Spatiotemporally controlled room-temperature exciton transport under dynamic strain

Kanak Datta¹, Zhengyang Lyu², Zidong Li¹, Takashi Taniguchi³, Kenji Watanabe⁴ and Parag B. Deotare*¹

Two-dimensional transition metal dichalcogenides provide an attractive platform for studying strain-dependent exciton transport at room temperature due to large exciton binding energy and strong bandgap sensitivity to mechanical stimuli. Here we use Rayleigh-type surface acoustic waves to demonstrate controlled and directional exciton transport under the weak coupling regime at room temperature. We screen the in-plane piezoelectric field using photogenerated carriers to study transport under type-I bandgap modulation and measure a maximum exciton drift velocity of 600 m s⁻¹. Furthermore, we demonstrate the precise steering of exciton flux by controlling the relative phase between the input RF excitation and exciton photogeneration. The results provide an important insight into the weak coupling regime between the dynamic strain wave and room-temperature excitons in a two-dimensional semiconductor system and pave the way to exciting applications of excitonic devices ranging from data communication and processing to sensing and energy conversion.

Coulombically bound electron–hole pairs (commonly known as excitons) provide an effective platform to transport energy at the nanoscale. Coupled with small dimension and seamless transition with photons, excitons have the potential to serve various applications in energy conversion¹, light emission¹, chemical sensing¹, and information processing and communication¹⁻⁵. With the recent emergence of two-dimensional semiconductors such as transition metal dichalcogenides (TMDs) that support excitons with high diffusivity and binding energy (>100 meV)⁶⁻⁸, the feasibility of room-temperature excitonic devices is no longer questionable⁹. However, the spatial manipulation of exciton flux that is critical to control energy flow at the nanoscale remains a challenge, especially at room temperature. Unlike charged particles that drift under an externally applied electric field, the directed transport of charge-neutral exciton flux is achieved by the spatial tuning of exciton potential by external stimuli such as mechanical strain¹⁰ or electric field¹¹⁻¹⁻⁵. Travelling surface acoustic waves (SAWs) can dynamically utilize both effects to achieve long-range transport. Although most of the reported SAW-assisted transport is based on transporting individual charges following exciton dissociation by a piezoelectric field, exciton transport under a dynamic strain from SAWs has also been achieved in a III–V quantum well system¹⁶⁻¹⁸. However, it is limited to cryogenic temperatures and utilizes indirect excitons that are created using an external electrical field. With large sensitivity of the bandgap to external strain⁹,¹¹, TMDs are well suited to achieve room-temperature directional transport of direct excitons solely under dynamic strain. In monolayer materials, SAWs have been successfully utilized for long-range carrier transport¹¹,¹²,¹³, improved photoconductivity¹³, modulation of phonons¹⁴ and exciton photoluminescence (PL)¹⁵,¹⁶. In this work, we study the spatiotemporal control of exciton flux in a monolayer tungsten diselenide (WSe₂) system at room temperature. High-frequency (resonance frequency, ~745 MHz) Rayleigh-type SAWs are generated in a piezoelectric 128° Y-cut lithium niobate (LiNbO₃) substrate using interdigitated electrodes (IDTs) (Supplementary Section 1 provides the acoustic response of the device). Mechanically exfoliated monolayer WSe₂ encapsulated in hexagonal boron nitride (hBN) was transferred on the SAW delay line using a dry transfer technique. Using phase-synchronized spatiotemporal measurements and utilizing photogenerated free carriers to screen the in-plane electric field, we report directed exciton transport under type-I bandgap modulation. Based on the experiments, we extract a drift velocity of 600 m s⁻¹ for a strain amplitude of ~0.086% and estimate the neutral exciton mobility to be about 900 cm² (eV s)⁻¹. In addition, we demonstrate precise manipulation over the exciton transport by controlling the phase delay between radio-frequency (RF) excitation and photexcitation. This work also provides important insights into the weak coupling regime between the dynamic strain wave and room-temperature excitons in a two-dimensional semiconductor system, leading to a potential pathway for long-range exciton transport at room temperature.

Results and discussion

Figure 1a shows a schematic of the device geometry used in this work. Figure 1b,c show a false-colour brightfield optical image of the sample and the PL map of the hBN-encapsulated monolayer WSe₂. Here hBN encapsulation is critical to improve the transport properties of the excitons by suppressing non-radiative recombination processes¹⁷, surface roughness¹⁸, energetic disorder¹⁹, and scattering from impurities and surface states²⁰. More importantly, the underlying bulk hBN moderates the dielectric environment surrounding the monolayer, thereby increasing the exciton binding energy compared with monolayer directly placed on a LiNbO₃ substrate. This reduces exciton dissociation under a SAW piezoelectric field (type-II modulation) due to increased binding energy²⁰,²¹. The dissociation can be further reduced by screening the piezoelectric field, thereby enabling the study of excitonic interactions with type-I modulation (bandgap change due to strain). In this

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work, we achieve it by utilizing the optically generated free carriers to screen the in-plane electric field of the travelling SAW wave. Figure 1d plots the PL quenching factor $r_Q = \frac{I_{PL, RF}}{I_{PL, -60dBm}}$ due to dissociation as a function of RF input power for various optical excitations. Here $I_{PL, RF} = \int I_{RF}^o(\lambda) d\lambda$ and $I_{PL, -60dBm} = \int I_{-60dBm}(\lambda) d\lambda$ refer to the integrated PL intensity measured at a given RF input power and under –60 dBm RF power, respectively.

**Fig. 1** | Modulation of hBN-encapsulated monolayer WSe$_2$ PL by the piezoelectric field of the travelling SAW. a, Schematic of the transferred monolayer WSe$_2$ on the SAW delay line. b, Brightfield optical image of hBN-encapsulated monolayer WSe$_2$ transferred onto a LiNbO$_3$ substrate. c, Integrated PL intensity map of monolayer area represented by the square in b. d, PL intensity modulation under increased RF power at various optical excitation densities. The monolayer PL emission decreases with an increase in the RF power. The net PL quenching, however, reduces with increasing optical power density due to screening from the optically generated free carriers.

**Fig. 2** | Spatiotemporal modulation of exciton density in monolayer WSe$_2$ at room temperature under travelling wave. a,b, Spatiotemporal exciton density profile (log scale) from phase-synchronized TCSPC measurement for RF excitation inputs of –60 dBm (a) and 13 dBm (b). c,d, Spatiotemporal map of exciton density (normalized for each time instance along space) for RF input powers of –60 dBm (c) and 13 dBm (d) at an optical fluence of 1.2 $\mu$J cm$^{-2}$. A spatial shift in exciton density can be observed under RF excitation. e,f, Normalized exciton density profiles at different time instances as a function of space along with the Gaussian fit under RF input powers of –60 dBm (e) and 13 dBm (f). At the minimum RF input power (~60 dBm), the exciton density symmetrically broadens in space due to exciton diffusion in the monolayer. At a high RF input power, an asymmetric spatial shift in the exciton density in the direction of acoustic wave propagation is observed with the observed shift increasing with time.

To improve the signal-to-noise ratio, the raw data were binned using a 100 ps window.
Acoustic-wave-mediated exciton transport in monolayer WSe₂.

Figure 2 shows the results of excitonic energy transport measured using a scanning single photon single photon avalanche diode (SPAD) described in our previous work. Figure 2a,b shows the spatiotemporal exciton density distributions at RF input powers of −60 dBm (−1 nW) and 13 dBm (−20 mW), respectively, at an optical fluence of 1.2 μJ cm⁻² (type-I modulation dominates since PL quenching was below ~2% under the experimental conditions). Figure 2c,d shows the exciton density distribution normalized along space at each time instance corresponding to the data in Fig. 2a,b, respectively. The symmetric exciton density distribution at −60 dBm resembles typical anomalous exciton diffusion in monolayer WSe₂.

The black arrow indicates the direction of propagation.

Figure 3a shows the spatial evolution of the Gaussian exciton density peak as a function of time. With increasing RF power, we see a gradual increase in the spatial shift. In addition, we observe periodic oscillations corresponding to the SAW period. This suggests weak coupling between the excitons and travelling strain wave (discussed later), where the exciton drift velocity is insufficient to keep up with the travelling wave and hence results in a net spatial shift in the exciton density over time. We model the evolution of the Gaussian peak under dynamic strain over time using a linear relationship (Fig. 3a, solid line):

\[ h(t) = v_{avg}t + h_{offset}. \]  

where \( h(t) \) refers to the net displacement of the generated exciton density over many SAW periods, \( v_{avg} \) refers to the average drift velocity of the exciton density under the applied dynamic strain and \( h_{offset} \) is a constant to accommodate the fitting error. Figure 3b plots the \( v_{avg} \) for various volumetric strains estimated at different RF input powers. A linear trend with volumetric strain indicates a proportional increase in exciton coupling efficiency with the dynamic strain field. Further improvement in \( v_{avg} \) can be achieved by increasing the strain gradient or increasing the exciton diffusivity by suppressing scattering from impurity states and surface roughness.

We determine the extent of exciton coupling to the dynamically varying strain field of the SAW by estimating the instantaneous velocity from the measured instantaneous displacement of the exciton density. The time derivative of the measured displacement at RF input power of 13 dBm gives the instantaneous velocity of the exciton flux (Fig. 3c). The instantaneous drift velocity reaches a maximum value of 600 m s⁻¹, which is smaller than the SAW velocity \( v_{SAW} \) in LiNbO₃ (3,979 m s⁻¹ (ref. 33)). Since the maximum drift velocity of the exciton flux is nearly six times smaller than the acoustic wave velocity \( v_{SAW} \), the exciton flux cannot keep pace with the travelling strain wave (weak coupling regime). This results in asymmetric exciton funneling (in opposite directions) during each SAW period. Hence, we observe oscillations in the peak position of the exciton distribution and a net spatial shift over time in the direction of the travelling strain field (Figs. 2d and 3a). The observation closely resembles carrier drift under a dynamic electric field in the weak coupling regime presented elsewhere. Under strong coupling, the excitons would funnel to the lowest potential, followed by transport of the trapped excitons with the strain wave. In such a case, the transport distance is expected to be limited by the radiative lifetime and sample size. Such a regime of exciton transport has been observed in III–V semiconducting quantum wells at cryogenic temperatures.

Under a dynamically varying strain field, the maximum exciton drift velocity can be written as

\[ v_{max} = \mu \left| \frac{\partial E_g}{\partial \varepsilon} \right|_{\varepsilon_{max}} = \mu \left| \frac{\partial E_g}{\partial \varepsilon} \right| \varepsilon_0 k. \]  

where \( \mu \), \( \frac{\partial E_g}{\partial \varepsilon} \), \( k = \frac{2\pi}{\lambda} \) and \( \varepsilon_0 \) refer to the exciton mobility, strain sensitivity of the monolayer bandgap, acoustic wave momentum and maximum dynamic strain in the monolayer, respectively. Here
\( \lambda_{\text{SAW}} \) refers to the wavelength of the acoustic wave (\( \lambda_{\text{SAW}} = 4.7 \mu m \)). Based on the measured PL quenching (Fig. 1d), we calculate the maximum dynamic strain amplitude (\( \varepsilon_d \)) to be \( \pm 0.086 \% \) (tension) at RF power of 13 dBm using the converse piezoelectric matrix of 128° LiNbO3 and the estimated piezoelectric field in the substrate (Supplementary Section 12). Using the estimated value of the maximum drift velocity at RF input power of 13 dBm (600 m s\(^{-1}\)), which is about 15% of the SAW velocity in equation (2), we extract the exciton mobility in the monolayer to be 900 cm\(^2\) (eV s\(^{-1}\)) compared with indirect excitons in III–V quantum well structures at cryogenic temperatures that show long-range transport\(^{16,17}\). The small exciton mobility, resulting from scattering with defects and phonons at room temperature, along with the short radiative lifetime result in a smaller transport distance (approximately micrometres) under a dynamic strain in monolayer TMDs. However, the results show the potential of the material system for future room-temperature excitonic devices, especially as defect densities get lowered with rapid progress in material growth.

We note that the total bandgap modulation (\( \delta E_g = 2 \times 0.086 \times 60 = 10.3 \text{meV} \)) is smaller than the room-temperature thermal energy (25.7 meV). However, the observed directional transport primarily results from the combination of the strong bandgap sensitivity of TMD monolayers and the strain gradient generated by the SAW wave, which leads to drift dominating over diffusion transport at room temperature (Supplementary Section 6). At the same time, we assume a negligible decoupling of the dynamic strain to the WSe\(_2\) monolayer through the underlying hBN flakes due to the high mechanical strength\(^{16,18}\) and small thickness (20–30 nm; measured using white-light interferometry) compared with the acoustic wavelength (4.7 \( \mu m \)). Therefore, the extracted values of the average drift velocity set the lower limit at room temperature. We also expect negligible contributions to the observed transport from strain-induced change in the binding energy of neutral excitons as such modulations are substantially smaller compared with strain-induced bandgap modulation based on theoretical calculations\(^{26–41}\). Finally, we expect negligible contribution to the quasi-particle conversion (exciton to trion) under strain\(^{41,42}\) and type-II bandgap modulation\(^{43}\) due to the much lower strain amplitude (<0.1%) and screening due to the underlying hBN spacer.

Exciton transport under type-I modulation takes place by spatial trapping at the minimum energy locations\(^{43}\) (Fig. 4a–d) shows the schematic for the exciton potential under a dynamic strain field. We verified that the observed spatiotemporal modulation in the exciton density is due to type-I modulation by performing three experiments. First, we conduct transport measurements at an order lower optical fluence. Since the effective mobility of the photogenerated excitons increase with optical fluence\(^{44}\), the excitons experience efficient coupling with the travelling strain field at high optical excitation densities. For the same set of RF powers (constant strain), we observed reduced modulation in the exciton density and a decrease in average exciton spatial drift for lower optical fluence (Supplementary Section 7). Second, we measured the time-resolved photoluminescence (TRPL) as a function of optical fluence. At low fluence, the TRPL decays faster with an increase in RF power due to increased ionization\(^{45,46}\) (additional decay) resulting from type-II modulation. At high fluence, the reduction in the TRPL decay rate is slower (Supplementary Section 8), confirming the screening of the in-plane piezoelectric field by the free carriers. Under this condition, type-I modulation dominates and the TRPL oscillates at the SAW frequency. The strain-induced oscillations result from the dynamic modulation of the energy separation between the K and Q valleys under the travelling strain wave\(^{40}\) (Supplementary Section 8 provides the model describing the strain-induced oscillations). Finally, we verify type-I band-edge modulation by extracting the neutral-exciton linewidth broadening, which remains

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**Fig. 4 | Spatiotemporally controlled energy transport under phase-modulated optical excitation for 15 dBm SAW excitation.** a–d. Schematic of the photogenerated excitons at different SAW phases along with exciton potential: \( r (a) \); \( r + T/4 (b) \); \( r + T/2 (c) \); \( r + 3T/4 (d) \). The vertical blue line represents the position of the excitation pulse on the SAW wave. For \( r \), the excitons are generated at a position such that the energetically stable position lies in the opposite direction of SAW propagation. Therefore, immediately after photogeneration, the exciton density distribution shifts in the opposite direction of SAW propagation. However, as the excitons couple to the travelling strain field, a net drift in the direction of SAW was observed. For \( r + T/4 \), excitons are generated at the positions of the lowest bandgap (position of the maximum tensile strain corresponding to the minimum exciton potential but the lowest energy gradient). Hence, the exciton density distribution does not undergo an immediate spatial shift after photogeneration. At \( r + T/2 \), following photoexcitation, the energy gradient drives the exciton flux forward towards the stable position of the maximum tensile strain. Therefore, the generated density distribution moves immediately in the direction of SAW propagation. For \( r + 3T/4 \), the excitons are generated at the position of the maximum compressive strain, that is, the maximum bandgap, and hence results in no spatial shift immediately after photogeneration. e. Phase-synchronized evolution of the Gaussian peak position (circles) extracted by fitting the normalized exciton density measured using the TCSPC technique. The Gaussian peak position oscillates with the period of the SAW wave. A progressive delay in the Gaussian peak is also observed as the time delay is increased at an increment of T/4. Alongside the oscillations, a net drift in the Gaussian peak is observed in the direction of acoustic wave propagation. The solid line shows the numerical fit using the model discussed in Supplementary Section 13.
constant for varying optical excitation under RF excitation (Supplementary Section 9 provides the PL broadening data under type-I and type-II bandgap modulations). Further discussion on the effect of type-I and type-II band-edge modulation on exciton dynamics is provided in Supplementary Section 10. We also note that the observations have been reproduced on multiple samples with different RF resonance frequencies (Supplementary Section 11).

**Acoustic steering of photogenerated exciton density.** The dynamic acoustic strain field generated by a SAW wave can be represented by

$$\varepsilon(x,t) = \varepsilon_0 \cos(\omega t - kx + \phi),$$  

where $\varepsilon_0$ refers to the amplitude of the strain field, $\omega$ refers to the angular frequency ($\omega = 2\pi f_{\text{SAW}}$), $f_{\text{SAW}}$ refers to the acoustic resonance frequency) and $\phi$ refers to the instantaneous phase of the acoustic wave. Therefore, for a given optical excitation position from the IDT, the generated exciton density interacts with a certain phase of the travelling strain field. Using equation (3), the dynamic strain-induced drift velocity of the exciton flux can be written as:

$$v_{ex}(x,t) = \mu_e \frac{\partial \varepsilon(x,t)}{\partial x} = \mu_e k \sin(\omega t - kx + \phi),$$  

where $\mu_e = \mu \frac{\partial \varepsilon_0}{\partial x}$ refers to the strain mobility of the excitons. At photoexcitation ($t=0$), the instantaneous velocity of the exciton flux is a function of the acoustic phase, namely, $v_{ex}(x,0) = \mu_e k \sin(-kx + \phi)$. Therefore, precise control over the direction of photogenerated exciton flux can be achieved by controlling the relative phases (Fig. 4a–d). We achieve such control by introducing a delay $\tau$ ($\phi = \tau \omega / f_{SAW}$) in the laser trigger signal (Supplementary Section 6 provides a schematic of the characterization setup). Figure 4e plots the spatiotemporal evolution of the centre of the exciton density distribution as a function of time delay ($\tau$) with increments of $\tau T / 4$ (Supplementary Section 12 provides the corresponding spatiotemporal exciton density maps), which were further numerically modelled using the following modified drift–diffusion equation:

$$\frac{\partial n_{ex}(x,t)}{\partial t} = D_{ex} \frac{\partial^2 n_{ex}(x,t)}{\partial x^2} + \frac{\partial (v_{ex} n_{ex}(x,t))}{\partial x} + G - \frac{\tau_{rec}}{\tau_{rec}} - \frac{\tau_{ion}}{\tau_{ion}},$$  

where $n_{ex}$, $D_{ex}$, $v_{ex}$, $\tau_{rec}$ and $\tau_{ion}$ refer to the population, diffusion coefficient, velocity, recombination time and ionization time of the neutral exciton$^{15}$, respectively. Further, $G$ refers to the photo-generated exciton density under optical excitation. The solid lines in Fig. 4e are the fits from the numerical simulation that successfully capture the observed dynamics (Supplementary Section 13). Consistently, we observed similar progressive evolution with the instantaneous phase in the TRPL data (Supplementary Section 12).

In conclusion, we demonstrated room-temperature directed exciton energy transport in monolayer WSe$_2$ under a travelling strain field. The dynamic strain gradient obtained under the experimental condition was sufficient to study the weak coupling regime between the excitons and strain wave. Our results show that weak coupling leads to oscillations in the transported exciton density since the exciton drift velocity is lower than the velocity of the travelling wave. Based on the measurement, we estimate the neutral exciton mobility in the monolayer to be $900 \text{cm}^2/(\text{eV s})^{-1}$, which is in good agreement with the values reported in the literature for the same material. The results show that the coupling between excitons and dynamic strain wave in TMDs strongly depends on the transport properties and is therefore expected to be influenced by various factors like intrinsic and extrinsic defect states, lattice disorders, substrate roughness, and scattering from charged and neutral impurities. Hence, an improved material system with higher strain gradients would provide a pathway to reach the strong coupling regime. In addition, the use of TMD heterostructures that offer longer lifetimes could aid in improving the overall transport length. Finally, we also demonstrated acoustic steering over the photogenerated excitons by precisely tuning the exciton photogeneration with respect to the phase of the acoustic wave and provide a modified drift–diffusion model to describe the observations. The reported results pave the way for exciting future applications that include efficient energy conversion, sensing, detection, and room-temperature on-chip excitonic information processing and communication.

**Online content**

Any methods, additional references, Nature Research reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at https://doi.org/10.1038/s41566-021-00951-3.

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Methods

SAW device fabrication. The SAW devices were fabricated using a standard photolithography-based metal lift-off process on a 128° Y-cut LiNbO3 substrate. A bilayer photoresist stack was used (LOR 3A+ S1813) to achieve high-resolution IDT features. The IDTs were patterned using a projection lithography tool (GCA AS200 AutoStep). After the development of the exposed features, 10 nm Cr and 100 nm Au were evaporated using the electron-beam evaporation technique. The lift-off was performed by immersing the samples in Remover PG for 12 h. The second step of lithography involved the patterning of the contact pads, which was carried out using SPR 220 (3.0) as the photoresist layer. The same projection lithography tool was used to pattern the contact pads. After the development of the exposed contact-pad features, 10 nm Cr and 480 nm Au were evaporated using the electron-beam evaporation technique. The lift-off was performed following the same process as the first step. The samples were bonded to a custom-made printed circuit board using wire bonding. Detailed parameters for the overall process of device fabrication can be found in our previous work.

Exfoliation and transfer of monolayer WSe2. The hBN and WSe2 flakes were mechanically exfoliated from bulk crystals (bulk hBN crystal was received from T. T. and K. W.; bulk WSe2 was purchased from hq graphene) using Scotch tape. The top/bottom hBN and monolayer WSe2 were exfoliated from the Scotch tape onto a thin polylithiumsiloxane stamp (GEL-FILM WF-40/1.5-X4) and identified under an optical microscope. Heterostructures were stacked in a layer-by-layer manner using a polymer-assisted dry transfer technique under a home-built setup with micromanipulators and a rotational stage. No heat was applied during the transfer process. Consequently, we expect some residual polymer layer between the hBN flake and WSe2 monolayer.

Acoustic response measurement from the SAW devices. The acoustic characterization of the SAW devices was carried out using a network analyser (Agilent 4396B) connected to an S-parameter test unit (Agilent 85046A). The characterization of the SAW devices was carried out using a network analyser (Agilent 85046A). The authors declare no competing interests.

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Author contributions

P.B.D. conceived the idea and supervised the project. K.D. fabricated and characterized the devices. Z.Lyu transferred the encapsulated monolayers on the SAW devices. P.B.D., K.D. and Z.Li. analysed the data. Growth of hBN was done by T.T. and K.W. P.B.D., K.D. and Z.Li. contributed to writing the manuscript.

Competing interests

The authors declare no competing interests.

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