

Modeling and Simulation of Exciton Dynamics in Transition Metal Dichalcogenide Heterostructures

Rohit Nimje*, Vignesh S*, Athira S†, Ashutosh Mahajan*‡

* Centre for Nanotechnology Research, Vellore Institute of Technology, Vellore-632 014, India.

†Department of Physics, School of Advanced Sciences, Vellore Institute of Technology, Vellore-632 014, India.

‡ Email: ashutosh.mahajan@vit.ac.in

Abstract—This work develops a numerical model to estimate binding energy, exciton energy, and radiative lifetime in layered transition metal dichalcogenide (TMD) materials. Electron and hole effective masses obtained from DFT band structures are used in the quantum well model to describe out-of-plane carrier confinement. The in-plane exciton motion is solved using a finite-difference Wannier-Mott equation with Keldysh screening. These wavefunctions are then used to calculate electron-hole overlap, oscillator strength, intrinsic radiative lifetime, thermally averaged effective lifetime, and absorption spectra. The model is tested for monolayer, multilayer, and heterobilayer TMDs. The calculated exciton transition energies, binding energies, and lifetime trends are compared with reported PL and theoretical values. The results indicate that indirect interlayer excitons have lower binding energies and reduced electron-hole overlap. This reduced overlap leads to longer recombination lifetimes, which are relevant for exciton transport in optoelectronic devices.

Index Terms—Indirect exciton, exciton lifetime, binding energy, Keldysh potential, TMD heterobilayers, excitonic devices.

I. INTRODUCTION

Two-dimensional (2D) transition metal dichalcogenides (TMDs) are widely studied for optoelectronic and excitonic device applications because of their strong optical response, reduced dielectric screening, and pronounced quantum confinement effects [1]–[3]. In monolayer TMDs, these features produce excitons with large binding energies that recombine radiatively even at 300 K [4]. Adding layers shifts the electronic structure toward the bulk limit, leading to changes in exciton transition energy, binding energy, and recombination dynamics.

Exciton lifetime directly controls carrier relaxation and optical response in layered TMDs. Experimental studies based on photoluminescence (PL) and time-resolved PL spectroscopy have shown that exciton dynamics strongly depend on thickness, dielectric environment, and indirect coupling [5]. In multilayer systems, enhanced dielectric screening and reduced confinement modify the exciton wavefunction overlap and radiative decay. In heterobilayers, spatial separation of electrons and holes can further suppress recombination and lead to indirect exciton lifetimes ranging from microseconds to hundreds of microseconds, depending on temperature, dielectric environment, and heterostructure configuration [6]–[9].

Although first-principles approaches such as GW-BSE can accurately describe excitonic properties, their application in systematic research that depends on thickness and structure is still computationally challenging [10]. Effective mass

approaches combined with the Keldysh interaction offer a simpler physics-based alternative, provided that reliable material parameters are available. In particular, growth-direction electron and hole effective masses and confinement-related parameters are essential for describing quantization and in-plane exciton motion in layered TMDs.

In this work, we present a DFT-informed exciton framework that connects material parameters to exciton energy, binding energy, and lifetime. QuantumATK band structures provide anisotropic electron and hole effective masses. These masses are used in the quantum-well model for out-of-plane confinement and in the Wannier-Mott solver for in-plane exciton motion. The calculated wavefunctions directly determine the electron-hole overlap, oscillator strength, radiative lifetime, and thermally averaged effective lifetime. This enables comparison across monolayers, multilayers, and heterobilayer TMDs. Particular emphasis is placed on indirect excitons in heterobilayer TMDs. In these systems, spatial separation of electrons and holes reduces oscillator strength and extends the recombination lifetime. This helps identify suitable heterostructures for exciton transport and gate-controlled excitonic devices.

II. MODELS

The excitonic Hamiltonian for TMD heterobilayers is

$$H = H_e + H_h + E_c + T_X + V_K(\rho), \quad (1)$$

Here, T_X denotes the in-plane kinetic energy and E_c is the conduction-band edge. The terms H_e and H_h are the confinement-direction Hamiltonians for the electron and hole, respectively, while $V_K(\rho)$ is the Keldysh interaction. H_e and H_h are solved with Dirichlet boundary conditions for the confined states $\psi_e(z)$, $\psi_h(z)$. DFT band gaps and anisotropic masses (K -point parabolic fit) give the reduced mass $\mu = (m_e^{-1} + m_h^{-1})^{-1}$, which enters the 2D Wannier-Mott equation,

$$\left[-\frac{\hbar^2}{2\mu} \left(\frac{\partial^2}{\partial \rho^2} + \frac{1}{\rho} \frac{\partial}{\partial \rho} - \frac{l^2}{\rho^2} \right) + V_K(\rho) \right] \psi_n(\rho) = E_{X,n} \psi_n(\rho) \quad (2)$$

This equation is solved in MATLAB on a uniform ρ grid. A Neumann condition is applied at $\rho = 0$, and a vanishing boundary condition is used at the outer edge. The exciton energy is obtained as $E_X = E_{QP} - E_B$, where E_B is the binding energy, E_{QP} is the quasiparticle gap, and n denotes

the exciton state. For a monolayer direct exciton, the Keldysh potential is

$$V_K(\rho) = -\frac{\pi e^2}{\chi_{2D}} \left[H_0\left(\frac{\kappa\rho}{r_0}\right) - Y_0\left(\frac{\kappa\rho}{r_0}\right) \right] \quad (3)$$

where χ_{2D} is the 2D polarizability, r_0 is the screening length, κ is the environmental dielectric constant, with Y_0 and H_0 denote the second-kind Bessel and Struve functions of zeroth order. For bilayer and indirect interlayer excitons, the vertical separation is included via $\varrho = \sqrt{\rho^2 + d^2}$, where d is the vertical separation listed in Table II, with $d = 0$ for the monolayer. The screening is summed over both layers,

$$V_K(\varrho) = -\frac{\pi e^2}{2\gamma} \left[H_0\left(\frac{\kappa\varrho}{\gamma}\right) - Y_0\left(\frac{\kappa\varrho}{\gamma}\right) \right] \quad (4)$$

where $\gamma = (r_0^{(1)} + r_0^{(2)})$, $r_0^{(1)}$ and $r_0^{(2)}$ are the monolayer screening lengths. The radiative decay rate Γ_0 and intrinsic lifetime are

$$\tau_{\text{rad}} = \frac{\hbar}{2\Gamma_0} \quad \Gamma_0 = \frac{\pi e^2}{\sqrt{\epsilon_b} m_0 c} f_v \frac{S}{A_{\text{uc}}} \quad (5)$$

where $S = \pi a_B^2$ is the coherence area (a_B the Bohr radius), A_{uc} the unit cell area, and ϵ_b the in-plane dielectric constant. The oscillator strength combines the interband dipole d_{cv} , the out-of-plane overlap, and the wavefunction at zero separation,

$$f_v = \frac{2m_0 E_X d_{cv}^2}{\hbar^2} \left| \int \psi_e(z) \psi_h(z) dz \right|^2 |\psi(0)|^2 \quad (6)$$

Only excitons inside the light cone radiate. The remaining excitons are momentum-dark states. Since excitons thermalize within ~ 1 -2 ps, only a small bright fraction occupies the cone at room temperature, giving the effective lifetime

$$\tau_{\text{eff}}(T) = \frac{3Mk_B T c^2}{E_X^2 \epsilon_b} \tau_{\text{rad}} \quad (7)$$

where M is the exciton mass. A large out-of-plane overlap gives a large oscillator strength and fast recombination, desirable for high-speed emission, whereas type-II indirect (e_2h_1) interlayer excitons have reduced overlap and longer effective lifetime.

The interband absorption is obtained by summing the bright ($l = 0$) states, with linewidth Γ_0 ,

$$\alpha(\omega) = \frac{1}{\hbar\omega} \sum_{n(l=0)} f_{v,n} \sum_{i=A,B} g_i \frac{\Gamma_0}{(E_n^i - \hbar\omega)^2 + \Gamma_0^2} \quad (8)$$

with $g_A = 1$, $g_B = \eta$. Here $E_n^A = E_{X,n}$ is the A-exciton energy and $E_n^B = E_n^A + \Delta_{AB}$ the B-exciton energy, and Δ_{AB} denotes the spin-orbit splitting between the A and B excitons ($E_X^A - E_X^B$). Only s states carry finite contact amplitude $|\psi(0)|^2$ and are bright, p and d states are dark.

III. SIMULATION FRAMEWORK

DFT-derived parameters are used as inputs to the numerical exciton solver. The solver computes binding energy, exciton transition energy, radiative lifetime and absorption in monolayer, multilayer, and interlayer TMD heterobilayers using material parameters listed in Tables I and II. Fig. 1a shows the device geometry, where the electron and hole either stay in the

same layer as a direct exciton or separate across the interface as an indirect exciton. The type-II alignment is treated as a double quantum well (Fig. 1b), with the band offsets defining the confinement regions. Band structures were computed in QuantumATK using LDA, SG15 pseudopotentials, and a Medium basis set. The in-plane and out-of-plane effective masses were then extracted by parabolic fitting near the K point as shown in Fig. 1d. The in-plane masses set the reduced mass for the Wannier-Mott equation and the out-of-plane masses enter the confinement calculation.

The electron and hole envelope functions follow from the one-dimensional effective mass Schrödinger equation along the out-of-plane direction with Dirichlet boundary conditions. Their overlap (Fig. 1c) enters the radiative rate quadratically, so the reduced overlap between e_2h_1 excitons gives a longer lifetime. The in-plane states follow from the radial Wannier-Mott equation with the Keldysh potential. The wavefunctions in Fig. 1e show a localized $1s$ ground state and more extended excited states. Figure 1f compares the direct and indirect potentials, where the weaker indirect e_2h_1 potential reflects the finite vertical separation that lowers the indirect binding energy.

TABLE I: Material parameters used for monolayer TMDs reported in previous work [11]. Multilayer parameters are scaled with layer number N .

Parameters	Unit	Monolayer TMDs			
		WS ₂	WSe ₂	MoS ₂	MoSe ₂
E_{QP}	(eV)	2.51	2.11	2.38	2.03
T	(nm)	0.60	0.75	0.65	0.70
V_e	(eV)	3.93	3.61	4.28	3.91
V_h	(eV)	5.48	4.86	5.87	5.24
ϵ_b		14.0	15.3	15.4	16.8
r_0	(nm)	4.017	4.757	4.468	5.316
χ_{2D}	(nm)	7.58	9.02	8.29	10.34
$m_{\parallel}^e (m_0)$		0.32	0.34	0.47	0.55
$m_{\parallel}^h (m_0)$		0.35	0.36	0.54	0.59
$\mu_{\parallel}^e$		0.167	0.175	0.249	0.284
$m_{\perp}^e (m_0)$		0.665	2.401	0.790	2.227
$m_{\perp}^h (m_0)$		2.851	5.196	2.441	5.556
d_{cv}	(nm)	0.60	0.60	0.47	0.50

TABLE II: Material parameters for the indirect (e_2h_1) exciton study in heterobilayer TMDs.

Parameters	Unit	TMD heterobilayers					
		WS ₂	MoS ₂	WSe ₂	MoS ₂	WSe ₂	MoSe ₂
Layer-1 (hole)		WS ₂	MoS ₂	WSe ₂	MoS ₂	WSe ₂	MoSe ₂
Layer-2 (electron)		WS ₂	MoS ₂	WSe ₂	MoS ₂	WSe ₂	MoSe ₂
E_{QP}	(eV)	2.15	1.89	2.08	2.26	1.95	
$T_1 + T_2$	(nm)	1.35	1.35	1.40	1.25	1.45	
V_{e1}	(eV)	3.93	4.28	3.61	4.28	3.91	
V_{e2}	(eV)	3.61	3.91	4.28	3.93	3.61	
V_{h1}	(eV)	5.48	5.87	4.86	5.87	5.24	
V_{h2}	(eV)	4.86	5.24	5.87	5.48	4.86	
ΔE_C	(eV)	0.32	0.37	0.67	0.35	0.30	
ΔE_V	(eV)	0.62	0.63	1.01	0.39	0.38	
d	(nm)	0.62	0.62	0.63	0.60	0.64	
$(\epsilon_{b1} + \epsilon_{b2})/2$		14.7	16.1	15.4	14.7	16.1	
$m_{\parallel 2}^e (m_0)$		0.36	0.56	0.48	0.32	0.36	
$m_{h1}^e (m_0)$		0.36	0.58	0.45	0.58	0.65	
$\mu_{\parallel 2}^e$		0.180	0.285	0.232	0.206	0.232	
$m_{e2}^e (m_0)$		2.400	2.220	0.790	0.665	2.400	
$m_{h1}^h (m_0)$		2.851	2.441	5.196	2.441	5.556	
d_{cv}	(nm)	0.60	0.63	0.60	0.8385	0.70	

The exciton radiative lifetime is first verified using the Al-GaAs/GaAs double quantum well case. The field-dependent increase in lifetime in Fig. 2d confirms that reduced electron-hole overlap correctly slows radiative recombination in the indirect limit.

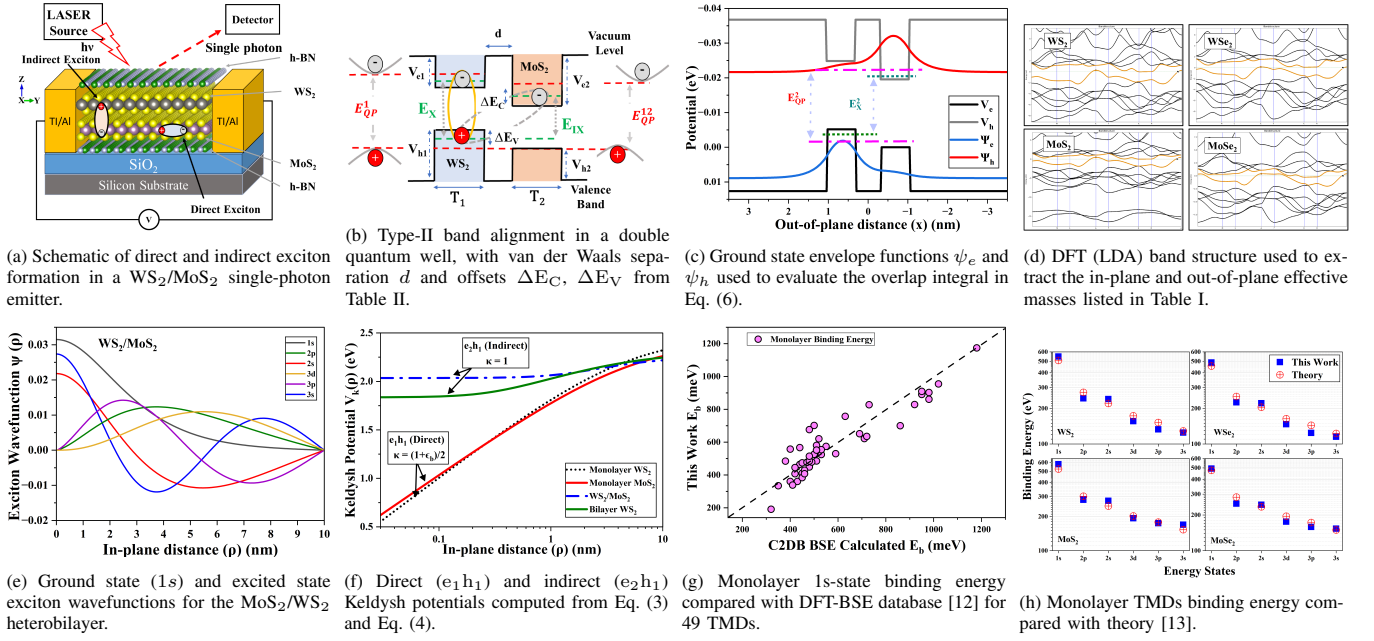


Fig. 1: Exciton formation in monolayer, multilayer, and $\text{WS}_2/\text{MoSe}_2$ heterobilayer systems, modeled through band alignment, effective masses, wavefunctions, Keldysh potentials, and binding energies.

IV. RESULTS AND DISCUSSION

The exciton-energy calculation is first validated for monolayer and multilayer WS_2 . For monolayer WS_2 , the calculated $1s - 5s$ transition energies in Fig. 2a agree closely with the reported PL peaks [4], particularly for the low-lying states. The small deviation in higher states is expected. They are weakly bound, extend over a larger radius, and sample the dielectric environment more strongly, making them sensitive to the screening length. In the multilayer case, the $1s$ and $2s$ energies in Fig. 2b decrease with layer number and then approach saturation. Two effects drive this trend, with the quasiparticle gap dropping toward its bulk value while stronger dielectric screening weakens the electron-hole attraction as more layers are added.

The binding energies show a similar trend across the DFT-BSE data for 49 TMD materials taken from the 2D material database, as shown in Fig. 1g. Exact agreement is not expected because the effective-mass description does not include band non-parabolicity and local-field effects. For monolayer TMDs, the excited state plot in Fig. 1h shows a clear reduction in binding energy from the $1s$ state to the higher states, because the higher states have a larger spatial extent and a correspondingly weaker electron-hole attraction. The non-hydrogenic spacing of the series is a direct signature of the nonlocal Keldysh screening. The monolayer binding energy comparison in Fig. 2c further shows that WS_2 , WSe_2 , MoS_2 , and MoSe_2 lie close to the reported DFT, existing model, and SVM values. The material-wise variation arises mainly from the reduced mass and the 2D polarizability χ_{2D} , with the heavier Mo-based compounds showing the larger binding energies.

For monolayer WS_2 , WSe_2 , MoS_2 , and MoSe_2 , the effective lifetimes at 300 K in Fig. 2e fall in the same range as the reported DFT and PL decay values. For multilayer TMDs,

the lifetime in Fig. 2g decreases with layer number and then saturates. This follows the thickness-dependent screening trend observed in the exciton-energy results. The changes in band gap, screening, and wavefunction overlap modify the transition energy and oscillator strength. The indirect lifetimes in Fig. 2h exceed the monolayer direct exciton values by orders of magnitude. The electron and hole sit in adjacent layers, so their envelope functions overlap only weakly across the van der Waals gap. This spatial separation suppresses the overlap integral in Eq. (6) and slows radiative recombination, in agreement with the long-lived indirect (e_2h_1) excitons reported experimentally [7], [8]. The spread across the five heterobilayers comes mainly from differences in band alignment, interlayer distance d , and reduced mass, which set the overlap and the thermal occupation factor in Eq. (7).

The absorption spectra in Fig. 2f confirm the calculated exciton transition energies. For monolayer TMDs, the dominant $1s$ peak agrees well with the reported PL energies marked in red, while the weaker $2s$ and $3s$ peaks appear at higher energies. Only s -type states are optically bright because the oscillator strength depends on $|\psi(0)|^2$. For heterobilayers, the spectra show interlayer e_2h_1 excitonic transitions. MoS_2/WS_2 and $\text{MoSe}_2/\text{WSe}_2$ are compared with the reported GW-BSE peak positions of Ref. [18], near 1.83 eV and 1.60 eV, respectively. Higher excited-state peaks arise from weaker binding, while their lower intensity results from reduced electron-hole overlap at zero separation.

V. CONCLUSION

A Wannier-Mott exciton model was developed for 2D TMDs by combining QuantumATK-derived parameters with Keldysh screening and oscillator-strength-based recombination. The model predicts binding energy, transition energy,

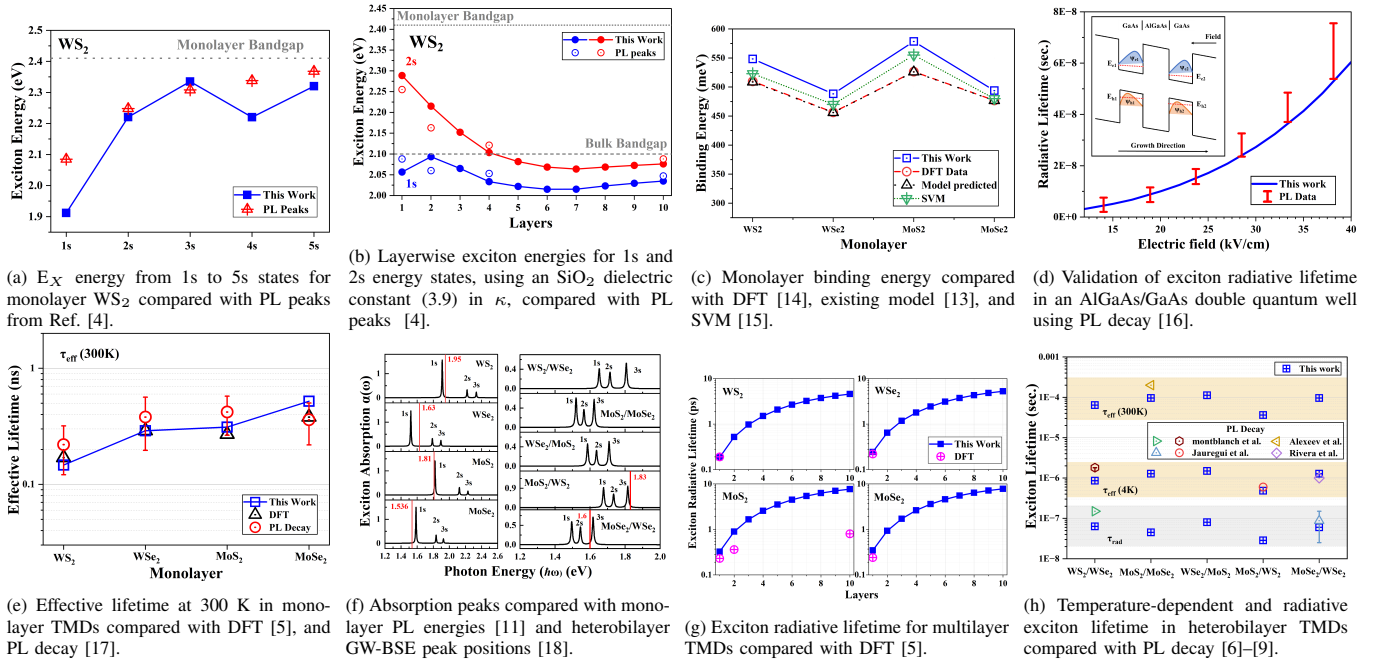


Fig. 2: Exciton energies, binding energies, absorption spectra, and radiative lifetimes across monolayer, multilayer, and heterobilayer TMDs.

effective lifetime, and absorption peaks across monolayer, multilayer, and heterobilayer TMDs. The calculated trends agree reasonably with reported data and show that the approach can estimate exciton lifetimes at low computational cost.

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