

Full-Band Semi-Classical Monte Carlo Simulation of Layer Number-Dependent Hole Transport in W- and Mo-Based Transition Metal Dichalcogenides

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Abstract—Using *ab initio*-based full-band semi-classical Monte Carlo simulations, we investigate intrinsic phonon-limited hole transport in monolayer, bilayer, and trilayer MoS₂, WS₂, and WSe₂. Increasing layer number shifts valence-band states near Γ upward, driving a transition from K/K' valley-dominated transport in monolayer to Γ valley-dominated transport in few-layer. Direct K–K' intervalley scattering is suppressed in all monolayers, however, in monolayer MoS₂ the small K– Γ energy separation allows the heavy Γ valley to contribute to transport, reducing the mobility. In contrast, monolayer WS₂ and WSe₂ maintain high mobility through K/K' valley-dominated transport with minimal involvement of the heavy Γ valley. In bilayers and trilayers, the mobility is governed by the Γ valley curvature and the energetic proximity of secondary Γ valleys, rather than by Γ valley occupation alone. These results identify valley alignment, valley effective mass, and valley-resolved phonon scattering as key descriptors governing the hole transport in MoS₂, WS₂, and WSe₂.

Keywords—transition metal dichalcogenides, hole transport, semi-classical Monte Carlo, electron-phonon scattering, first-principles calculations

I. INTRODUCTION

Two-dimensional (2D) transition metal dichalcogenides (TMDs) are promising channel materials for aggressively scaled field-effect transistors, as their atomically thin bodies provide strong electrostatic control of the gate over the channel [1]. In many 2D TMDs, hole transport is intrinsically determined by a complex multivalley valence band structure. In particular, for molybdenum- and tungsten-based TMDs such as MoS₂, WS₂, and WSe₂, the relative energies, band curvatures, and spin splittings of the valence bands near the K/K' and Γ points depends strongly on both the material and the layer number [2], [3]. Consequently, the hole mobility cannot be accurately predicted from the effective mass of a single valley alone.

This work was supported by Samsung Advanced Institute of Technology, Samsung Electronics Co., Ltd. This work was supported by K-CHIPS (Korea Collaborative & High-tech Initiative for Prospective Semiconductor Research) (RS-2026-25523338, 2410018350, 26085-45FC) funded by the Ministry of Trade, Industry & Energy (MOTIE, Korea). This work was supported by the National Research Foundation of Korea (NRF) grant funded by the Korea government (MSIT) (RS-2026-25542094).

Intrinsic phonon-limited calculations provide a useful basis for evaluating the intrinsic limits of TMDs by excluding extrinsic effects such as contact resistance, impurities, defects, and dielectric or substrate interactions. Although earlier first-principles studies presented the importance of electron-phonon scattering in monolayer TMDs transport [4], a systematic comparison of MoS₂, WS₂, and WSe₂ across different layer numbers within a unified full-band transport framework remain limited. In this work, we employ an *ab initio*-based semi-classical Monte Carlo (SCMC) method to correlate layer-dependent valence-band structures with low-field mobility and field-dependent drift velocity, thereby identifying the valley-dependent transport mechanisms which govern the performance of p-type TMDs channel.

II. COMPUTATIONAL METHODS

A. First-principles Calculations

Electronic band structures, phonon dispersions, and electron–phonon matrix elements were obtained through the Quantum ESPRESSO–EPW calculation [5], [6]. For each material and layer number, the atomic structures were first relaxed with Grimme-D3 van der Waals corrections, followed by self-consistent-field calculations, D3-corrected force-constant calculations, density-functional perturbation theory (DFPT) phonon calculations, non-self-consistent calculations, and EPW interpolation. Fully relativistic SG15 optimized norm-conserving Vanderbilt (ONCV) pseudopotentials [7] were used together with noncollinear spin–orbit coupling. Isolated 2D boundary conditions and pairwise Grimme-D3 van der Waals corrections [8] were applied consistently throughout the electronic and phonon calculations.

The plane-wave cutoff and self-consistent k -point mesh were converged to within 1 meV/atom. The EPW coarse grids were set to $12 \times 12 \times 1$ for electrons and $6 \times 6 \times 1$ for phonons. Dipole and quadrupole long-range corrections were included in EPW using the polar, quadrupole, and 2D treatments [9]. The dynamical quadrupoles were calculated in ABINIT and imported into EPW. Wannierization was performed using the selected columns of the density matrix (SCDM) method [10]. The electronic bands, phonon bands, and electron–phonon matrix elements were then interpolated onto $90 \times 90 \times 1$ fine k - and q -point grids. The EPW-interpolated bands and electron–

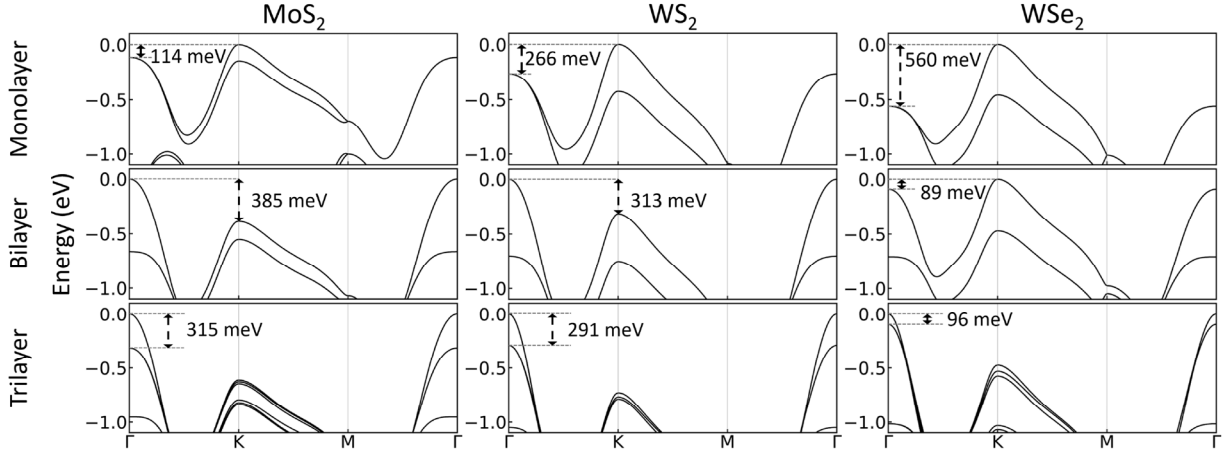


Fig. 1. Band structures of monolayer, bilayer, and trilayer MoS₂, WS₂, and WSe₂ along the Γ -K-M- Γ high-symmetry path. The energy in each panel is referenced to the corresponding valence-band maximum, set to 0 eV.

phonon matrix elements were used as input to the in-house SCMC simulations.

B. Electron-Phonon Scattering Rates

The phonon-limited transition rates were evaluated over the full Brillouin zone from interpolated full-band electronic states and electron-phonon matrix elements. To reduce memory usage, the EPW-generated matrix elements were stored in the irreducible-wedge representation used by EPW and unfolded through the corresponding symmetry mapping during SCMC post-processing. For a valence-band transition from an initial electronic state $(n, \mathbf{k})$ to a final state $(m, \mathbf{k} + \mathbf{q})$ mediated by phonon mode ν at $\mathbf{q}$, the hole scattering-rate contribution was computed from Fermi's golden rule as

$$\Gamma_{n, \mathbf{k} \rightarrow m, \mathbf{k} + \mathbf{q}}^{\nu} = \frac{2\pi}{\hbar} |g_{mn\nu}(\mathbf{k}, \mathbf{q})|^2 \left(N_{q\nu} + \frac{1}{2} \pm \frac{1}{2} \right) \times \delta[\varepsilon_{n, \mathbf{k}} \pm \hbar\omega_{q\nu} - \varepsilon_{m, \mathbf{k} + \mathbf{q}}]. \quad (1)$$

Here, $g_{mn\nu}(\mathbf{k}, \mathbf{q})$ is the electron-phonon matrix element, $N_{q\nu}$ is the Bose-Einstein occupation, $\hbar\omega_{q\nu}$ is the phonon energy, and ε denotes the electronic band energy within the transport window. Since Eq. (1) is expressed in terms of electronic band energies, the upper and lower signs correspond to hole emission and absorption, respectively. The energy-conservation delta functions were evaluated directly in momentum space using triangle-based Gilat-Raubenheimer (GR) integration [11], which accounts for the variation of final-state energies within each triangle element.

Final-state availability was evaluated from the instantaneous occupation of the Monte Carlo ensemble rather than from an equilibrium Fermi-Dirac distribution. The Brillouin zone was divided into the same $90 \times 90 \times 1$ cells used for the full-band transport grid. For a candidate final state in band m and cell C_l , the hole occupation was estimated as

$$f_h(m, C_l, t) = \min \left[\frac{N_{MC}(m, C_l, t)}{N_{max}(m, C_l)}, 1 \right] \quad (2)$$

where $N_{MC}(m, C_l, t)$ is the number of Monte Carlo holes occupying the final band-cell at time t , and $N_{max}(m, C_l)$ is the corresponding particle count for complete occupation of the available single-particle states. A proposed scattering event into this cell was accepted with probability $1 - f_h$. Rejected

events were treated as self-scattering events, leaving the carrier state unchanged. A hole density of $1 \times 10^{10} \text{ cm}^{-2}$ was used to set the particle weight in the occupation estimate.

C. Full-band SCMC Transport Simulation

Hole transport under in-plane electric fields applied along the (100) direction was simulated using the full-band SCMC method. Each simulation used 100,000 particles and a time step of 1.0×10^{-2} ps. During free flight, carriers were accelerated by applied electric field, and scattering events were selected from the cumulative phonon-limited transition rates obtained from Eq. (1). For each electric field, the drift velocity was calculated by averaging the carrier velocity projected onto the field direction over the Monte Carlo ensemble and the steady-state time window after removing the initial transient. Statistical uncertainty was estimated using block averages, and low-field mobility was extracted from the linear-response regime.

III. RESULTS AND DISCUSSION

The upper valence bands of the studied TMDs are located at the K/K' and Γ points, and their relative energies vary strongly with the layer number. As shown in Fig. 1, the valence-band maximum (VBM) is located at the K valley in monolayers. As the layer number increases, however, the valence-band states near the Γ point shift upward relative to those near the K valley. This thickness-dependent evolution is consistent with previous theoretical and spectroscopic studies of few-layer TMDs [2], [3]. It is commonly attributed to the

TABLE I. LAYER-DEPENDENT VALLEY EFFECTIVE MASSES (M^*/M_0)

		Effective Mass		
		MoS ₂	WS ₂	WSe ₂
1L	Γ_1	3.19	2.52	3.93
	K ₁	0.55	0.39	0.41
2L	Γ_1	1.02	0.74	1.10
	K ₁	0.55	0.39	0.42
3L	Γ_1	0.66	0.37	0.36
	Γ_2	1.29	0.60	0.47

Γ_1 and Γ_2 denote the first and second Γ valleys, respectively, and K₁ denotes the first K valley.

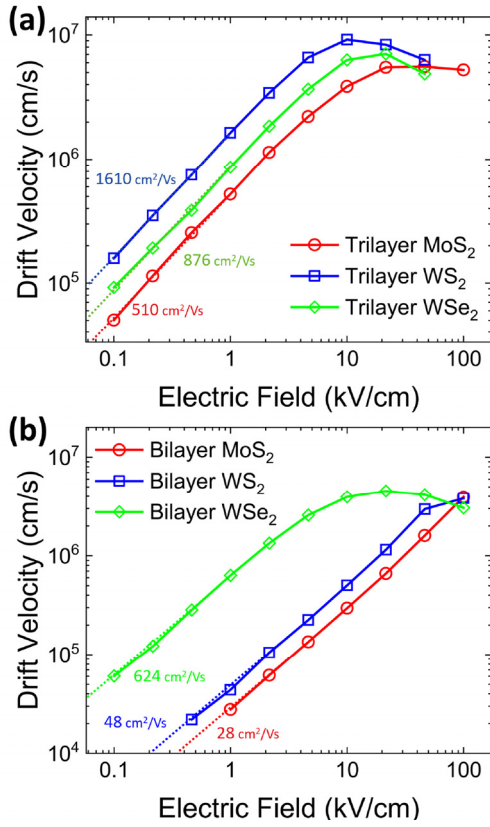


Fig. 2. Drift velocity as a function of electric field for (a) trilayer and (b) bilayer MoS₂, WS₂, and WSe₂, on a log-log scale. The values indicated in each panel represent the low-field mobilities extracted from the linear-response regime.

different orbital characters of the relevant valleys: the states near Γ contain stronger out-of-plane components and are therefore more sensitive to interlayer coupling, whereas the states near K are dominated by more localized in-plane orbitals. Consequently, the K valley remains the VBM in monolayer MoS₂, WS₂, and WSe₂. In bilayer MoS₂ and WS₂, the Γ valley becomes the highest valence-band valley, while bilayer WSe₂ retains a K valley VBM with a relatively small K- Γ separation. For all trilayers, the Γ valley becomes the VBM.

The layer-dependent valley shift is accompanied by a pronounced change in the curvature of the Γ valleys. As summarized in Table 1, the Γ_1 effective mass decreases substantially as the layer number increases, whereas the K_1 effective mass remains nearly unchanged. Therefore, once the Γ valley moves close to or above the K valley, the transport behavior becomes sensitive not only to the VBM location but also to the Γ valley effective mass and the occupation of nearby secondary valleys.

The drift-velocity characteristics of bilayer and trilayer are shown in Fig. 2. In these few-layers, several upper valence-band states are close in energy, and the mobility trends are mainly governed by the effective mass of the occupied valleys and the number of accessible intervalley scattering channels. For trilayers, the Γ valley is the VBM for all three materials. Trilayer MoS₂ shows the lowest mobility among the trilayers because its Γ_1 valley remains relatively heavy. In contrast, trilayer WS₂ benefits from a much lighter Γ_1 valley and exhibits the highest trilayer mobility. Although trilayer WSe₂ has a similarly light Γ_1 mass, its mobility is lower than that of

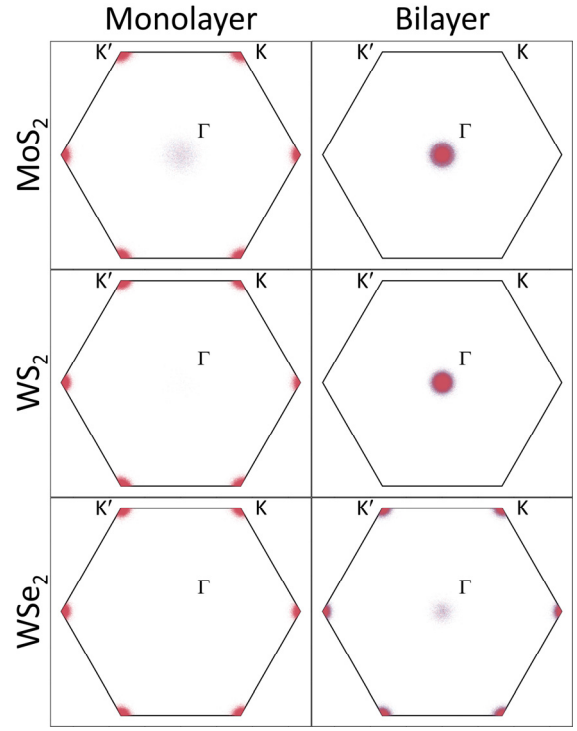


Fig. 3. Hole distributions in the first Brillouin zone for monolayer and bilayer MoS₂, WS₂, and WSe₂ at an applied electric field of 1 kV/cm. Red and blue points denote holes in the highest and second-highest valence bands, respectively; Γ , K, and K' mark the zone-center and two inequivalent corner valleys.

trilayer WS₂. This reduction is attributed to the small separation between the first and second Γ valleys, which allows the Γ_2 valley to become substantially occupied and introduces additional intervalley scattering channels. Thus, the trilayer mobility is not determined by the band-edge effective mass alone, but also by the energetic proximity of secondary valleys.

The bilayer results in Fig. 2(b) show that the mobility trend depends strongly on whether transport is dominated by the Γ valley or the K valley. In bilayer MoS₂ and WS₂, the Γ valley is the dominant occupied valley. Bilayer MoS₂ exhibits particularly low mobility because transport occurs mainly through the relatively heavy Γ_1 valley. Bilayer WS₂ also transports primarily through the Γ valley, but its lighter Γ_1 mass leads to higher mobility than in bilayer MoS₂. Bilayer WSe₂ differs from the other two materials because the K valley remains the VBM, as indicated in Fig. 1. The hole distribution in Fig. 3 shows that the Γ valley is still partially occupied, but the substantial K valley occupation prevents the mobility from being degraded as strongly as in the Γ -dominated bilayers.

Monolayer requires an additional interpretation beyond the effective mass of the valley. As shown in Fig. 4, monolayer WS₂ and WSe₂ exhibit much higher drift velocities than monolayer MoS₂. Part of this difference comes from the valley occupation shown in Fig. 3. In monolayer MoS₂, the Γ valley lies relatively close to the K valley VBM and has a very heavy effective mass, so even partial occupation near the Γ region can strongly reduce the mobility. In contrast, monolayer WS₂ and WSe₂ mainly occupy the K/K' valleys, while the heavy Γ valleys lie farther below the VBM. However, the large mobility enhancement in monolayer WS₂

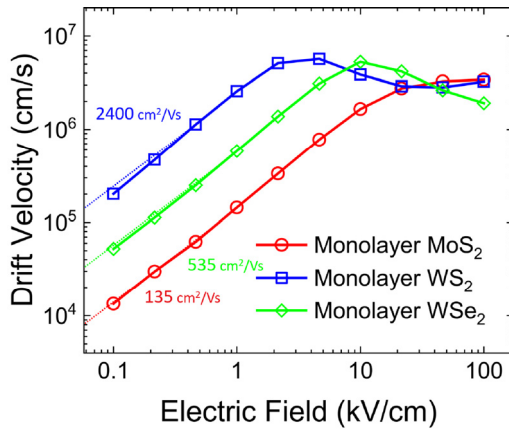


Fig. 4. Drift velocity as a function of electric field for monolayer MoS₂, WS₂, and WSe₂ on a log-log scale. The indicated values represent the low-field mobilities extracted from the linear-response regime.

and WSe₂ cannot be explained by effective mass alone, because their K valley effective masses differ only moderately from that of monolayer MoS₂.

The high mobility in monolayer can be explained by the scattering rates shown in Fig. 5. First, direct K–K' intervalley scattering is strongly suppressed in all three monolayers due to spin-dependent valley states, which are produced by spin-orbit coupling at the K and K' valleys. The critical difference in mobility arises from the Γ valley's position relative to the K valley. In MoS₂, the small energy separation between Γ and K valleys allows holes to access the Γ valley by the intervalley scattering; the resulting partial occupation of the heavier Γ valley significantly reduces mobility. In contrast, the Γ valleys in WS₂ and WSe₂ lie much farther below the K valley. This deep energy separation confines hole transport strictly to the K/K' valleys, minimizing effective scattering pathways to the Γ valley. Therefore, the superior mobility of WS₂ and WSe₂ originates from the synergistic effects of dominant K/K' valley occupation, suppressed direct K–K' scattering, and relatively light K valley effective masses.

Overall, the layer dependence of hole transport in these TMDs is governed by the combined effects of valley alignment, valley effective mass, valley occupation, and intervalley scattering. In bilayers and trilayers, the upward shift of the Γ valley makes the Γ valley effective mass and the

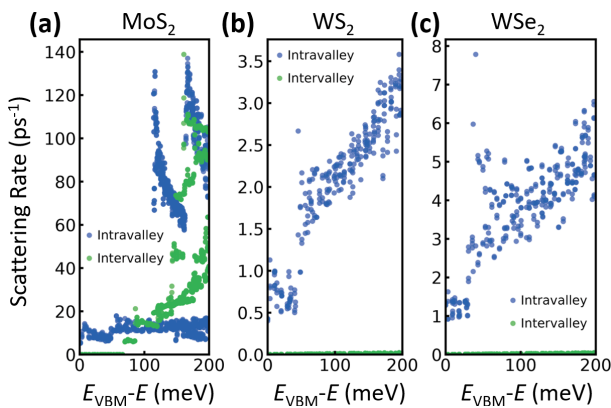


Fig. 5. Intravalley and intervalley scattering rates as functions of hole energy measured from the valence-band maximum, $E_{\text{VBM}} - E$, for monolayer (a) MoS₂, (b) WS₂, and (c) WSe₂.

occupation of nearby secondary Γ valleys the dominant factors. In monolayers, by contrast, these Γ valley-related limitations are much weaker in WS₂ and WSe₂ because holes remain mainly in the K/K' valleys over the relevant energy range. Direct K–K' intervalley scattering is suppressed by the spin-valley character of the K/K' valley states, while scattering to the heavy Γ valley is also limited by the large K– Γ valley energy separation. As a result, their mobility becomes much higher than expected from the effective mass alone.

IV. CONCLUSION

The full-band SCMC simulations show that the layer-dependent hole transport of MoS₂, WS₂, and WSe₂ is controlled by the interplay between valence-band valley alignment and phonon-mediated scattering. Increasing the layer number raises valence-band states near Γ relative to states near K, making Γ valley transport increasingly important in bilayers and trilayers. In monolayers, the high mobilities of WS₂ and WSe₂ originate from dominant K/K' occupation and strongly suppressed intervalley scattering, whereas MoS₂ is limited by stronger scattering and partial occupation of a heavy Γ valley. In few-layer structures, the mobility is further affected by the Γ valley curvature and the energetic proximity of secondary valleys. These results indicate that valley alignment, band-edge curvature, and valley-resolved scattering pathways must be considered when selecting TMDs channels for high-performance p-type devices.

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