves systematically with E. With Fourier analysis, we can determine the inverse fringe period k_{ρ} = $2\pi/\rho$ [marked by arrows in Fig. 3(a)], and hence $\lambda_0/\rho \equiv k_\rho/k_0$, where $k_0 = 2\pi/\lambda_0$ is the free-space photon wavevector. We can then obtain λ_0/λ_{ep} that equals to $\lambda_0/\rho + 0.866$ based on Eq. (1). Note that λ_0/λ_{ep} is equal to the normalized in-plane mode wavevector k_{ep}/k_0 , where $k_{ep} = 2\pi/\lambda_{ep}$. Based on the Fourier transform results in Fig. 3(a), we obtained the $(k_{ep}/k_0, E)$ data points that describe the dispersion relation of the measured TE mode. As shown in Fig. 3(b), these data points are consistent with the calculated dispersion relation revealed in the dispersion colormap. In the colormap, we plot the imaginary part of the s-polarized reflection coefficient, which reflects the TE photonic density of states (DOS) (see Supplemental Material [37]). Both the experimental and theoretical dispersion relations in Fig. 3(b) reveals a clear mode back-bending feature (marked by a blue dashed curve) as E goes through the exciton energy E_A . In comparison, the pure TE photonic mode of the bare SiO₂/Si substrate appears to be a straight line (Fig. S2). The observed back-bending dispersion is the hallmark of polaritons measured by fix-energy measurements (e.g. imaging or angular scans) [35,38,39]. Anti-crossing polariton dispersion, on the other hand, is obtained mainly through fix-momentum spectroscopy measurements [24-34].

In addition to the fringe period, the fringe decay also shows a sensitive dependence on the excitation energy *E*. Clearly, the fringes extend further into the sample interior at lower excitation energies, indicating longer propagation length $L_{ep} \equiv 1/[2Im(k_{ep})]$ of the EPs. For quantitative analysis, we plot in Fig. 4(a) the extracted L_{ep} of the EPs by fitting the decay trend of the fringe profiles in Fig. 2(b) (see Fig. S9 in the Supplemental Material [37]). In Fig. 4(a), we also plot the calculated L_{ep} from COMSOL simulations (red curve), which agrees well with experimental data points. Representative COMSOL simulation maps are shown in Fig. S5 of the Supplemental Material [37], where EPs are propagating from the left to the right as plane waves.

From Fig. 4(a), one can see that L_{ep} drops rapidly as *E* gets closer to or above the A exciton energy E_{A} .

The increased damping of EPs at higher *E* is mainly due to the optical absorption by the WSe₂ sample layer, which is directly linked to the imaginary part of the in-plane dielectric function $\varepsilon_2(E)$. Indeed, our simulations prove that L_{ep} scales monotonically with $1/\varepsilon_2$ (Fig. S11 in the Supplementary Material [37]). For further discussions, we plot in Fig. 4(b) energy-dependent ε_2 data (grey curve) adopted from Ref. 40. In the same plot, we decompose ε_2 into different contributions: A, B and C excitons, and the background absorption continuum mainly due to strong interband transitions peaked at around 3 eV [40]. The A, B and C exciton resonances are constructed with standard Lorentzian oscillators peaked at about 1.60 eV, 2.16 eV and 2.57 eV, respectively. The background absorption continuum is obtained by subtracting the exciton resonances from the ε_2 data [41].

From Fig. 4(b), one can see that A exciton broadening is responsible for the EP damping below the A exciton energy $E_A \approx 1.6$ eV. According to previous studies [42,43], exciton broadening is mainly due to the scattering of excitons with both longitudinal optical photons and acoustic phonons. As *E* increases above E_A , background continuum mainly due to interband transitions is the dominant damping source with additional contribution from the B excitons. Therefore, the lower energy polariton branch ($E < E_A$) is preferred for practical applications related to EP transport. As discussed below, the other key damping mechanism of the EPs in the current WSe₂ devices is radiative decay into the silicon substrate that shows a clear dependence on WSe₂ thickness.

Finally, we found that TE EPs can be tailored by varying the sample thickness. In Fig. 5(a), we plot the *s*-polarized near-field amplitude images of WSe₂ with various thicknesses. The laser energy was set to be E = 1.38 eV below the exciton resonance and the sample thickness varies from 33 to 3 nm. The corresponding line profiles taken perpendicular to the sample edge are shown in Fig. 5(b). From both the s-SNOM images and profiles, one can see that the fringe period ρ increases systematically with decreasing sample thickness. Based on the fringe period data measured from Figs. 5(a) and 5(b), we extracted the EP wavelength λ_{ep} with Eq. (1). The thickness-dependent λ_{ep} is shown in Fig. 5(c) (squares), where λ_{ep} varies from 440 nm to 770 nm for sample thicknesses from 33 nm to 3 nm. Stronger mode confinement and higher-order modes could be seen in even thicker samples (see Fig. S3 in the Supplemental Material [37]). In Fig. 5(c), we also plot the calculated λ_{ep} (red curve) based on COMSOL simulations (see Fig. S5 in the Supplemental Material [37]), which is consistent with experimental data points.

Figures 5(a) and 5(b) also show a clear dependence of the propagation length L_{ep} of EPs on sample thickness. By fitting the decaying trend of polariton fringes in Fig. 5(b) (see Fig. S10 in the Supplemental Material [37]), we were able to extract the thickness dependent L_{ep} [black squares in Fig. 5(d)] at E = 1.38 eV, which is consistent with the calculated L_{ep} [red curve in Fig. 5(d)] based on COMSOL simulations (Fig. S5). From Fig. 5(d), one can see that L_{ep} reaches over 10 µm in samples thicker than 30 nm in the current sample geometry but drops rapidly to less than 2 µm in WSe₂ atomic layers. The main damping mechanism here in the atomic-layer samples is due to radiative decay into the silicon substrate. As shown in Fig. S5(b) in the Supplemental Material [37], the EP mode is better confined to the sample layer (marked by black arrows) in thicker WSe₂ samples due to the smaller λ_{ep} , so radiative decay into silicon is weaker. As for thinner samples, less confined EPs leads to stronger decay into silicon. To suppress the radiative decay, replacing silicon with noble metals is a good solution. As demonstrated in Figs. 4(a) and 5(d), if replacing silicon with silver (Ag), L_{ep} is orders of magnitude higher and could

reach 100 μ m, sufficient for general nanophotonic device applications. Note that using silver substrate instead of silicon has negligible modifications on the EP mode pattern (see Fig. S6 in the Supplemental Material [37]) and thus the EP wavelength [Fig. 5(c)].

By applying s-SNOM under s-polarized laser excitations, we imaged TE EPs in ultra-thin WSe₂ nanoflakes. We found that the TE modes couple strongly with excitons due to the in-plane orientation of the electric field. As a result, TE EPs are observed in WSe₂ samples down to a few atomic layers and can be turned on and off via laser polarization control. In addition, we showed that the polariton wavelength varies dramatically with sample thickness, which points to an effective means to tailor the EPs in TMDs. Furthermore, we found that TE EPs in WSe₂ can propagate over many microns below the A exciton energy. Implementation of a metal substrate that shields radiative decay could in principle increases L_{ep} to over 100 µm at ambient conditions. With rigorous analysis and quantitative simulations, we uncover fully the nano-optical physics related to the observed real-space characteristics of the transverse-electric EPs in TMD atomic layers and sheds light on applications of WSe₂ in polaritonic transistors and modulators in the near-infrared telecommunication bands.

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Figure captions

FIG. 1. (a) Illustration of tip excitation of TE EPs in WSe₂. (b) Sketch of the experimental setup, sample geometry, and signal collection. (c) Polarization-dependent s-SNOM images of a 9-nm-thick WSe₂ sample taken at an excitation energy of E = 1.44 eV. The white dashed lines mark the edge of the sample. From top to the bottom, we tune the laser polarization step by step from *s* to *p*. (d,e) COMSOL simulation of transverse electric (TE) and transverse magnetic (TM) modes on the sample surface, respectively. The *y*- (*p_y*) or *z*-directional (*p_z*) dipoles used to excite the TE or TM modes are marked red at the sample center. The double-sided arrow in (d) marks the TE mode wavelength. Scale bars: 1 µm.

FIG. 2. (a) Near-field amplitude images of a 9-nm-thick WSe_2 flake at various excitation energies. The white dashed lines mark the edge of the sample. Scale bar: 2 µm. (b) Energy-dependent line profiles taken perpendicular to the sample edge from the data images in (a).

FIG. 3. (a) Fourier transform (FT) of the fringe profiles in Fig. 2(b). The blue arrows mark the resonance peak due to the EP mode. Additional peaks at small k_{ρ} are due to far-field photons. (b) Dispersion diagram of the EPs in the 9-nm-thick WSe₂ sample from both experiment (squares) and calculation (colormap). The colormap plots the photonic density of states (DOS). The blue dashed curve is a guide to the eye. Note that the unit for the wavevector k is the free-space photon wavevector k_0 .

FIG 4. (a) Energy-dependent propagation length of the EPs (L_{ep}) from both experiment (squares) and simulation (red solid and blue dashed curves for two types of devices, see discussions in the main text). (b) The imaginary part of the dielectric function of WSe₂ (ε_2 , grey curve) from Beal et al. [40]. The color dashed curves sketch separately the contributions to ε_2 from A (red), B (blue), and C (orange) excitons, and the background absorption continuum (green).

FIG. 5. (a,b) The near-field amplitude images and fringe profiles of the EPs in WSe₂ thin layers with various thicknesses. The vertical dashed lines in (a) mark the edge of the sample. Scale bar: 1 μ m. (c,d) Experimental (squares) and calculated (red solid and blue dashed curves for two types of devices, see discussions in the main text) EP wavelength λ_{ep} and propagation length L_{ep} with various WSe₂ thicknesses. In all panels, the excitation energy is E = 1.38 eV.