

Tuning Band Tails in Mono- and Multilayered Transition-Metal Dichalcogenides: A Detailed Assessment and a Quick-Reference Guide

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Transition-metal dichalcogenides (TMDs) are promising candidates for a wide variety of ultrascaled electronic, optoelectronic, and quantum-computing applications. The electronic density of states exponentially decaying into the band gap (also known as the band tail) has a strong impact on the performance of TMD applications. In this work, the band tails of various TMD monolayer and multilayer systems is predicted with density-functional-theory-based nonequilibrium Green's functions when placed on several dielectric substrates such as HfO₂, Al₂O₃, SiO₂, and *h*-BN. Nonlocal scattering of electrons on polar optical phonons, charged impurities, and remote scattering on phonons in the dielectric materials is included in the self-consistent Born approximation. The band tails are found to critically depend on the layer thickness, temperature, doping concentration, and particularly on the chosen dielectric substrate. The underlying physical mechanisms are studied in high detail and an analytical interpolation formula is given to provide a quick reference for Urbach parameters and to guide the design work in MoS₂, WS₂, and WSe₂.

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I. INTRODUCTION

Two-dimensional materials have attracted considerable attention due to their unique electronic, optical, and mechanical properties [1–5]. Transition-metal dichalcogenides (TMDs) have a finite band gap making it an attractive alternative in electronics for Si/SiGe-based transistors [6–9], in optoelectronics as possible materials in light-emitting diodes [10–15] and solar-cell applications [16–18]. TMD layers are coupled by weak van der Waals forces, which allows for mechanical cleavage of bulk TMD materials into mono- and multilayer systems. These systems yield electronic and optical properties that depend strongly on the number of layers [19]. Stacking multiple TMD layers on top of each other significantly widens the available material design space [20,21] resulting in a plethora of ultrathin devices such as stacked optoelectronic *p-n* junctions [4,22,23], photovoltaics [24–26], as well as ultrascaled nonvolatile and neuromorphic memory devices [27–29]. Electrons in TMDs scatter on phonons, defects, and charged impurities, which leads to band tails (also known as Urbach tails), i.e., exponentially decaying density of states in the band gap [30–36]. The

slope of the exponential density of states' tail is known as the Urbach parameter. Urbach tails can significantly alter the device performance: the switching of transistors is drastically affected by these tails [37–39]. They affect the optical behavior such as absorption spectra and absorption-recombination coefficients in optoelectronic devices [40–42]. They also set a fundamental limit on the subthreshold performance of semiconductor devices at cryogenic temperatures for large-scale quantum-computing applications [43,44]. Since Urbach tails are formed due to electron-phonon and electron-defect interactions, the Urbach parameter is strongly dependent on temperature and doping concentration [30,34,45,46]. State-of-the-art models for the Urbach parameter of specific materials are either heuristic or the parameters are directly extracted from experimental observations [30,32–35,39,46,47]. In addition, TMD-based nanodevices typically interface the TMD layers with various oxides. Therefore, TMD device electrons scatter on remote phonons as well [48–50]. Experiments for several TMDs have shown that their Urbach tails strongly depend on the oxide type [51,52]. Different scattering mechanisms can interfere with each other and impact the electronic density of states rendering it hard to estimate without detailed calculations [31,53,54]. This is further complicated by the

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strong dependence of electronic and scattering properties in TMD systems on the number of atomic layers and their environment [49,55,56]. Therefore, this work predicts Urbach parameters for MoS₂, WS₂, and WSe₂ layer systems as a function of layer thickness, temperature, doping concentration, and oxide type (comprising HfO₂, Al₂O₃, SiO₂, and *h*-BN) with density-functional-theory (DFT) based quantum-transport calculations [57,58] for electronic Green's functions including scattering on various types of phonons, charged impurities, and remote scattering on oxide phonons with NEMO5 [31,59,60]. We delineate the contribution of different scattering mechanisms towards the Urbach parameter and gain insight into its dependence on layer thickness and oxide type. All calculations in this work are based on the nonequilibrium Green's function (NEGF) implementation of NEMO5. NEGF is well suited to analyze Urbach parameters [31,61] since it is a method of choice to predict electronic behavior when incoherent scattering and coherent quantum effects are equally relevant. Electronic, thermal, and optoelectronic systems with nanoscale dimensions or pronounced nonequilibrium conditions are a few such examples [62–69]. This work summarizes the quantum-transport calculations with an easily accessible lookup formula to predict Urbach parameters of MoS₂, WS₂, and WSe₂ layer systems as a function of layer thickness, temperature, doping concentration, and oxide type. The presented results and formula enable easy assessments of Urbach parameters and ways to customize them for specific TMD-based device concepts.

II. SIMULATION APPROACH

All TMD structures considered in this work are represented in their native atomic lattice. Electrons are subatomically resolved with maximally localized Wannier functions (MLWFs) derived from DFT Hamiltonians [19,57]. The process of generating electronic Hamiltonian operators first requires a self-consistent electronic structure calculation with the DFT tool VASP [70] with a convergence criterion of 10^{-8} eV. A momentum mesh of $5 \times 5 \times 5$ Monkhorst-Pack grid and an energy cutoff of 520 eV is used along with van der Waals force included following Ref. [71]. The applied DFT model is the generalized gradient approximation (GGA) employing the Perdew-Burke-Ernzerhof (PBE) functionals. The resulting DFT Hamiltonian operators are then transformed into a MLWF representation using the Wannier90 software [72–74] with *d* orbitals for the metal sites and *sp*³-hybridized orbitals for the chalcogenide sites as the initial projection. The spreading of the Wannier functions is reduced iteratively until it converges to around $2A^2$. The atom positions and their corresponding electronic Hamiltonian operator of TMD structures are then imported into NEMO5 for NEGF calculations. All electronic Green's functions are solved in the

self-consistent Born approximation with self-energies representing each incoherent scattering mechanism [scattering of electrons with acoustic phonons (APs), polar optical phonons (POPs), charged impurities (CIs), and remote phonons (RPs) from the oxide]. Since the structures are assumed to be periodic in the transverse *Y* direction (see Fig. 1), the electronic Green's functions and self-energies depend on the electronic energy *E* and in-plane momentum *k*. The corresponding Brillouin zones are resolved with 25 points. All Green's functions and self-energies are matrices with respect to the MLWF orbitals along the $\vec{x} = (X, Z)$ directions. (see Fig. 1). Each scattering process is modeled with a corresponding retarded and lesser scattering self-energy [53,62]. The imaginary part of the retarded self-energy provides information about the scattering rate of the electrons [75], which gives rise to the band tails. The real part of the retarded self-energies yields an energy shift of electronic states [31,76]. Since this work focuses on the Urbach parameter only, the real part of all retarded scattering self-energies is ignored.

In this work, Green's functions of electrons are explicitly solved, whereas Green's functions of phonons are approximated as plane waves occupied with the equilibrium Bose distribution N_{ph} . The three acoustic phonon types (LA, TA, ZA) are averaged into a single effective deformation potential and sound velocity. The corresponding self-energy given in Ref. [65] is multiplied by $3 \times$ accordingly. POPs are modeled with a constant, material-dependent phonon energy $\hbar\omega_{\text{LO}}$. Scattering on CIs is assumed to be elastic. Scatterings on POP and CI are based on long-range Coulomb interaction and therefore yield nonlocal scattering self-energies [77,78]. To limit the numerical burden of solving self-energies and Green's functions but still faithfully predict the POP and CI scattering, the respective self-energies are approximated to be local and multiplied with a material and device-dependent compensation factor. The detailed expressions for POP and

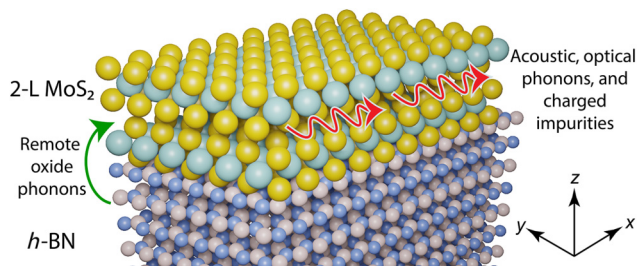


FIG. 1. Representative schematic of the simulated atomic TMD layers placed on top of oxide. Electrons are solved with scattering on acoustic and optical phonons and charged impurities within the TMD layers. In addition, electrons scattering remotely on oxide phonons are included. The *x* direction and *z* direction are resolved in real space, the *y* direction is assumed to be periodic.

CI scattering self-energies and the compensation factor are given in Ref. [31]. The interaction potential of electrons and remote oxide phonons is taken from Ref. [79]. The

resulting lesser and retarded scattering self-energies for electrons remotely scattering on the two surface optical-oxide-phonon modes ($\nu = 1, 2$) are given by

$$\begin{aligned}\Sigma^<(\vec{x}_1, \vec{x}_2, k, E) &= \sum_{\nu} \frac{e^2}{(2\pi)} \frac{\hbar\omega_{\text{SO},\nu}}{2\epsilon_o} \left(\frac{1}{\epsilon_{\text{ox}}^{\infty} + \epsilon_s^{\infty}} - \frac{1}{\epsilon_{\text{ox}}^s + \epsilon_s^{\infty}} \right) \int dk' I(k, k', \vec{x}_1, \vec{x}_2, z_1, z_2) \\ &\quad \times [N_{\text{ph}} G^<(\vec{x}_1, \vec{x}_2, k', E - \hbar\omega_{\text{SO},\nu}) + (1 + N_{\text{ph}}) G^<(\vec{x}_1, \vec{x}_2, k', E + \hbar\omega_{\text{SO},\nu})] \\ \Sigma^R(\vec{x}_1, \vec{x}_2, k, E) &= \sum_{\nu} \frac{e^2}{(2\pi)} \frac{\hbar\omega_{\text{SO},\nu}}{2\epsilon_o} \left(\frac{1}{\epsilon_{\text{ox}}^{\infty} + \epsilon_s^{\infty}} - \frac{1}{\epsilon_{\text{ox}}^s + \epsilon_s^{\infty}} \right) \int dk' I(k, k', \vec{x}_1, \vec{x}_2, z_1, z_2) \\ &\quad \times \left[(1 + N_{\text{ph}}) G^R(\vec{x}_1, \vec{x}_2, l, E - \hbar\omega_{\text{SO},\nu}) + N_{\text{ph}} G^R(\vec{x}_1, \vec{x}_2, k', E + \hbar\omega_{\text{SO},\nu}) \right. \\ &\quad \left. + \frac{1}{2} G^<(\vec{x}_1, \vec{x}_2, k', E - \hbar\omega_{\text{SO},\nu}) - \frac{1}{2} G^<(\vec{x}_1, \vec{x}_2, k', E + \hbar\omega_{\text{SO},\nu}) \right. \\ &\quad \left. + i \int \frac{d\tilde{E}}{2\pi} G^<(\vec{x}_1, \vec{x}_2, k', \tilde{E}) \left(\text{Pr} \frac{1}{E - \tilde{E} - \hbar\omega_{\text{SO},\nu}} - \text{Pr} \frac{1}{E - \tilde{E} + \hbar\omega_{\text{SO},\nu}} \right) \right],\end{aligned}\quad (1)$$

where

$$\begin{aligned}I(k, k', \vec{x}_1, \vec{x}_2, z_1, z_2) &= 2\pi J_0 \left(\sqrt{(k - k')^2 + \zeta^{-2} |\vec{x}_1 - \vec{x}_2|} \right) \\ &\quad \times e^{-\sqrt{(k - k')^2 + \zeta^{-2} (z_1 + z_2 - 2t)}}.\end{aligned}\quad (2)$$

$\hbar\omega_{\text{SO},\nu}$ is the ν^{th} optical-phonon frequency of the underlying oxide, ϵ_{ox}^s and $\epsilon_{\text{ox}}^{\infty}$ are the static and infinite frequency dielectric constants of the oxide, ϵ_s^{∞} is the infinite frequency dielectric constant of the TMD. The oxide-phonon modes are considered to decay exponentially into the TMD with z_1 and z_2 the distances of the two electron propagation coordinates from the oxide-semiconductor interface (see Fig. 1). t is the thickness of the TMD system. J_0 is the Bessel- J function of 0th order. The electrostatic screening of electrons and holes is represented by ζ and calculated with the Lindhard formula [80]

$$\zeta_{\text{Lindhard}} = \left(\frac{e^2}{\epsilon_o \epsilon_s} \frac{-2}{(2\pi)^3} \int d\vec{q} \frac{\partial f}{\partial \epsilon} \Big|_{\epsilon(\vec{q})} \right)^{-1/2}, \quad (3)$$

where f is the Fermi distribution function and the momentum integral runs over the first Brillouin zone. Scattering on remote phonons is nonlocal and long ranged as well. Similar to the nonlocal POP and CI self-energies, the remote phonon-scattering self-energy is approximated to be local and multiplied with a compensation factor following the approach of Ref. [31]. For later discussion of the results, we define an intrinsic phonon-scattering prefactor (IP) and a remote oxide-phonon-scattering prefactor

(ROP) as

$$P_{\text{IP}} = \frac{\hbar\omega_{\text{LO}}}{2\epsilon_o} \left(\frac{1}{\epsilon_s^{\infty}} - \frac{1}{\epsilon_s} \right), \quad (4)$$

$$P_{\text{ROP}} = \sum_{\nu} \frac{\hbar\omega_{\text{SO},\nu}}{2\epsilon_o} \left(\frac{1}{\epsilon_{\text{ox}}^{\infty}} - \frac{1}{\epsilon_{\text{ox}}^s} \right), \quad (5)$$

where ϵ_s is the static dielectric constant of the TMD material. For all the results shown in subsequent sections, the electrons are solved in equilibrium. The electronic Fermi level is determined such that the spatially integrated electron density agrees with the integrated doping concentration to achieve global charge neutrality. Green's functions are solved with the Dyson and Keldysh equations:

$$\begin{aligned}G^R &= (EI - H - \Sigma_{\text{AP}}^R - \Sigma_{\text{POP}}^R - \Sigma_{\text{CI}}^R - \Sigma_{\text{ROP}}^R \\ &\quad - \Sigma_{\text{source}}^R - \Sigma_{\text{drain}}^R)^{-1}, \\ G^< &= G^R (\Sigma_{\text{AP}}^< + \Sigma_{\text{POP}}^< + \Sigma_{\text{CI}}^< + \Sigma_{\text{ROP}}^< \\ &\quad + \Sigma_{\text{source}}^< + \Sigma_{\text{drain}}^<) G^{R\dagger}.\end{aligned}\quad (6)$$

All scattering self-energies are self-consistently solved with Green's functions until the relative particle current variation is less than 1×10^{-5} throughout the device. The source and drain contact self-energies are solved following Ref. [81]. The Urbach parameter is extracted from the exponentially decaying spatially averaged density of states below (above) the conduction (valence) band [31]. The extracted Urbach parameters correspond to the ones measured in transport experiments. In contrast, the parameter

TABLE I. Sound velocity v_s , material density ρ , LO phonon frequency ($\hbar\omega_{\text{LO}}$), deformation potentials (D), dielectric constants (ϵ_s and ϵ_∞), and IP scattering prefactor for one–four layers of MoS₂, WS₂, and WSe₂ used in this work. Parameters are taken from Refs. [55] and [82].

Material	Number of layers	v_s (m/s)	ρ (kg/m ³)	$\hbar\omega_{\text{LO}}$ (meV)	D (eV/nm)	ϵ_s	ϵ_∞	IP scattering prefactor
MoS ₂	1	7200	5060	48	4.5	3.8	3.2	0.4069
	2				5.37	5.65	4.8	0.2584
	3				6.25	6.47	5.5	0.2248
	4				7.12	7.3	6.2	0.2004
WS ₂	1	6670	7500	33	3.2	3.65	3.1	0.5847
	2				4.07	5.15	4.37	0.4169
	3				4.95	5.92	5.03	0.3595
	4				5.82	6.7	5.69	0.3187
WSe ₂	1	5550	9320	30	3.2	3.7	3.145	0.6167
	2				4.07	5.3	4.5	0.4337
	3				4.95	6.1	5.18	0.3765
	4				5.82	6.9	5.86	0.3326

extracted from optical measurements are related to excitons, which require a different scattering model accounting for the excitonic interaction. All other material parameters for the TMDs and oxides are taken from Refs. [55,56,82] and listed in Tables I and III.

III. RESULTS

A. Band tails: Intrinsic to the material

Electronic scattering on APs and POPs creates finite density of states above (below) the valence (the conduction) band that decays exponentially into the band gap (see Fig. 2). The slope of this band tail, i.e., the Urbach parameter increases with incoherent scattering of electrons on charged impurities (as also observed in Ref. [31]). The scattering rates are proportional to the imaginary retarded scattering self-energies (Σ^R) [75]. In the self-consistent Born approximation Σ^R is proportional to the retarded Green's function (G^R) [see Eq. (1) and Refs. [31,59]]. Therefore, the strength of the scattering processes that form Urbach tails is determined by the imaginary part of G^R , i.e., the density of states at the band edges [31]. This is the root cause for the effective-mass dependency of scattering rates in Fermi Golden rule models (see Refs. [77,78]). This results in the Urbach parameter of monolayer MoS₂ valence band being larger than its conduction band as shown in Fig. 2. The effective masses of different valleys and their relative contribution to the Urbach parameter are described in the subsequent paragraph. The phonon- (charged impurity) scattering self-energies are proportional to the phonon number (doping concentration) [31].

Accordingly, Figs. 3(a) and 3(b) show that the Urbach parameters of MoS₂ valence-band electrons increase with doping concentration and temperature. Figures 3(a) and 3(b) also show a reduction of the Urbach parameter with the number of MoS₂ layers. This is due to the

fact the density of states at the top of the valence band of monolayer MoS₂ is larger than the ones of any multilayer MoS₂ system. While the valence-band maximum of monolayer MoS₂ lies at the K valley with an effective mass of $0.543m_e$, the K -valley energy of monolayer MoS₂ valence bands is only 59 meV higher than the energy of the Γ valley and marginally higher than the polar optical-phonon energy of 48 meV. The phonon occupancy of 48-meV modes at 300 K is high enough for holes at the top of the MoS₂ valence band to experience significant intervalley scattering and to yield a density of states effectively composed of contribution from both valleys. The effective mass of the Γ valley ($2.886m_e$) is significantly larger than the one of the K valley. Therefore, intervalley scattering raises

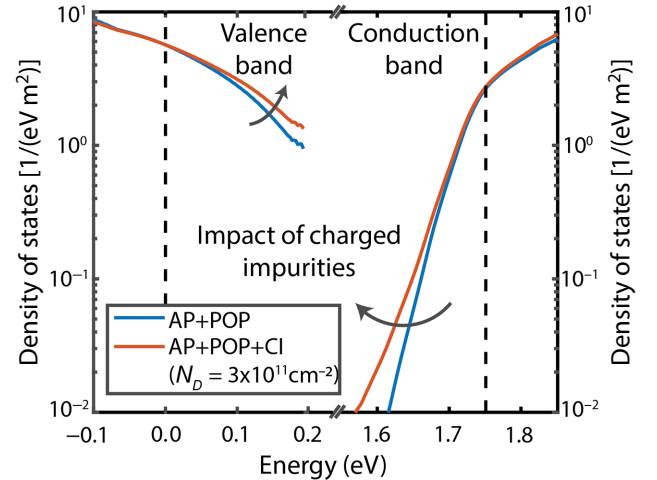


FIG. 2. Density of states of monolayer MoS₂ close to valence- and conduction-band-edges. Scattering on APs and POPs creates Urbach tails into the band gap (blue). Scattering on CIs (red) increases the Urbach tail further. The black dashed lines indicate the valence- and conduction-band-edges without incoherent scattering.

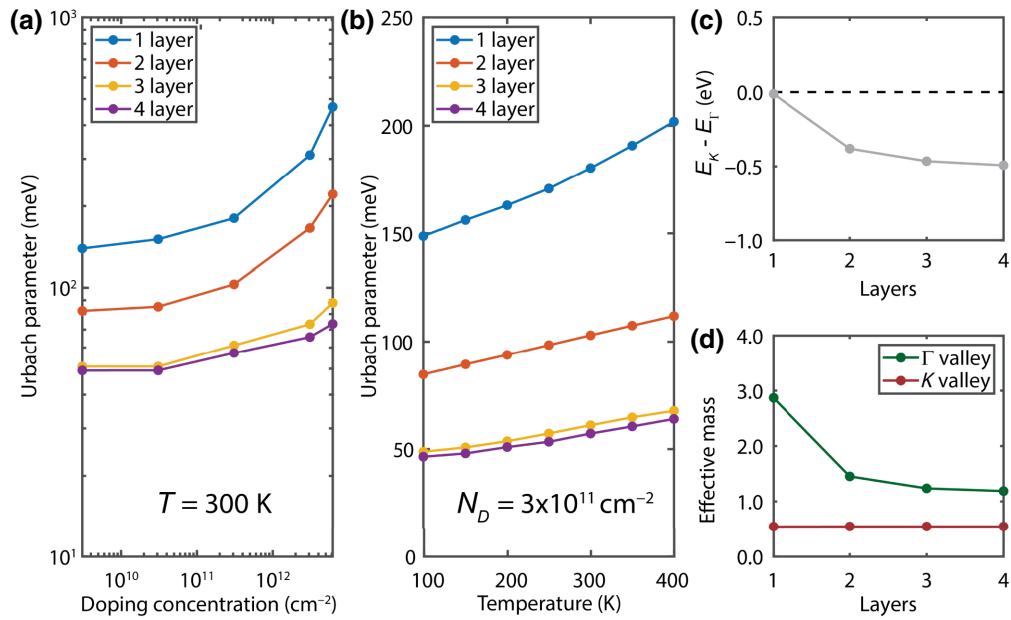


FIG. 3. (a) Urbach parameter of the MoS₂ valence band as a function of doping concentration for a different number of layers at 300 K. With increasing doping concentration, the Urbach parameter increases due to stronger impurity scattering. (b) Urbach parameter of the MoS₂ valence band as a function of temperature for a different number of layers at a doping concentration of $3 \times 10^{11} \text{ cm}^{-2}$. With increasing temperature, the Urbach parameter increases due to stronger phonon scattering. Both (a) and (b) show smaller Urbach parameters with larger number of layers because of the reduction of valence-band-edge density of states with more layers: only monolayer MoS₂ exhibits degenerate valence-band *K* and Γ valleys as depicted with their energy differences in (c). The density of states of the Γ valley is proportional to its effective mass, which reduces with the number of MoS₂ layers, shown in (d).

the effective density of states of monolayer MoS₂ holes and the corresponding Urbach parameter. This effective degeneracy of *K* and Γ valleys is lifted as soon as more than one layer of MoS₂ is present [see Fig. 3(c)]. The effective mass of the Γ valley, i.e., the highest valence-band valley of multilayer MoS₂ systems, and with it the density of

states at the top of the valence band declines continuously with the number of layers with the steepest decline happening between monolayer and bilayer [see Fig. 3(d)]. That is why adding a second MoS₂ layer gives the largest reduction in the Urbach parameter. In addition, the polar optical-phonon-scattering potential decreases with larger

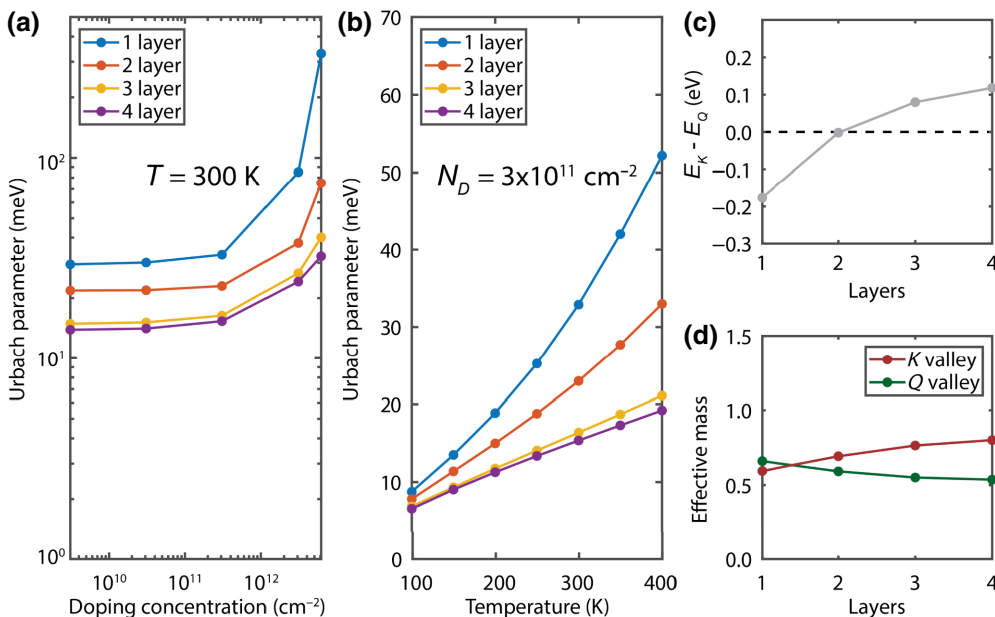


FIG. 4. Similar to Fig. 3, the Urbach parameter of the MoS₂ conduction band as a function of doping concentration (a) and temperature (b). Differing from the valence-band results of Fig. 3, the *K* and *Q* valleys are degenerate in the bilayer case, [see (c)] and the conduction-band-edge effective mass has a relatively small layer thickness dependence as shown in (d). In spite of the bilayer valley degeneracy, the Urbach parameter still decreases monotonously with the number of layers, since the reduction in scattering potential of the bilayer overcompensates its increase in band-edge density of states (see Table I).

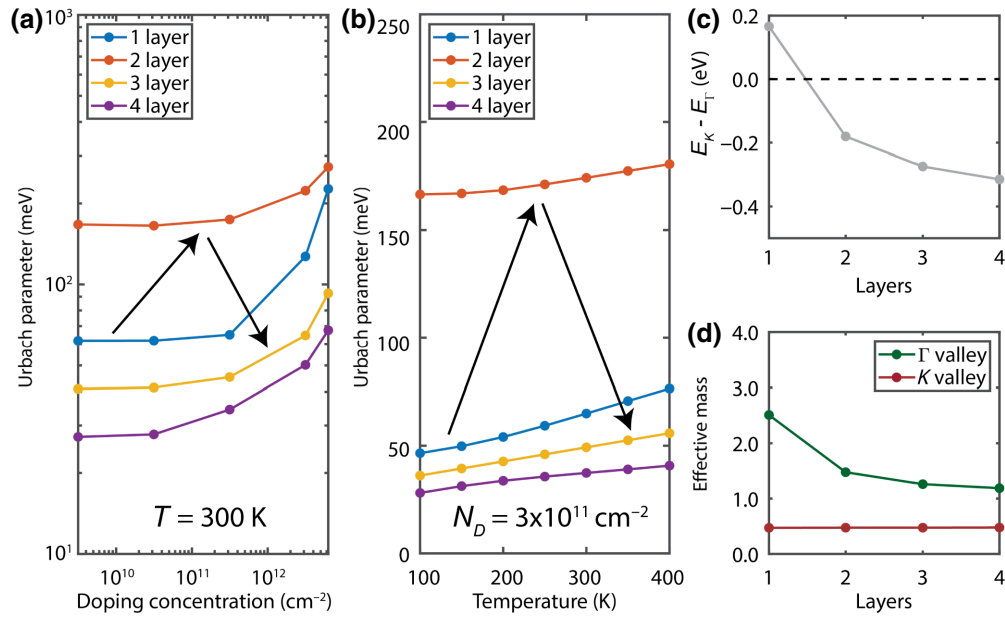


FIG. 5. (a) Urbach parameter of the WS₂ valence band as a function of doping concentration for a different number of layers at 300 K (a) and as a function of temperature for a doping density of $3 \times 10^{11} \text{ cm}^{-2}$. Similar to the valence-band Urbach parameter of MoS₂ (Fig. 3), the Urbach parameter of WS₂ increases with doping concentration and temperature. The energy of *K* valleys exceed those of the Γ valleys only for monolayer WS₂ as shown in (c). The transition of the valence-band-edge from *K* to Γ valley and the large difference of *K*- and Γ -valley effective masses [shown in (d)] cause a maximum of the WS₂ band-edge density of states and accordingly a maximum of the Urbach parameter for the bilayer configuration (indicated with arrows).

dielectric constants [31,65,83], which has been observed to increase in thicker MoS₂ layers [55,82]. It is worthwhile to mention that we also observe the decreasing impact of scattering in thicker ultrathin bodies of III-V materials in Ref. [31].

The conduction band of MoS₂ has a similar dependence on doping concentration and temperature as the

valence band [see Figs. 4(a) and 4(b)]. However, the conduction-band valleys *K* and *Q* are degenerate only in the bilayer case [see Fig. 4(c)]. In spite of the expected increase in band-edge density of states and with it an increase of the conduction-band Urbach parameter of the bilayer MoS₂, the Urbach parameter shows a monotonous decrease with the number of MoS₂ layers [see Figs. 4(a)

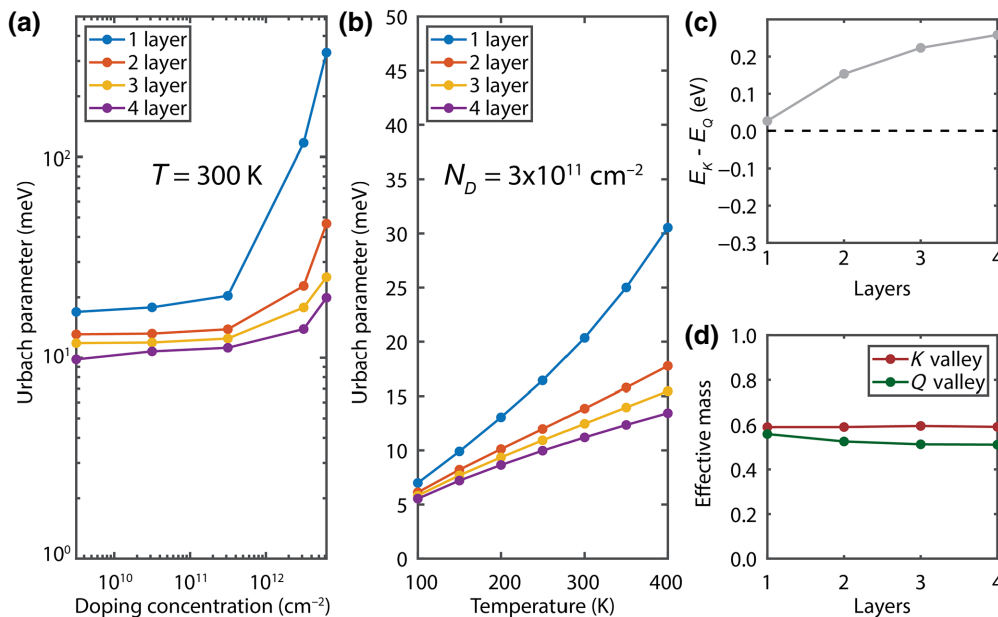


FIG. 6. (a) Urbach parameter of the WS₂ conduction band as a function of doping concentration (a) and temperature (b) similar for the conduction band of MoS₂ in Fig. 4. The Urbach parameter declines with increasing number of layers mainly due to the reduction of the scattering potential (see Table I), since neither valley degeneracy in (c) nor the valley effective mass in (d) changes significantly with the number of layers.

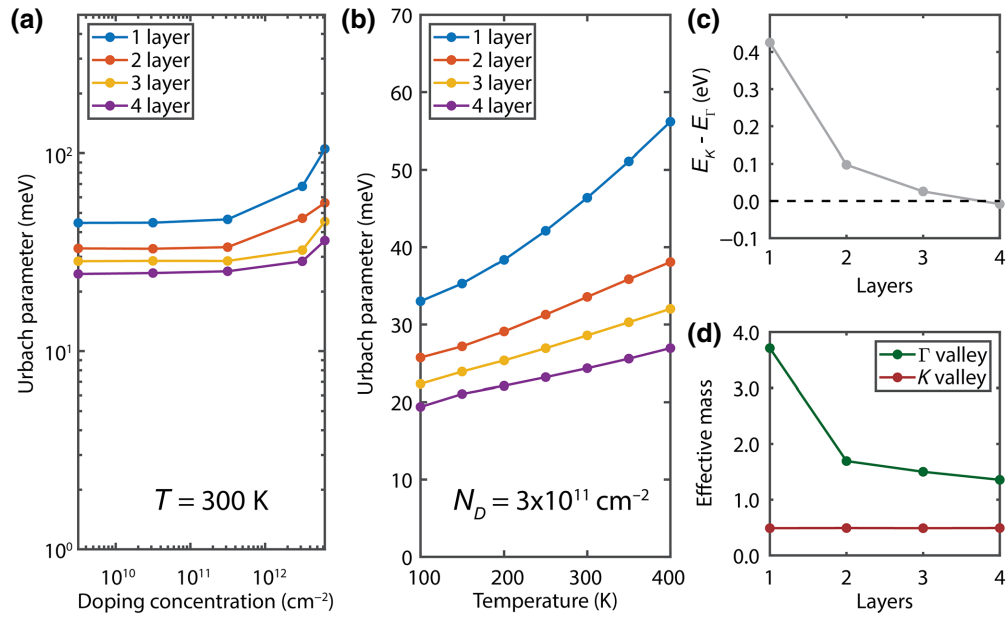


FIG. 7. (a) Urbach parameter of the WSe₂ valence band as a function of doping concentration for a different number of layers at 300 K. With increasing doping concentration, the Urbach parameter increases due to stronger impurity scattering. (b) Urbach parameter of the WSe₂ valence band as a function of temperature for a different number of layers at a doping concentration of $3 \times 10^{11} \text{ cm}^{-2}$. With increasing temperature, the Urbach parameter increases due to stronger phonon scattering. Both (a) and (b) show smaller Urbach parameters with a larger number of layers because of the reduction of valence-band-edge density of states with more layers: only four-layer WSe₂ exhibits degenerate valence-band K and Γ valleys as depicted with their energy differences in (c). Otherwise, the Γ valley contributes only to the conduction. The density of states of the Γ valley is proportional to its effective mass, which reduces with the number of WSe₂ layers, shown in (d).

and 4(b)]. This is due to a significant reduction of the calculated scattering self-energy prefactor of polar optical-phonon-scattering from monolayer to bilayer MoS₂ (see Table I). Overall, the conduction-band Urbach parameters of MoS₂ layers are lower than those of the valence

band due the lower conduction-band effective masses [see Fig. 4(d)].

The valence-band Urbach parameters of WS₂ layers in Figs. 5(a) and 5(b) show similar changes with doping concentration and temperature as the MoS₂ valence-band

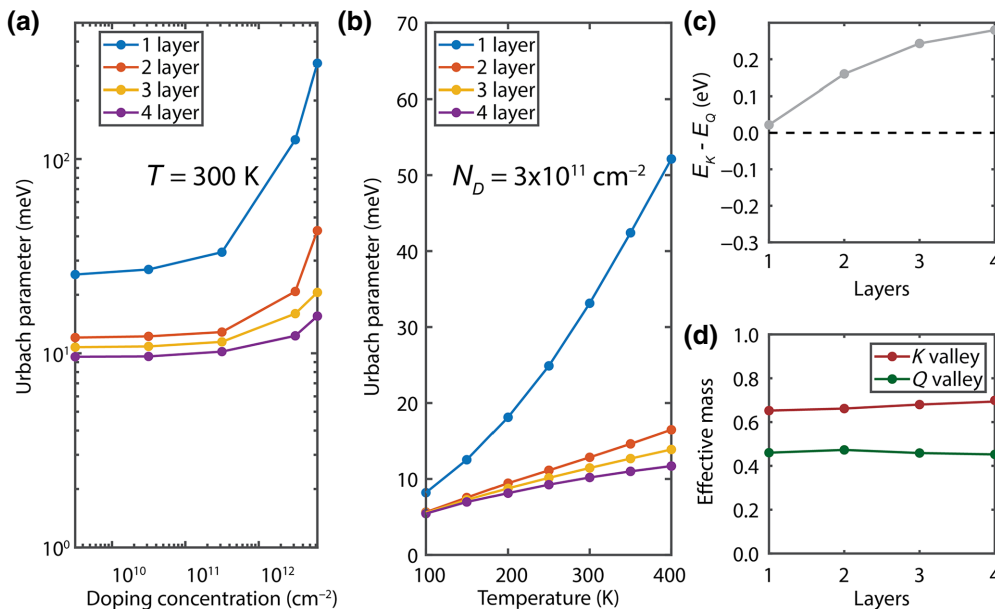


FIG. 8. (a) Urbach parameter of the WSe₂ conduction band as a function of doping concentration (a) and temperature (b) similar to the conduction band of WSe₂ in Fig. 8. The Urbach parameter declines with increasing number of layers mainly due to the reduction of the scattering potential (see Table I), since neither valley degeneracy in (c) nor the valley effective mass in (d) changes significantly with the number of layers.

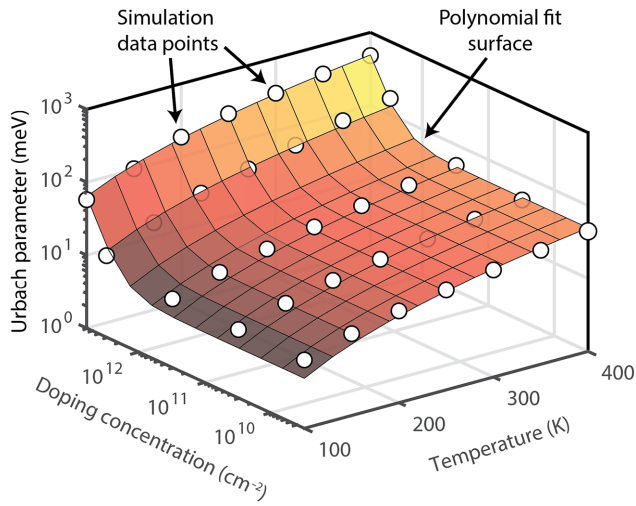


FIG. 9. Urbach parameter as a function of doping concentration and temperature for monolayer MoS₂. The white circles represent the simulation data points and the contour represents its least-squares polynomial fit. The fit parameters for this and all other systems are given in Table II.

results of Fig. 3. However, the Urbach parameter is largest for bilayer WS₂. This is due to a transition of the valence-band-edge from *K* to Γ valley when more than one layer of

WS₂ is present [see Fig. 5(c)]. Given the large difference of *K*- and Γ -valley effective masses [see Fig. 5(d)], this transition causes a maximum in the valence-band-edge density of states and therefore a maximum in the scattering strength for bilayer WS₂. Accordingly, the Urbach parameter follows this trend [indicated with arrows in Figs. 5(a) and 5(b)]. WS₂ with more than two layers show the similar reduction in the Urbach parameter as discussed already for MoS₂ in Fig. 3.

The conduction-band Urbach parameter of WS₂ (see Fig. 6) is very similar to the one of MoS₂ (see Fig. 4) in its overall dependence on doping and temperature. The scattering potential of the conduction band of WS₂ and with it the Urbach parameter decays with the number of layers (see Table I). Although the conduction band *K* and *Q* valleys of monolayer WS₂ are close enough in energy to allow for intervalley scattering, the masses of *K* and *Q* valleys are very similar. Thus, neither significant effective-mass changes nor valley degeneracies influence the Urbach-parameter behavior for WS₂ conduction bands. The same is true for valence and conduction bands of WSe₂ layers as shown in Figs. 7 and 8.

For completeness and to ease the estimation of the Urbach parameter as a function of doping concentration, temperature, layer thickness, and material, all calculated Urbach parameters are input to a polynomial fit given by

TABLE II. Parameters for predicting the Urbach tail with Eq. (7) as a function of temperature and doping concentration for MoS₂, WS₂, and WSe₂ along with the *R*² deviation from the calculated quantum transport result. The parameters are determined with the MATLAB curve fitting toolbox [84].

Material	Band type	Number of layers	<i>a</i>	<i>b</i>	<i>p</i>	<i>q</i>	<i>r</i>	<i>R</i> ² fit value
MoS ₂	Valence	1	149.57	61.162	0.1461	0.8073	0.9078	0.9955
		2	82.981	39.460	0.1116	0.73851	0.6793	0.9978
		3	50.096	14.623	0.1014	1.3003	0.4714	0.9817
		4	49.101	0.1101	0.1002	3.7729	2.3337	0.9271
	Conduction	1	30.567	5.7612	1.6752	1.4527	2.1574	0.9965
		2	22.461	2.4506	1.0747	1.6788	1.6752	0.9988
		3	15.368	2.9972	0.8734	0.0885	1.1907	0.9923
		4	48.497	1.8010	0.0590	6.5438	0.3661	0.9009
WS ₂	Valence	1	63.210	11.144	0.4238	1.3126	1.5879	0.9951
		2	172.43	18.337	0.1102	0.4899	0.8087	0.9222
		3	40.343	12.360	0.2296	0.9083	0.4475	0.9848
		4	27.545	13.102	0.1408	0.8728	0.5770	0.9782
	Conduction	1	17.814	15.216	1.4801	1.0604	1.6482	0.9982
		2	13.449	1.2230	0.7822	0.8175	1.8035	0.9970
		3	12.046	1.3597	0.7226	0.0675	1.2610	0.9930
		4	10.196	1.0997	0.4620	0	1.2088	0.9721
WSe ₂	Valence	1	45.762	4.3223	0.3396	1.0847	1.4928	0.9797
		2	33.271	0.8838	0.2733	0	1.1190	0.9129
		3	28.843	0.0825	0.2060	3.0934	2.8990	0.9810
		4	24.699	0.5682	0.2214	1.7593	1.6473	0.9564
	Conduction	1	28.203	18.465	1.4141	1.0200	1.4832	0.9990
		2	12.450	1.0889	0.7411	0.8994	1.8314	0.9936
		3	10.684	2.1195	0.6217	0	0.8655	0.9917
		4	9.2162	1.6687	0.3946	0	0.7384	0.9311

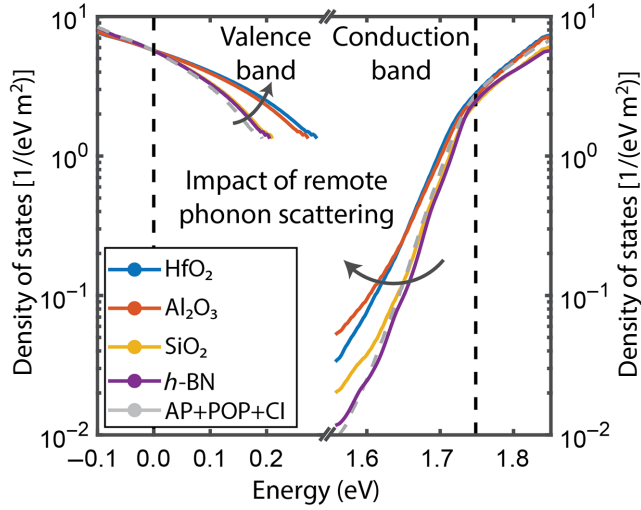


FIG. 10. Density of states of monolayer MoS₂ when exfoliated on various dielectric materials, close to valence- and conduction-band-edges (solid lines). The remote scattering on the dielectric material phonons increases the Urbach tail compared to the intrinsic value of monolayer MoS₂ (gray dashed lines). The impact of the dielectric material phonon scattering follows the same order that is given in Table III, i.e., the strength of the remote phonon-scattering prefactor. The black dashed lines indicate the valence- and conduction-band-edges without incoherent scattering.

$$\text{Urbach}(T, N_D) = a(T/300)^p + b(T/300)^q (N_D/1e12)^r. \quad (7)$$

Figure 9 exemplifies this fit for the conduction-band Urbach parameter of MoS₂. Since the Urbach parameter depends nonmonotonically on the number of layers, the least-squares fitting is performed for each layer separately. The fit parameters of all considered TMD systems together with their R^2 fit values can be found in Table II.

B. Band tails: Impacted by dielectric materials

In typical TMD-based nanodevices, the TMD layers are capped with dielectric materials. The remote scattering on phonons in those dielectric materials contribute significantly to the Urbach band tail, as shown in Fig. 10.

The relative impact of remote scattering on phonons in the dielectric materials is directly proportional to the

scattering self-energy prefactor listed in Table III. This prefactor is determined by the difference of the dielectric constants and the energies of the soft optical-phonon modes [see Eq. (1)]. It is worthwhile to mention, HfO₂ has a large scattering impact given its high difference in dielectric constants and the comparably low soft optical-phonon-mode energies. Following the same arguments, the low scattering contributions of *h*-BN and SiO₂ originate in their high phonon-mode energies and small difference in their dielectric constants.

Figure 11 shows how the ROP scattering impacts the Urbach parameter more in thinner TMD systems than in the thicker ones. This is due to the smaller spatial confinement in larger TMD systems, which allows the charge density in the TMD layers to move farther away from the insulator-TMD interface. Accordingly, the difference in the contributions of the various insulators decreases with increasing TMD thickness. Beyond three layers, the Urbach parameter starts to saturate and slowly approaches the intrinsic value of the respective TMD. As seen already in Fig. 10, the Urbach parameters are consistently higher for Al₂O₃ and HfO₂ due to their strong ROP scattering potentials.

It is worth mentioning that a similar trend for mobility is observed in Refs. [50,56] for capped TMD systems with HfO₂/MoS₂ and Al₂O₃/MoS₂ exhibiting the strongest suppression of mobility (strong scattering) and SiO₂/MoS₂ exhibiting the weakest suppression of mobility (weak scattering) in comparison to the vacuum case. In addition, Ref. [50] demonstrates increasing mobility with increasing MoS₂ thickness and saturation to the bulk value around five layers similar to our observed variation of Urbach parameter with the number of layers for each type of oxide.

To roughly benchmark our simulation results against experimental data, we extract the Urbach parameters from several published PL measurements. Note that a direct comparison with the calculated values of this work cannot be quantitative, since all the predicted Urbach parameters of this work are transport band tails, which are known to deviate from the optical, excitonic band tails [85]. We compute the Urbach parameter from the PL spectrum of experimental results by extracting the slope below the band edge. Reference [51] for the conduction band of monolayer MoS₂ exfoliated on SiO₂ presents an Urbach parameter

TABLE III. Soft optical-phonon frequencies ($\hbar\omega_{\text{SO},1}$ and $\hbar\omega_{\text{SO},2}$), static and infinite frequency dielectric constants of the oxides used in this work taken from Ref. [56] along with the respective ROP scattering prefactors.

Oxide	$\hbar\omega_{\text{SO},1}$ (meV)	$\hbar\omega_{\text{SO},2}$ (meV)	ϵ_s	ϵ_∞	ROP scattering prefactor
HfO ₂	12.40	48.35	23.00	5.03	3.6363
Al ₂ O ₃	48.18	71.41	12.53	3.20	2.9231
SiO ₂	55.60	138.10	3.90	2.50	1.0478
<i>h</i> -BN	93.07	179.10	5.09	4.10	0.1159

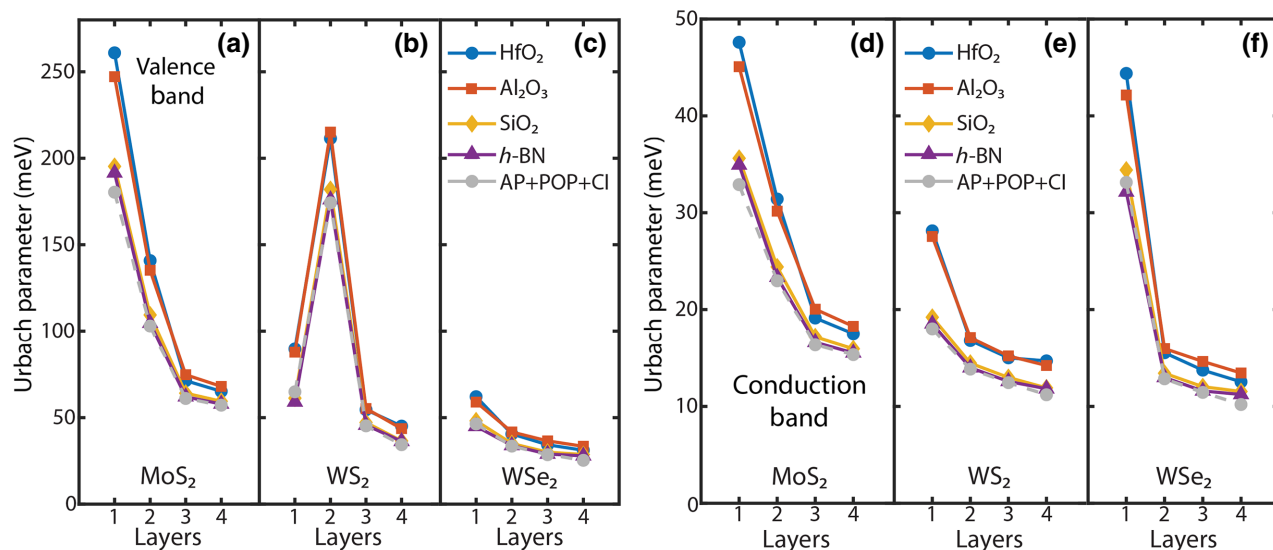


FIG. 11. Conduction-band (a)–(c) and valence-band (d)–(f) Urbach parameters of MoS_2 , WS_2 , and WSe_2 systems capped with HfO_2 (dots), Al_2O_3 (squares), SiO_2 (diamonds), or $h\text{-BN}$ (triangles) along with their intrinsic values (gray dashed lines). As the TMD thickness increases, the maximum of the electron density is located farther from the semiconductor-insulator interface resulting in bands and their Urbach parameters being less affected by the remote scattering on oxide phonons. HfO_2 and Al_2O_3 increase the Urbach parameter more than the other substrates due to their large scattering potentials (see also Fig. 10). Bilayer WS_2 valence bands show an elevated Urbach parameter due to a transition of the valence-band maximum from the K to the Γ valley at that layer thickness.

of 40.06 meV before and 30.62 meV after the chemical removal of native defects. Monolayer MoS_2 fabricated as a type-II heterointerface with perylene in Ref. [86] shows 46.04 meV and 52.94 meV conduction-band Urbach parameter for two perylene hosts—one providing a clean interface and the other hosting trap states, respectively. Reference [52] lists a conduction-band tail parameter of 30.04 meV for monolayer $\text{MoS}_2\text{-WSe}_2$ van der Waals heterostructure. All the above values correlate well with the findings in Fig. 11.

IV. CONCLUSION

Quantum-transport calculations of electrons in TMD systems are used to predict the formation of band tails due to scattering on phonons, charged impurities, and remote scattering on substrate dielectric phonons. All materials are atomically resolved, electronic Hamiltonian operators are based on DFT, and incoherent, nonlocal scattering is modeled in self-consistent Born approximation. It is shown that the Urbach band-tail parameter strongly depends on temperature, impurity concentration and TMD layer thickness as well as on the type of dielectric substrate. The details of the Urbach parameter depend on a balance of valley degeneracies, scattering potentials, and phonon occupancies. This can result in nonmonotonic behavior of the Urbach parameter as seen in WS_2 . To ease reproducibility of the sophisticated quantum-transport calculations, we present analytical approximations to the observed Urbach-parameter predictions. These analytical formula allow fast

assessments on how far the band tails are tunable within a given TMD-based device concept. Among the dielectric materials considered, Al_2O_3 and HfO_2 are shown to contribute strongest to the remote phonon-scattering-enhanced band tails. With increasing TMD-layer count, electrons spread farther away from the dielectric and thus the contribution of remote scattering on dielectric phonons decreases. Monolayer MoS_2 results agree well with published observations in experiments.

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