Long-range propagation of indirect excitons in van der Waals heterostructure: 1/e decay distance reaches ~ 100 microns

L. H. Fowler-Gerace,1 Zhiwen Zhou,1 E. A. Szwed,1 and L. V. Butov1
1Department of Physics, University of California at San Diego, La Jolla, CA 92093, USA
(Dated: April 20, 2022)

A long-range propagation of indirect excitons (IXs), also known as interlayer excitons, is observed in a van der Waals MoSe2/WSe2 heterostructure. The 1/e decay distance of IX luminescence reaches ~ 100 µm. The long-range IX propagation is realized using an optical excitation with the energy close to the MoSe2 or WSe2 direct exciton energy. The range of excitation powers and temperatures for the long-range IX propagation is determined.

PACS numbers:

INTRODUCTION

A spatially indirect exciton (IX) is a bound pair of an electron and a hole confined in separated layers [1]. Due to the spatial separation of electrons and holes, the lifetimes of IXs can exceed the lifetimes of spatially direct excitons (DXs) by orders of magnitude. The long lifetimes allow IXs to travel long distances before recombination. This gives an opportunity to explore exciton transport by imaging spectroscopy.

The long-range IX transport has been extensively studied in GaAs heterostructures where the 1/e decay distances of IX luminescence d1/e reach tens and hundreds of microns [2–22]. The long IX propagation distances in GaAs heterostructures allowed uncovering a number of exciton transport phenomena, including the inner ring in exciton emission patterns [5, 7], the exciton localization-delocalization transition in random [5], periodic [10] and moving [14] potentials, the transistor effect for excitons [9], the long-range coherent spin transport [15, 17, 21], and the dissipationless exciton transport over long times evidencing the exciton superfluidity [22].

Excitons exist at temperatures roughly below $E_X/k_B$ ($E_X$ is the exciton binding energy, $k_B$ the Boltzmann constant) [23]. IXs in GaAs heterostructures exist at low temperatures due to their low binding energies: $E_X$ is typically ~ 4 meV in GaAs/AlGaAs heterostructures [24] and ~ 10 meV in GaAs/AlAs heterostructures [25]. Furthermore, the temperature of quantum degeneracy, which can be achieved with increasing density before excitons dissociate to electron-hole plasma, scales proportionally to $E_X$ [26]. Therefore, material systems where IXs have high-binding energies can provide the medium for the realization of both high-temperature quantum phenomena and high-temperature excitonic devices. IXs are explored in III-V and II-VI semiconductor heterostructures based on GaAs [2–22], GaN [27–30], and ZnO [31, 32]. Among these materials, the highest IX binding energy ~ 30 meV is in ZnO heterostructures [31].

Excitons with high binding energies can be realized in van der Waals heterostructures composed of atomically thin layers of transition-metal dichalcogenides (TMDs) [33–36]. The IX binding energies in TMD heterostructures reach hundreds of meV [26, 37], making IXs stable at room temperature [38, 39]. Propagation of both DXs in TMD monolayers [40–46] and IXs in TMD heterostructures [47–55] is intensively studied. However, in spite of long IX lifetimes in the TMD heterostructures, orders of magnitude longer than DX lifetimes, a relatively short-range IX propagation with $d_{1/e}$ up to ~ 3 µm was reported in the studies of TMD heterostructures.

The short-range of IX propagation likely originates from substantial in-plane potentials in TMD heterostructures. In-plane potentials localize excitons and suppress exciton transport. In addition to a long IX lifetime, a long-range IX propagation requires that the in-plane potential is small. In particular, the long-range IX propagation with a high IX diffusion coefficient is realized in GaAs heterostructures with small in-plane disorder potentials [20].

In contrast to GaAs heterostructures, in mechanically stacked TMD heterostructures, the layers are not perfectly aligned and the misalignment can cause significant moiré superlattice potentials. For MoSe2/WSe2 heterostructures, similar to the heterostructure studied in this work, the IX energy modulations in moiré superlattice potentials are predicted to be in the range of tens of meV [56–60]. Moiré superlattices enable studying excitons in in-plane potentials, the period $b = a/\sqrt{\delta^2 + \delta^2}$ is typically in the ~ 10 nm range ($a$ is the lattice constant, $\delta$ the lattice mismatch, $\theta$ the twist angle deviation from $n\pi/3$, $n$ is an integer) [61–66], and the moiré potential landscapes can be affected by atomic reconstruction [67–71]. However, for the exciton propagation, the moiré potentials can cause an obstacle and, along with in-plane disorder potentials, can be responsible for limiting the IX propagation distances to $d_{1/e} \sim 3$ µm in the TMD heterostructures [47–55].

Recently, IX propagation with $d_{1/e} \sim 10$ µm was observed in a TMD heterostructure [72]: While the IX propagation was suppressed at zero bias, the IX propagation with $d_{1/e} \sim 10$ µm was realized by applying voltage
between the top and bottom electrodes. The origin of this voltage-controlled IX propagation was discussed in terms of the theoretically predicted [59] tuning of the moiré potential by electric-field, enabling the IX delocalization.

In this work, we realize in a MoSe$_2$/WSe$_2$ TMD heterostructure a macroscopically long-range IX propagation with $d_{1/e}$ reaching $\sim 100$ µm. The long-range IX propagation is realized using an optical excitation with the energy close to the MoSe$_2$ or WSe$_2$ DX energy.

**RESULTS AND DISCUSSION**

The MoSe$_2$/WSe$_2$ heterostructure is assembled by stacking mechanically exfoliated 2D crystals on a graphite substrate. The MoSe$_2$ and WSe$_2$ monolayers are encapsulated by dielectric cladding layers of hexagonal boron nitride (hBN). IXs are formed from electrons and holes confined in adjacent MoSe$_2$ and WSe$_2$ monolayers, respectively. No electrodes are attached to the MoSe$_2$ layer or the WSe$_2$ layer or the top hBN layer and no voltage is applied in the heterostructure. In the absence of applied voltage, IXs form the ground state in the MoSe$_2$/WSe$_2$ heterostructure. The heterostructure details are presented in Supporting Information (SI).

Figure 1 shows that the long-range IX propagation with $d_{1/e}$ reaching $\sim 100$ µm (Fig. 1a,b) is realized when the optical excitation has the energy $E_{ex}$ close to the MoSe$_2$ or WSe$_2$ DX energy (Fig. 1e). The IX propagation with even longer $d_{1/e}$ is likely realized for $E_{ex}$ close to the MoSe$_2$ or WSe$_2$ DX energy, however, the finite heterostructure dimensions limit the longest $d_{1/e}$, which can be reliably established, to $\sim 100$ µm (Fig. 1a,b). In contrast, for a non-resonant excitation, the range of IX propagation is substantially shorter (Fig. 1a,b). The IX luminescence is traced along the entire IX propagation path from the excitation spot to the edge of the heterostructure (Fig. 1c). Spatial intensity modulations along the IX propagation will be considered elsewhere.

Below, we discuss the factors, which may contribute to the observed long-range of IX propagation. IXs have built-in electric dipoles $\sim e d_e$ ($d_e$ is the separation between the electron and hole layers) and the interaction between IXs is repulsive. The repulsive interaction screens the in-plane potential thus facilitating the IX transport [5, 7, 10]. The screening is more effective when the excitation is close to the DX resonances. For instance, heating of the exciton system by the nearly resonant excitation is, in general, smaller than for non-resonant excitation, and the colder IXs can screen the in-plane potential more effectively [7, 10]. The most efficient screening of the in-plane potential by IXs is realized below the temperature of IX condensation [73]. In this experiment, a lower temperature of the exciton system for the nearly resonant excitation is, in particular, evidenced by a lower relative intensity of higher-energy direct excitons (Fig. 1c,d).

The nearly resonant excitation produces a higher IX density due to a higher absorption. The higher-density IXs (i) better screen the in-plane potential and (ii) create a higher IX energy enhancement at the excitation spot facilitating the IX drift from the origin [7] (the drift-diffusion description of IX transport [7] is outlined in SI). The higher IX densities can be also achieved for non-resonant excitation using higher excitation powers $P_{ex}$. However, increasing $P_{ex}$ of nonresonant excitation to achieve the same IX signal as for the nearly resonant excitation produces a shorter $d_{1/e}$, see SI. This indicates that the nearly resonant excitation is essential for the achievement of the long-range IX propagation.

The IX energy increase due to the repulsive interaction $\delta E$ (Fig. 4c) can be used for estimating the IX density $n$. For instance, for the optimal IX propagation conditions, that is for the nearly resonant excitation (Fig. 1) and...
FIG. 2: Time-resolved IX propagation. (a) IX luminescence profiles during the laser excitation pulse at different times. (b) Normalized IX luminescence intensity vs. time at different positions. (c) R vs. 𝜏 dt. 𝜏 dt is the time to reach 70% of the maximum intensity at distance R from the excitation spot relative to that time in the excitation spot. (d) R² vs. 𝜏 dt. The dashed line is a fit to the data with D² = R²/𝜏 dt = 70 cm²/s. For all data, the times are given at the ends of the 10 ns signal integration windows, the excitation pulse starts at t = 0, Eex = 1.694 eV, Pex = 0.15 mW, T = 1.7K, the ~ 2 μm laser spot is centered at x = 0.

FIG. 3: Excitation power and temperature dependence of IX propagation. (a-c) Normalized IX luminescence profiles for different Pex (a,b) and temperatures (c). The greatest IX propagation occurs at T ~ 6 K and Pex ~ 0.2 mW. The dashed line shows the IX luminescence profile in the WSe₂ monolayer, this profile is close to the laser excitation profile for a short IX propagation. T = 6 K (a,b,d), Pex = 0.2 mW (c,e). (d,e) The 1/e decay distance of IX luminescence d(1/e) vs. Pex (d) and vs. temperature (e), extracted from fitting exponential decays to the profiles in (a-c) at x = 0 – 11 μm. (f) Contour plot showing the decay distance d(1/e) vs. both IX density in the excitation spot n and temperature. n is estimated in the mean-field approximation from δE = nμ0, δE is the energy shift of the IX line relative to its energy at the lowest Pex (Fig. 4c). For all data, Eex = 1.623 eV, the ~ 1.5 μm laser spot is centered at x = 0.

the temperature and Pex corresponding to the long d(1/e) (Fig. 3), δE ~ 3 meV (Fig. 4c) and an estimate for n using the mean-field “plate capacitor” formula δE = nμ0 [74] gives n ~ 4 × 10¹¹ cm⁻² (μ0 = 4πe²δz/ε, δz ~ 0.6 nm, the dielectric constant ε ~ 7.4 [75]). This estimate can be improved by taking into account IX correlations that increases n in comparison to the mean-field estimate (by ~ 3 times for the IX systems in GaAs heterostructures [10, 76], with similar estimates for correlations in TMD heterostructures yet unavailable).

Figure 2 shows the kinetics of IX propagation from the near field for the optimal propagation conditions. The IX kinetics is measured during the rectangular-shaped laser excitation pulses with the duration 100 ns and period 300 ns. The 200 ns off time exceeds the IX lifetime (Fig. 4) and is sufficient for a substantial decay of the IX signal. The IX luminescence at locations away from the excitation spot is delayed in comparison to the IX luminescence in the excitation spot (Fig 2a,b). The delay times 𝜏 dt for the IX cloud to expand to the locations separated by distance R from the origin allow estimating the IX transport characteristics. Fitting R vs. 𝜏 dt by R² ~ D² 𝜏 dt (Fig. 2d), gives an estimate for the effective IX diffusion coefficient D² ~ 70 cm²/s.

For a diffusive IX propagation, both IX drift and diffusion contribute to the expansion of IX cloud. With increasing distance from the origin, both the IX density n and IX interaction energy δE ~ nμ0 decrease. The IX energy gradient causes the IX drift away from the origin. Fitting an IX cloud expansion by R² ~ D² 𝜏 dt probes an effective IX diffusion coefficient D² = D + μμ0, which includes both the diffusion and drift due to the density gradient [77] (a drift due to the in-plane potential landscape in the heterostructure is not included in this equation). The IX mobility μ can be estimated using the Einstein relation μ = D/k_BT, giving D² = D(1 + nμ0/k_BT). For the IX energy nμ0 ~ 3 meV at Pex = 0.15 mW (Fig. 4c), D² ~ 70 cm²/s (Fig. 2d), and T = 1.7 K, this equation gives an estimate for the IX diffusion coefficient D ~ 4 cm²/s. In turn, the estimated IX mobility μ = D/k_BT ~ 3 × 10⁴ cm²/(eV s).

The data R vs. 𝜏 dt (Fig. 2c) indicate that an IX cloud expansion occurs with a nearly constant velocity after first few ns. The estimated average velocity of the IX cloud expansion for the time range Δt = 1 – 20 ns, v = ΔR/Δt ~ 5 × 10⁴ cm/s (Fig. 2c).

The excitation power and temperature dependence of
eV

1.382
1.384
1.386
1.388

Energy (eV)

Intensity

0.1

0 10 20
Intensity

0.1

0 10 20

0.15 mW

2.25 mW

0.1

0 10 20

1.7 K

20 K

0.1

0 10 20

τ (ns)

0 10 20 30

0 10 20 30

P (mW)

T (K)

τ (ns)

0.1

0 10 20

Intensity

0.1

0 10 20

0 10 20

0 10 20

0 10 20

FIG. 4: Excitation power and temperature dependence of IX decay kinetics after the excitation pulse end. (a,b) Spatially integrated IX luminescence intensity vs. time for different $P_{\text{ex}}$ (a) and temperatures (b). The times are given at the ends of the 10 ns signal integration windows. The excitation pulse ends at $t = 0$. $T = 1.7$ K (a,c), $P_{\text{ex}} = 0.15$ mW (b,d). (c,d) The IX lifetime $\tau$ (black points) vs. $P_{\text{ex}}$ (c) and temperature (d). $\tau$ is the initial decay time after the excitation pulse end. $E_{\text{ex}} = 1.694$ eV for the kinetics data. Red triangles in (c) show the IX energy in the excitation spot vs. $P_{\text{ex}}$. $E_{\text{ex}} = 1.623$ eV for the $\delta E(P_{\text{ex}})$ data.

IX propagation is presented in Fig. 3. With increasing $P_{\text{ex}}$, the IX propagation distance $d_{1/e}$ changes nonmonotonically. A relatively short-range IX propagation is observed at the lowest $P_{\text{ex}}$ (Fig. 3a,d). This is consistent with the IX localization in the in-plane potential landscape. Increasing $P_{\text{ex}}$ and, in turn, IX density $n$ improves the screening of the in-plane potential and results in an enhancement of IX propagation. The longest IX propagation is achieved at $P_{\text{ex}} \sim 0.2$ mW and the further enhancement of $P_{\text{ex}}$ leads to the suppression of IX propagation (Fig. 3b,d). This suppression is likely caused by the reduction of the IX lifetime at high $P_{\text{ex}}$ (Fig. 4a,c).

With increasing temperature, the IX propagation first enhances, reaches maximum around $T \sim 6$ K, and reduces at higher temperatures (Fig. 3c,e). The enhancement may be related to the thermal delocalization in the in-plane potential landscape, while the suppression at high temperatures is likely caused by the reduction of IX lifetime (Fig. 4b,d). $P_{\text{ex}} - T$ and $n - T$ diagrams for the IX propagation distance $d_{1/e}$ are shown in SI and Fig. 3f, respectively. For the latter, the IX density $n$ is estimated from the IX energy shift (Fig. 4c) using the “plate capacitor” formula $\delta E = n\mu_0$.

Figure 4 shows the IX luminescence decay kinetics after the laser excitation pulse is off. The IX decay times $\tau$ (Fig. 4) are orders of magnitude longer than the DX decay times [78]. Both increasing $P_{\text{ex}}$ (Fig. 4a,c) and temperature (Fig. 4b,d) lead to a reduction of $\tau$. This reduction of $\tau$ suppresses the IX propagation at the high excitation powers and temperatures as outlined above. Rough estimates of the radiative and nonradiative lifetimes from the measured integrated IX luminescence intensity and $\tau$ indicate that the reduction of $\tau$ is mainly caused by the enhanced nonradiative recombination, see SI.

In summary, the macroscopically long-range IX propagation with the decay distances $d_{1/e}$ reaching $\sim 100 \mu$m is observed in a van der Waals MoSe$_2$/WSe$_2$ heterostructure. The long-range IX propagation is realized using an optical excitation with the energy close to the MoSe$_2$ or WSe$_2$ DX energy. The range of excitation powers and temperatures for the long-range IX propagation is determined.

ACKNOWLEDGMENTS

We thank Darius Choksy and Michael Fowler for discussions. These studies were supported by DOE Office of Basic Energy Sciences under Award No. DE-FG02-07ER46449. The heterostructure fabrication and data analysis were supported by NSF Grant No. 1905478.

References

[1] Y.E. Lozovik, V.I. Yudson, A new mechanism for superconductivity: pairing between spatially separated electrons and holes, Sov. Phys. JETP 44, 389 (1976).
[2] M. Hagn, A. Zrenner, G. Böhm, G. Weimann, Electric-field-induced exciton transport in coupled quantum well structures, Appl. Phys. Lett. 67, 232 (1995).
[3] L.V. Butov, A.I. Filin, Anomalous transport and luminescence of indirect excitons in AlAs/GaAs coupled quantum wells as evidence for exciton condensation, Phys. Rev. B 58, 1980 (1998).
[4] A.V. Larionov, V.B. Timofeev, J. Hvam, K. Soerensen, Intermwell Excitons in GaAs/AlGaAs Double Quantum Wells and Their Collective Properties, J. Exp. Theor. Phys. 90, 1093 (2000).
[5] L.V. Butov, A.C. Gossard, D.S. Chemla, Macroscopically ordered state in an exciton system, Nature 418, 751 (2002).
[6] Z. Voros, R. Balili, D.W. Snoke, L. Pleiffer, K. West, Long-Distance Diffusion of Excitons in Double Quantum Well Structures, Phys. Rev. Lett. 94, 226401 (2005).
[7] A.L. Ivanov, L.E. Smallwood, A.T. Hammad, Sen Yang, L.V. Butov, A.C. Gossard, Origin of the inner ring in photoluminescence patterns of quantum well excitons, Europhys. Lett. 73, 920 (2006).
[8] A. Gärtnert, A.W. Holleitner, J.P. Kotthaus, D. Schuh, Drift mobility of long-living excitons in coupled GaAs quantum wells, Appl. Phys. Lett. 89, 052108 (2006).
[9] A.A. High, E.E. Novitskaya, L.V. Butov, M. Hanson, A.C. Gossard, Control of exciton fluxes in an excitonic integrated circuit, Science 321, 229 (2008).
[10] M. Remeika, J.C. Graves, A.T. Hammack, A.D. Meyerhagen, M.M. Fogler, L.V. Butov, M. Hanson and A.C. Gossard, Localization-Delocalization Transition of Indirect Excitons in Lateral Electrostatic Lattices, Phys. Rev. Lett. 102, 186803 (2009).

[11] X.P. Vigele, D. Schuh, W. Wegscheider, J.P. Kotthaus, A.W. Holleitner, Density Enhanced Diffusion of Dipolar Excitons within a One-Dimensional Channel, Phys. Rev. Lett. 103, 126402 (2009).

[12] J.R. Leonard, Y.Y. Kuznetsova, S. Yang, L.V. Butov, T. Ostatnický, A. Kavokin, A.C. Gossard, Nano Lett. 9, 4204 (2009).

[13] S. Lazić, P.V. Santos, R. Hey, Exciton transport by moving strain dots in GaAs quantum wells, Phys. E 42, 2640 (2010).

[14] A.G. Winbow, J.R. Leonard, M. Remeika, Y.Y. Kuznetsova, A.A. High, A.T. Hammack, L.V. Butov, J. Wilkes, A.A. Guenther, A.L. Ivanov, M. Hanson, A.C. Gossard, Electrostatic Conveyor for Excitons, Phys. Rev. Lett. 106, 196806 (2011).

[15] A.A. High, J.R. Leonard, A.T. Hammack, M.M. Fogler, L.V. Butov, A.V. Kavokin, K.L. Campman, A.C. Gossard, Spontaneous coherence in a cold exciton gas, Nature 483, 584 (2012).

[16] M. Alloing, A. Lemaître, E. Galopin, F. Dubin, Nonlinear dynamics and inner-ring photoluminescence pattern of indirect excitons, Phys. Rev. B 85, 245106 (2012).

[17] A.A. High, A.T. Hammack, J.R. Leonard, Sen Yang, L.V. Butov, T. Ostatnický, M. Vladimirova, A.C. Gossard, T.C.H. Liew, K.L. Campman, A.C. Gossard, Spin currents in a coherent exciton gas, Phys. Rev. Lett. 110, 246403 (2013).

[18] S. Lazić, A. Violante, K. Cohen, R. Hey, R. Rapaport, P.V. Santos, Scalable interconnections for remote indirect exciton systems based on acoustic transport, Phys. Rev. B 89, 085313 (2014).

[19] R. Finkelstein, K. Cohen, B. Jouault, K. West, L.N. Pfeiffer, M. Vladimirova, R. Rapaport, Transition from spin-orbit to hyperfine interaction dominated spin relaxation in a cold fluid of dipolar excitons, Phys. Rev. B 96, 085404 (2017).

[20] C.J. Dorow, M.W. Hasling, D.J. Choksy, J.R. Leonard, L.V. Butov, K.W. West, L.N. Pfeiffer, High-mobility indirect excitons in wide single quantum well, Appl. Phys. Lett. 113, 212102 (2018).

[21] J.R. Leonard, A.A. High, A.T. Hammack, M.M. Fogler, L.V. Butov, K.L. Campman, A.C. Gossard, Pancharatnam-Berry phase in condensate of indirect excitons, Nat. Commun. 9, 2158 (2018).

[22] J.R. Leonard, Lunhui Hu, A.A. High, A.T. Hammack, Congjun Wu, L.V. Butov, K.L. Campman, A.C. Gossard, Moiré pattern of interference dislocations in condensate of indirect excitons, Nat. Commun. 12, 1175 (2021).

[23] D.S. Chemla, D.A.B. Miller, P.W. Smith, A.C. Gossard, W. Wiegmann, Room temperature excitonic nonlinear absorption and refraction in GaAs/AlGaAs multiple quantum well structures, IEEE J. Quantum Electron. 20, 265 (1984).

[24] K. Sivalertporn, I. Mouchliadis, A.L. Ivanov, R. Philip, E.A. Muljarov, Direct and indirect excitons in semiconductor coupled quantum wells in an applied electric field, Phys. Rev. B 85, 045207 (2012).

[25] A. Zrenner, P. Leeb, J. Schäfler, G. Böhm, G. Weimann, J.M. Worlock, L.T. Florez, J.P. Harbison, Indirect excitons in coupled quantum well structures, Surf. Sci. 263, 496 (1992).

[26] M.M. Fogler, L.V. Butov, K.S. Novoselov, High-temperature superfluidity with indirect excitons in van der Waals heterostructures, Nature Commun. 5, 4555 (2014).

[27] P. Lefebvre, S. Kalliakos, T. Bretagnon, P. Valvin, T. Taliercio, B. Gil, N. Grandjean, J. Massies, Observation and modeling of the time-dependent descreening of internal electric field in a wurtzite GaN/AlZnO:GaAzN quantum well after high photoexcitation, Phys. Rev. B 69, 035307 (2004).

[28] F. Fedichkin, P. Andreakou, B. Jouault, M. Vladimirova, T. Guillet, C. Brimont, P. Valvin, T. Bretagnon, A. Dussaigne, N. Grandjean, P. Lefebvre Transport of dipolar excitons in (Al,Ga)N/GaN quantum wells, Phys. Rev. B 91, 205424 (2015).

[29] F. Fedichkin, T. Guillet, P. Valvin, B. Jouault, C. Brimont, T. Bretagnon, L. Labourcade, N. Grandjean, P. Lefebvre, M. Vladimirova, Room-temperature transport of indirect excitons in (Al,Ga)N/GaN quantum wells, Phys. Rev. Appl. 6, 014011 (2016).

[30] François Chiaruttini, Thierry Guillot, Christelle Brimont, Benoît Jouault, Pierre Lefebvre, Jessica Vives, Sébastien Chenot, Yvon Corder, Benjamin Damilano, Maria Vladimirova, Trapping Dipolar Exciton Fluids in GaN/(Al,Ga)N Nanostructures, Nano Lett. 19, 4911 (2019).

[31] C. Morhain, T. Bretagnon, P. Lefebvre, X. Tang, P. Valvin, T. Guillet, B. Gil, T. Taliercio, M. Teissere-Doninelli, B. Vinter, C. Deparis, Internal electric field in wurtzite ZnO/ZnO: Mg52O2 quantum wells, Phys. Rev. B 72, 241305(R) (2005).

[32] Y.Y. Kuznetsova, F. Fedichkin, P. Andreakou, E.V. Calman, L.V. Butov, P. Lefebvre, T. Bretagnon, T. Guillet, M. Vladimirova, C. Morhain, J.-M. Chauveau, Transport of indirect excitons in ZnO quantum wells, Opt. Lett. 40, 3667 (2015).

[33] A.K. Geim, I.V. Grigorieva, Van der Waals heterostructures, Nature 499, 419 (2013).

[34] Ziliang Ye, Ting Cao, Kevin O’Brien, Hanyu Zhu, Xiaobo Yin, Yuan Wang, Steven G. Louie, Xiang Zhang, Probing excitonic dark states in single-layer tungsten disulphide, Nature 513, 214 (2014).

[35] Alexey Chernikov, Timothy C. Berkelbach, Heather M. Hill, Albert Rigosi, Yilei Li, Ozgur Burak Aslan, David Hill, Albert Rigosi, Yilei Li, Ozgur Burak Aslan, David C. Smalley, Andrei Kuznetsova, Yoon Kwang Seo, Hyung Min Heo, J. Tang, Xiang Zhang, Probing excitonic dark states in single-layer tungsten disulphide, Nature 513, 214 (2014).

[36] M. Goryca, J. Li, A.V. Stier, T. Taniguchi, K. Watanabe, E. Courtade, S. Shree, C. Robert, B. Urbaszek, X. Marie, S.A. Crooker, Revealing exciton masses and dielectric properties of monolayer semiconductors with high magnetic fields, Nature Commun. 10, 4172 (2019).

[37] Thorsten Deilmann, Kristian Sommer Thygesen, Interlayer Trions in the MoS2/WSe2 van der Waals Heterostructure, Nano Lett. 18, 1460 (2018).

[38] E.V. Calman, M.M. Fogler, L.V. Butov, S. Hu, A. Mishchenko, A.K. Geim, Indirect excitons in van der Waals heterostructures at room temperature, Nature Commun. 9, 1895 (2018).

[39] E.V. Calman, L.H. Fowler-Gerae, D.J. Choksy, L.V. Butov, D.E. Nikonov, I.A. Young, S. Hu, A. Mishchenko, A.K. Geim, Indirect Excitons and Trions in MoSe2/WSe2 van der Waals Heterostructures, Nano Lett. 20, 1869 (2020).

[40] Nardeep Kumar, Qiannan Cui, Frank Ceballos, Dawei He, Yongsheng Wang, Hui Zhao, Exciton diffusion in mono-
layer and bulk MoSe₂, Nanoscale 6, 4915, (2014).

[41] Marvín Külig, Jonas Zipfel, Philipp Nagler, Sofia Blanter, Christian Schüller, Tobias Korn, Nicola Paradiso, Mikhail M. Glazov, Alexey Chernukov, Exciton Diffusion and Halo Effects in Monolayer Semiconductors, Phys. Rev. Lett. 120, 207401 (2018).

[42] F. Cadiz, C. Robert, E. Courtade, M. Manca, L. Martinelli, T. Taniguchi, K. Watanabe, T. Amand, A.C.H. Rowe, D. Paget, B. Urbaszek, X. Marie, Exciton diffusion in WSe₂ monolayers embedded in a van der Waals heterostructure, Appl. Phys. Lett. 112, 152106 (2018).

[43] Darwin F. Cordovilla Leon, Zidong Li, Sung Woon Jang, C.-Hsuan Cheng, Parag B. Deotare, Exciton transport in strained monolayer WSe₂, Appl. Phys. Lett. 113, 252101 (2018).

[44] Darwin F. Cordovilla Leon, Zidong Li, Sung Woon Jang, Parag B. Deotare, Hot exciton transport in WSe₂ monolayers, Phys. Rev. B 100, 241401(R) (2019).

[45] Shengcai Hao, Matthew Z. Bellus, Dawei He, Yongsheng Wang, Hui Zhao, Controlling exciton transport in monolayer MoSe₂ by dielectric screening, Nanoscale Horiz. 5, 139 (2020).

[46] Kanak Datta, Zhengyang Lyu, Zidong Li, Takashi Taniguchi, Kenji Watanabe, Parag B. Deotare, Spatiotemporally controlled room-temperature exciton transport under dynamic strain, Nature Photonics 16, 242 (2022).

[47] Pasqual Rivera, Kyle L. Seyler, Hongyi Yu, John R. Schabley, Jiaqiang Yan, David G. Mandrus, Wang Yao, Xiaodong Xu, Valley-polarized exciton dynamics in a 2D semiconductor heterostructure, Science 351, 688 (2016).

[48] Luis J. Arcos, Andrew Y. Jee, Katelyna Piscitello, Dominik S. Wild, Alexander A. High, You Zhou, Giovanni Scuri, Kristaian De Greve, Andrey Sushko, Che-Hang Yu, Takashi Taniguchi, Kenji Watanabe, Daniel J. needleman, Mikhail D. Lukin, Hongkun Park, Philip Kim, Electrical control of interlayer exciton dynamics in atomically thin heterostructures, Science 366, 870 (2019).

[49] Dmitrii Unuchek, Alberto Ciarrocchi, Ahmet Avsar, Kenji Watanabe, Takashi Taniguchi, Andras Kis, Room-temperature electrical control of exciton flux in a van der Waals heterostructure, Nature 560, 340 (2019).

[50] Dmitrii Unuchek, Alberto Ciarrocchi, Ahmet Avsar, Zhe Sun, Kenji Watanabe, Takashi Taniguchi, Andras Kis, Valley-polarized exciton currents in a van der Waals heterostructure, Nature Nanotechnology 14, 1104 (2019).

[51] Yuanda Liu, Kevin Dini, Qinghao Tai, Timothy Liew, Kostya S. Novoselov, Weibo Gao, Electrically controllable router of interlayer excitons, Sci. Adv. 6, eaba1830 (2020).

[52] Zunpeng Huang, Yuanda Liu, Kevin Dini, Qinghao Tai, Zhuojun Liu, Hanlin Fang, Jiu Liu, Timothy Liew, Weibo Gao, Robust room temperature valley Hall effect of interlayer excitons, Nano Lett. 20, 1345 (2020).

[53] Long Yuan, Biiyuan Zheng, Jens Kunstmann, Thomas Brumme, Agnieszka Beata Kuc, Chao Ma, Shihbin Deng, Daria Blach, Anli Pan, Libai Huang, Twist-angle-dependent interlayer exciton diffusion in WS₂/WSe₂ heterobilayers, Nat. Mat. 19, 617 (2020).

[54] Jue Wang, Qianhui Shi, En-Min Shih, Lin Zhou, Wenjing Wu, Yusong Bai, Daniel Rhodes, Katayun Barmak, James Hone, Cory R. Dean, X.-Y. Zhu, Diffusivity Reveals Three Distinct Phases of Interlayer Excitons in MoSe₂/WSe₂ Heterobilayers, Phys. Rev. Lett. 126, 106804 (2021).

[55] Zhe Sun, Alberto Ciarrocchi, Fedele Tagarelli, Juan Francisco Gonzalez Marin, Kenji Watanabe, Takashi Taniguchi, Andras Kis, Excitonic transport driven by repulsive dipolar interaction in a van der Waals heterostructure, Nature Photonics 16, 79 (2022).

[56] Fengchong Wu, Timothy Lovorn, A.H. MacDonald, Theory of optical absorption by interlayer excitons in transition metal dichalcogenide heterobilayers, Phys. Rev. B 97, 035306 (2018).

[57] Hongyi Yu, Gui-Bin Liu, Wang Yao, Brightened spin-polarized interlayer excitons and optical selection rules in van der Waals heterobilayers, 2D Mater. 5, 035021 (2018).

[58] Fengchong Wu, Timothy Lovorn, A.H. MacDonald, Topological Exciton Bands in Moiré Heterojunctions, Phys. Rev. Lett. 118, 147401 (2017).

[59] Hongyi Yu, Gui-Bin Liu, Jianu Tang, Xiaodong Xu, Wang Yao, Moiré excitons: From programmable quantum emitter arrays to spin-orbit-coupled artificial lattices, Sci. Adv. 3, e1701696 (2017).

[60] Chandong Zhang, Chih-Piao Chuu, Xiaobo Ren, Ming-Yang Li, Lain-Jong Li, Chuanhong Jin, Mei-Yin Chou, Cih-Kang Shih, Interlayer couplings, Moiré patterns, and 2D electronic superlattices in MoS₂/WSe₂ heterobilayers, Sci. Adv. 3, e1601459 (2017).

[61] Pasqual Rivera, Hongyi Yu, Kyle L. Seyler, Nathan P. Wilson, Wang Yao, Xiaodong Xu, Interlayer valley excitons in heterobilayers of transition metal dichalcogenides, Nature Nanotech. 13, 1004 (2018).

[62] Kyle L. Seyler, Pasqual Rivera, Hongyi Yu, Nathan P. Wilson, Essence L. Ray, David G. Mandrus, Jiaqiang Yan, Wang Yao, Xiaodong Xu, Signatures of moiré-trapped valley excitons in MoS₂/WSe₂ heterobilayers, Nature 567, 66 (2019).

[63] Kha Tran, Galan Moody, Fengcheng Wu, Xiaobo Lu, Junho Choi, Kyoungwhan Kim, Amitesh Rai, Daniel A. Sanchez, Jiamin Quan, Akshay Singh, Jacob Embley, André Zepeda, Marshall Campbell, Travis Autry, Takashi Taniguchi, Kenji Watanabe, Nanshu Lu, Sanjay K. Banerjee, Kevin L. Silverman, Suennie Kim, Emanuel Tutuc, Li Yang, Allan H. MacDonald, Xiaqin Li, Evidence for moiré excitons in van der Waals heterostructures, Nature 567, 71 (2019).

[64] Olechao Jin, Emma C. Regan, Aiming Yan, M. Iqbal Bakti Utama, Danqing Wang, Sihan Zhao, Ying Qin, Sijie Yang, Zhiren Zheng, Shenyang Shi, Kenji Watanabe, Takashi Taniguchi, Sefaattin Tongay, Alex Zettl, Feng Wang, Observation of moiré excitons in WS₂/WSe₂ heterostructures, Nature 567, 76 (2019).

[65] Evgeny M. Alexeev, David A. Ruiz-Tijerina, Mark Danovich, Matthew J. Hamer, Daniel J. Terry, Pramod K. Nayak, Seongjoon Ahn, Sangyeon Pak, Juwon Lee, Jung Inn Sohn, Maciej R. Molas, Maciej Koperski, Kenji Watanabe, Takashi Taniguchi, Kostya S. Novoselov, Roman V. Gorbachev, Hyeon Suk Shin, Vladimir I. Fal’ko, Alexander I. Tartakovskii, Resonantly hybridized excitons in moiré superlattices in van der Waals heterostructures, Nature 567, 81 (2019).

[66] Nathan P. Wilson, Wang Yao, Jie Shan, Xiaodong Xu, Excitons and emergent quantum phenomena in stacked 2D semiconductors, Nature 599, 383 (2021).

[67] Jonathan S. Alden, Adam W. Tsen, Pinshane H. Huang, Robert Hovden, Lola Brown, Jiwoong Park, David A. Muller, and Paul L. McEuen, Strain solitons and topological defects in bilayer graphene, PNAS 110, 11256 (2013).

[68] C.R. Woods, L. Britnell, A. Eckmann, R.S. Ma, J.C. Lu, H.M. Guo, X. Lin, G.L. Yu, Y. Cao, R.V. Gorbachev, A.V.
Kretinin, J. Park, L.A. Ponomarenko, M.I. Katsnelson, Yu.N. Gornostyrev, K. Watanabe, T. Taniguchi, C. Casiraghi, H-J. Gao, A.K. Geim, and K.S. Novoselov, Commensurate–incommensurate transition in graphene on hexagonal boron nitride, Nature Phys. 10, 451 (2014).

[69] Hyobin Yoo, Rebecca Engelke, Stephen Carr, Shiang Fang, Kuan Zhang, Paul Cazeaux, Suk Hyun Sung, Robert Hovden, Adam W. Tsen, Takashi Taniguchi, Kenji Watanabe, Gyu-Chul Yi, Miyoung Kim, Mitchell Luskin, Ellad B. Tadmor, Efthimios Kaxiras, and Philip Kim, Atomic and electronic reconstruction at the van der Waals interface in twisted bilayer graphene, Nature Mat. 18, 448 (2019).

[70] Astrid Weston, Yichao Zou, Vladimir Enaldiev, Alex Summerfield, Nicholas Clark, Viktor Zólyomi, Abigail Graham, Celal Yelgel, Samuel Magorrian, Mingwei Zhou, Johanna Zultak, David Hopkinson, Alexei Barinov, Thomas H. Bointon, Andrey Kretinin, Neil R. Wilson, Peter H. Beton, Vladimir I. Fal'ko, Sarah J. Haigh, and Roman Gorbachev, Atomic reconstruction in twisted bilayers of transition metal dichalcogenides, Nature Nano. 15, 592 (2020).

[71] Matthew R. Rosenberger, Hsun-Jen Chuang, Madeleine Phillips, Vladimir P. Oleshko, Kathleen M. McCreary, Saujan V. Sivaram, C. Stephen Hellberg, and Berend T. Jonker, Twist Angle-Dependent Atomic Reconstruction and Moiré Patterns in Transition Metal Dichalcogenide Heterostructures, ACS Nano 14, 4580 (2020).

[72] L.H. Fowler-Gerace, D.J. Choksy, L.V. Butov, Voltage-controlled long-range propagation of indirect excitons in a van der Waals heterostructure, Phys. Rev. B 104, 165302 (2021).

[73] Dmitri E. Nikonov, Atac Imamoglu, Bose condensation in two dimensions with disorder: Gross-Pitaevskii approach, arXiv:quant-ph/9806003 (1998).

[74] D. Yoshioka and A. H. Macdonald, Double quantum well electron-hole systems in strong magnetic fields, J. Phys. Soc. Jpn. 59, 4211 (1990).

[75] Akash Laturia, Maarten L. Van de Put, William G. Vandenberghhe, Dielectric properties of hexagonal boron nitride and transition metal dichalcogenides: from monolayer to bulk, 2D Materials and Applications 2, 6 (2018).

[76] D.J. Choksy, Chao Xu, M.M. Fogler, L.V. Butov, J. Norman, A.C. Gossard, Attractive and repulsive dipole interaction in bilayers of indirect excitons, Phys. Rev. B 103, 045126 (2021).

[77] C.J. Dorow, M.W. Hasling, E.V. Calman, L.V. Butov, J. Wilkes, K.L. Campman, A.C. Gossard, Spatially resolved and time-resolved imaging of transport of indirect excitons in high magnetic fields, Phys. Rev. B 95, 235308 (2017).

[78] T. Korn, S. Heydrich, M. Hirmer, J. Schmutzler, C. Schütter, Low-temperature photocarrier dynamics in monolayer MoS₂, Appl. Phys. Lett. 99, 102109 (2011).
Supporting Information for
Long-range propagation of indirect excitons in van der Waals heterostructure: 1/e decay distance reaches ∼ 100 microns

L. H. Fowler-Gerace,1 Zhiwen Zhou,1 E. A. Szwed,1 and L. V. Butov1

1Department of Physics, University of California at San Diego, La Jolla, CA 92093, USA
(Dated: April 20, 2022)

PACS numbers:

The heterostructure details

The van der Waals heterostructure was assembled using the dry-transfer peel-and-lift technique [1]. Crystals of hBN, MoSe2, and WSe2 were first mechanically exfoliated onto different Si substrates that were coated with a double polymer layer consisting of polymethyl glutarimide (PMGI) and polymethyl methacrylate (PMMA). The bottom PMGI was then dissolved with the tetramethylammonium hydroxide based solvent CD-26, causing the top PMMA membrane with the target 2D crystal to float on top of the solvent. The PMMA membrane functions both as a support substrate for transferring the crystal and as a barrier to protect the crystal from the solvent. Separately, a large graphite crystal was exfoliated onto an oxidized Si wafer, which served as the basis for the heterostructure. The PMMA membrane supporting the target crystal was then flipped over and aligned above a flat region of the graphite crystal using a micromechanical transfer stage. The two crystals were brought into contact and the temperature of the stage was ramped to 800 °C in order to increase adhesion between the 2D crystals. Then, the PMMA membrane was peeled off leaving the bilayer stack on the wafer. The procedure was repeated leading to a multicrystal stack with the desired layer sequence. Sample annealing was performed by immersing the sample in Remover PG, an N-methyl-2-pyrrolidone (NMP) based solvent stripper, at 70°C for 12 hours.

No intentional sample doping was done; however, unintentional n-type doping is typical for TMD layers [1]. The thickness of bottom and top hBN layers is about 40 and 30 nm, respectively. The MoSe2 layer is on top of the WSe2 layer. The long WSe2 and MoSe2 edges reach ∼ 30 and ∼ 20 µm, respectively, which enables a rotational alignment between the WSe2 and MoSe2 monolayers. The twist angle estimated from the angle between the long WSe2 and MoSe2 edges δθ = 0.5° ± 0.8° (Fig. S1).

Figure S1 presents a microscope image showing the layer pattern of the heterostructure. The layer boundaries are indicated. The hBN layers cover the entire areas of MoSe2 and WSe2 layers. There was a narrow graphene electrode on the top of the heterostructure around x = 2 µm for y = 0, Fig. S1. This electrode was detached. The IX luminescence reduction around x = 2 µm can be related with residual graphene layers on the heterostructure.

The discussions in this work apply to both R and H (AA and AB) stacking; therefore we did not verify stacking in the sample. Our work shows that the long-range IX propagation can be realized in TMD heterostructures using an optical excitation with the energy close to the DX energy. The studies of sample statistics, and in particular, verifying the role of the rotational alignment between crystals on the IX propagation, is the subject for future works.

FIG. S1: (a) Schematic energy-band diagram for the heterostructure. The ovals indicate a direct exciton (DX) and an indirect exciton (IX) composed of an electron (−) and a hole (+). (b) A microscope image showing the layer pattern of the heterostructure. The green, red, cyan, and orange lines indicate the boundaries of WSe2 and MoSe2 monolayers and bottom and top hBN layers, respectively.
Optical measurements

In the cw experiments, excitons were generated by a cw Ti:Sapphire laser with tunable excitation energy or a cw HeNe laser with excitation energy $E_{ex} = 1.96$ eV. Luminescence spectra were measured using a spectrometer with resolution 0.2 meV and a liquid-nitrogen-cooled CCD.

The IX luminescence kinetics was measured using a pulsed semiconductor laser with $E_{ex} = 1.694$ eV nearly resonant to WSe$_2$ DX energy. The emitted light was detected by a liquid-nitrogen-cooled CCD coupled to a PicoStar HR TauTec time-gated intensifier.

The experiments were performed in a variable-temperature 4He cryostat. The sample was mounted on an Attocube xyz piezo translation stage allowing adjusting the sample position relative to a focusing lens inside the cryostat.

The drift-diffusion description of IX transport

The drift-diffusion model of IX transport [2–4], used for the estimates for IX diffusion coefficient and mobility in the main text, is outlined in this section. Within this model, IX transport is described by the equation for IX density $n$

$$\frac{\partial n}{\partial t} = \nabla \left[ D \nabla n + \mu n \nabla (u_0 n) \right] + \Lambda - \frac{n}{\tau}$$  (1)

The first and second terms in square brackets in Eq. 1 describe IX diffusion and drift currents, respectively. The latter originates from the IX repulsive dipolar interactions and is approximated by the mean field “plate capacitor” formula for the IX energy shift with density $\delta E = n u_0$, $u_0 = 4 \pi e^2 d / \epsilon$ [5]. The diffusion coefficient

$$D = D(0) \exp\left[-U(0) / (k_B T + n u_0)\right]$$  (2)

accounts for the temperature- and density-dependent screening of the long-range-correlated in-plane potential landscape by interacting IXs, $D(0)$ is the diffusion coefficient in the absence of in-plane potential and $U(0)/2$ is the amplitude of the in-plane potential [2–4]. The IX mobility $\mu$ is given by the Einstein relation $\mu = D / (k_B T)$. The IX generation rate $\Lambda$ has a profile of the laser excitation spot. $\tau$ is the IX lifetime.

Both the IX-interaction-induced screening of in-plane potential and the IX-interaction-induced drift from the origin are important for the enhancement of IX transport with increasing IX density $n$. In particular, within this model, the enhancement of IX transport due to the IX-interaction-induced screening of in-plane potential is described by Eq. 2, while the enhancement of IX transport due to the IX-interaction-induced drift from the origin is described by Eq. 3 as outlined in the main text

$$D' = D[1 + n u_0 / (k_B T)].$$  (3)

The drift-diffusion Eq. 1 can be supplemented by the thermalization equation, which describes heating of excitons by photoexcitation and cooling via interaction with phonons, and by including the heterostructure in-plane potential landscape in the drift term [3, 4]. In particular, the exciton thermalization can lead to the appearance of the inner ring in exciton luminescence patterns [3, 4]. The Einstein relation can be extended to the generalized Einstein relation $\mu = D(e^{T_0/T} - 1) / (k_B T_0)$, which gives the classical $\mu = D / (k_B T)$ for $T \gg T_0$, where $T_0 = 2 \pi h^2 n / (g_k m)$ is the temperature of quantum degeneracy, $g$ the spin degeneracy, $m$ the exciton mass [2–4]. Including these factors to the analysis of IX propagation forms a subject for future work.

IX interaction energies and the long-range IX propagation

Below, we briefly discuss the IX interaction energies $\delta E$ corresponding to the long-range IX propagation. In GaAs heterostructures, the strong enhancement of IX propagation, the IX delocalization, is observed when the IX interaction energy becomes comparable to the amplitude of the in-plane potential, which is, in turn, comparable to the IX luminescence linewidth at low IX densities [6]. For the long-range IX propagation in the MoSe$_2$/WSe$_2$ heterostructure (Fig. 3), the IX interaction energy $\delta E \sim 3$ meV (Fig. 4c). This value is comparable to the smallest IX linewidth $\sim 3$ meV at low IX densities in the MoSe$_2$/WSe$_2$ heterostructure (Fig. S2), in qualitative agreement with the data in GaAs heterostructures [6].

However, this value, $\delta E \sim 3$ meV, is significantly smaller than the predicted IX energy modulations in moiré superlattice potentials in MoSe$_2$/WSe$_2$ heterostructures that are in the range of tens of meV [7–11]. Understanding
FIG. S2: IX luminescence spectrum at a low IX density in the MoSe$_2$/WSe$_2$ heterostructure. $P_{ex} = 0.005$ mW, $T = 1.7$ K, $E_{ex} = 1.96$ eV.

FIG. S3: $d_{1/e}$ extracted from fitting exponential decays to IX luminescence profiles at $x = 0 - 11 \mu$m vs. integrated IX intensity in the entire MoSe$_2$/WSe$_2$ heterostructure. Excitation is either nonresonant ($E_{ex} = 1.96$ eV, blue), or near resonant ($E_{ex} = 1.687$ eV, red) to the DX. $T = 1.7$ K, the $\sim 1.5 \mu$m laser spot is centered at $x = 0$.

The long-range IX propagation in the predicted high-amplitude in-plane potential forms a subject for future work. In particular, the scale of the moiré superlattice potentials, with estimated period $\sim 20$ nm small in comparison to the long-scale potentials considered in [6], and effects of atomic reconstruction on the in-plane potential [12–16] can be considered.

**IX propagation decay distance for nearly resonant excitation and nonresonant excitation**

The nearly resonant excitation produces a higher IX density and, in turn, a stronger IX luminescence signal due to a higher absorption (Fig. 1). The higher IX densities can be also achieved by nonresonant excitation with higher excitation powers $P_{ex}$. Figure S3 compares the IX propagation decay distances $d_{1/e}$ for the nearly resonant and nonresonant excitations. Figure S3 shows that in the regime of the long-range IX propagation, for the same IX signal in the heterostructure, achieved by a higher $P_{ex}$ for nonresonant excitation, $d_{1/e}$ is longer in the case of the nearly resonant excitation. This indicates that the nearly resonant excitation is essential for the achievement of the long-range IX propagation.

**Rough estimates of the radiative and nonradiative lifetime variation with $P_{ex}$ and temperature**

The IX radiative and nonradiative lifetimes, $\tau_r$ and $\tau_{nr}$, can be roughly estimated from the measured IX total luminescence intensity $I$ and decay time $\tau$. Figure S4 shows $\tau_r$ and $\tau_{nr}$ estimated using $\tau_r^{-1} = (I/\Lambda)^{-1}$ and $\tau_{nr}^{-1} = \tau^{-1} - \tau_r^{-1}$. Within this estimate, the IX luminescence kinetics is approximated as monoeponential. It is also assumed that there is no IX spatial escape from the system. This is approximated by integrating the IX luminescence signal over the heterostructure. For an estimate of the generation rate $\Lambda$, we use $I_D = \alpha \Lambda$, where $I_D$ is the DX luminescence signal for the excitation at the MoSe$_2$ monolayer outside the heterostructure and $\alpha$ is the quantum efficiency for this DX luminescence. Qualitatively similar variations of $\tau_r$ and $\tau_{nr}$ with $P_{ex}$ and temperature are obtained for different values of $\alpha$ (Fig. S4). These estimates suggest that the reduction of $\tau$ at the high excitation powers and temperatures is mainly caused by the enhanced nonradiative recombination (Fig. S4).
FIG. S4: Rough estimate of radiative and nonradiative lifetimes, $\tau_r$ and $\tau_{nr}$, (a,b) Spatially integrated IX luminescence intensity $I$ (green) and IX lifetime $\tau$ (blue) vs. $P_{ex}$ (a) and temperature (b). $T = 1.7$ K (a), $P_{ex} = 0.15$ mW (b), and $E_{ex} = 1.694$ eV (a,b). $\tau$ is the initial decay time after the excitation pulse end. (c-f) Estimated $\tau_r$ and $\tau_{nr}$ vs. $P_{ex}$ (c,e) and temperature (d,f) for the quantum efficiency of DX luminescence in the MoSe$_2$ monolayer $\alpha = 100\%$ (c,d) and $25\%$ (e,f).

FIG. S5: Normalized IX luminescence profiles for different laser excitation energies $E_{ex}$. The dashed line shows the DX luminescence profile in the WSe$_2$ monolayer, this profile is close to the laser excitation profile for a short DX propagation. $P_{ex} = 0.2$ mW, $T = 1.7$ K, the $\sim 1.5$ $\mu$m laser spot is centered at $x = 0$.

IX luminescence profiles for different laser excitation energies

Normalized IX luminescence profiles for different laser excitation energies $E_{ex}$ are shown in Fig. S5. The long-range IX propagation with high decay distances $d_{1/e}$ is realized for $E_{ex}$ close to the MoSe$_2$ or WSe$_2$ DX energy (Fig. 1 in the main text). For some of the IX luminescence profiles the IX luminescence intensity increases with separation from the origin and no decay is observed within the finite heterostructure dimensions. The description of this intensity increase is beyond Eq. 1 and may require supplementing this equation by the thermalization equation, which describes heating
FIG. S6: Contour plot showing the decay distance $d_{1/e}$ vs. both laser excitation power $P_{ex}$ and temperature. $E_{ex} = 1.623$ eV, the $\sim 1.5 \mu m$ laser spot is centered at $x = 0$.

of excitons by photoexcitation and cooling via interaction with phonons, and including the heterostructure in-plane potential landscape in the drift term [3, 4]. In particular, the exciton thermalization can lead to the appearance of the inner ring in exciton luminescence patterns, that is an exciton luminescence intensity increase with separation from the origin [3, 4]. The data with the fit indicating $d_{1/e} > 100 \mu m$ and, in particular, showing the IX luminescence intensity increase with separation from the origin, are presented in Fig. 1b by points on the edge.

$P_{ex} - T$ diagram for the IX propagation distance $d_{1/e}$

$P_{ex} - T$ diagram for the IX propagation distance $d_{1/e}$ is shown in Fig. S6. The $n - T$ diagram for $d_{1/e}$ in Fig. 3f in the main text is obtained from this diagram with the IX density $n$ estimated from the IX energy shift (Fig. 4c) using the “plate capacitor” formula $\delta E = nu_0$.

References

[1] F. Withers, O. Del Pozo-Zamudio, A. Mishchenko, A.P. Rooney, A. Gholinia, A. Watanabe, T. Taniguchi, S.J. Haigh, A.K. Geim, A.I. Tartakovskii, K.S. Novoselov, Light-emitting diodes by band-structure engineering in van der Waals heterostructures, Nat. Mater. 14, 301 (2015).
[2] A.L. Ivanov, Quantum diffusion of dipole-oriented indirect excitons in coupled quantum wells, Europhys. Lett. 59, 586 (2002).
[3] A.L. Ivanov, L.E. Smallwood, A.T. Hammack, Sen Yang, L.V. Butov, A.C. Gossard, Origin of the inner ring in photoluminescence patterns of quantum well excitons, Europhys. Lett. 73, 920 (2006).
[4] A.T. Hammack, L.V. Butov, J. Wilkes, L. Mouchliadis, E.A. Muljarov, A.L. Ivanov, A.C. Gossard, Kinetics of the inner ring in the exciton emission pattern in coupled GaAs quantum wells, Phys. Rev. B 80, 155331 (2009).
[5] D. Yoshioka, A.H. Macdonald, Double quantum well electron-hole systems in strong magnetic fields, J. Phys. Soc. Jpn. 59, 4211 (1990).
[6] M. Remeika, J.C. Graves, A.T. Hammack, A.D. Meyertholen, M.M. Fogler, L.V. Butov, M. Hanson and A.C. Gossard, Localization-Delocalization Transition of Indirect Excitons in Lateral Electrostatic Lattices, Phys. Rev. Lett. 102, 186803 (2009).
[7] Fengcheng Wu, Timothy Lovorn, A.H. MacDonald, Theory of optical absorption by interlayer excitons in transition metal dichalcogenide heterobilayers, Phys. Rev. B 97, 035306 (2018).
[8] Hongyi Yu, Gui-Bin Liu, Wang Yao, Brightened spin-triplet interlayer excitons and optical selection rules in van der Waals heterobilayers, 2D Mater. 5, 035021 (2018).
[9] Fengcheng Wu, Timothy Lovorn, A.H. MacDonald, Topological Exciton Bands in Moiré Heterojunctions, Phys. Rev. Lett. 118, 147401 (2017).
[10] Hongyi Yu, Gui-Bin Liu, Jianju Tang, Xiaodong Xu, Wang Yao, Moiré excitons: From programmable quantum emitter arrays to spin-orbit–coupled artificial lattices, Sci. Adv. 3, e1701696 (2017).
[11] Chendong Zhang, Chih-Piao Chuu, Xibiao Ren, Ming-Yang Li, Lain-Jong Li, Chuanhong Jin, Mei-Yin Chou, Chih-Kang Shih, Interlayer couplings, Moiré patterns, and 2D electronic superlattices in MoS$_2$/WSe$_2$ hetero-bilayers, Sci. Adv. 3, e1601459 (2017).

[12] Jonathan S. Alden, Adam W. Tsen, Pinshane Y. Huang, Robert Hovden, Lola Brown, Jiwoong Park, David A. Muller, and Paul L. McEuen, Strain solitons and topological defects in bilayer graphene, PNAS 110, 11256 (2013).

[13] C.R. Woods, L. Britnell, A. Eckmann, R.S. Ma, J.C. Lu, H.M. Guo, X. Lin, G.L. Yu, Y. Cao, R.V. Gorbachev, A.V. Kretinin, J. Park, L.A. Ponomarenko, M.I. Katsnelson, Yu.N. Gornostyrev, K. Watanabe, T. Taniguchi, C. Casiraghi, H-J. Gao, A.K. Geim, and K.S. Novoselov, Commensurate–incommensurate transition in graphene on hexagonal boron nitride, Nature Phys. 10, 451 (2014).

[14] Hyobin Yoo, Rebecca Engelke, Stephen Carr, Shiang Fang, Kuan Zhang, Paul Cazeaux, Suk Hyun Sung, Robert Hovden, Adam W. Tsen, Takashi Taniguchi, Kenji Watanabe, Gyu-Chul Yi, Miyoung Kim, Mitchell Luskin, Ellad B. Tadmor, Efthimios Kaxiras, and Philip Kim, Atomic and electronic reconstruction at the van der Waals interface in twisted bilayer graphene, Nature Mat. 18, 448 (2019).

[15] Astrid Weston, Yichao Zou, Vladimir Enaldiev, Alex Summerfield, Nicholas Clark, Viktor Zólyomi, Abigail Graham, Celal Yelgel, Samuel Magorrian, Mingwei Zhou, Johanna Zultak, David Hopkinson, Alexei Barinov, Thomas H. Bointon, Andrey Kretinin, Neil R. Wilson, Peter H. Beton, Vladimir I. Fal’ko, Sarah J. Haigh, and Roman Gorbachev, Atomic reconstruction in twisted bilayers of transition metal dichalcogenides, Nature Nano. 15, 592 (2020).

[16] Matthew R. Rosenberger, Hsun-Jen Chuang, Madeleine Phillips, Vladimir P. Oleshko, Kathleen M. McCreary, Saujan V. Sivaram, C. Stephen Hellberg, and Berend T. Jonker, Twist Angle-Dependent Atomic Reconstruction and Moiré Patterns in Transition Metal Dichalcogenide Heterostructures, ACS Nano 14, 4550 (2020).