

Universal Thickness-Dependent Absorption in Solids at the Nanoscale

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Through systematic experimental and theoretical studies of layer-thickness-dependent absorption in semiconducting MoSe₂ and WS₂ across the visible to near-infrared spectral range, we demonstrate a universal absorption behavior in solids at nanoscale thicknesses. With increasing thickness, a non-monotonic evolution of absorption integrated over the measured spectral region is revealed which is accompanied by pronounced oscillatory features. This strongly deviates from the expected Beer-Lambert's law. The observed behavior has origins in the electromagnetic interference effects taking place between the two surfaces of the thin crystals. The present work on 2D semiconductors is extendable to all kinds of solids such as conventional semiconductors (e.g. Si, GaAs, GaN, InP), (semi) metals (e.g. Al, Ag, Au, c-HOPG) and 2D magnetic material (e.g. CrSBr and NiPS₃). Our results provide fundamental insights into light-matter interactions in solids at the nanoscale and are vital for optimally designing the new generation of absorption-based flexible optoelectronic devices.

Low-dimensional solids such as quantum wells and van der Waals (vdW) materials are excellent platforms to study two-dimensional quantum effects and for their potential towards the next generation of optoelectronic, spintronic and valleytronic devices [1–5]. Group III-V and II-VI semiconductor quantum wells and heterostructures have already revolutionized modern optoelectronics [6]. Recently, vdW materials such as transition metal dichalcogenides (TMDCs) of chemical formula MX_2 ($M = \text{Mo, W, Re etc.}, X = \text{S, Se, Te etc.}$), graphene, hexagonal boron nitride (hBN), and the newly emerging 2D magnetic materials (2DMM e.g. CrX_3 with $X = \text{Cl, Br, I}$; MPX_3 with $M = \text{Mn, Fe, Zn, Ni}$ and $X = \text{S, Se}$; Fe_nGeTe_2 with $n = 3, 4$ and 5) have opened immense potential towards flexible and energy-efficient device technologies [3,7]. Semiconductors are active layers in the absorption-based optoelectronic devices such as photodiodes, photovoltaics, phototransistors, avalanche photodiodes, charge coupled devices (CCDs) and complementary metal-oxide-semiconductor (CMOS) devices [6]. Interestingly, monolayer TMDCs, despite their ultrathin thickness (less than 1 nm), can absorb a considerable fraction of the visible light (approximately 5-10 %) [8]. Additionally, spintronics and valleytronics require heterostructures of thin semiconductor films (such as TMDCs) with magnetic films (such as 2DMMs) for efficient control of spin-valley degrees of freedom in TMDCs [4]. Indeed, a complete device also requires electrical contacts through thin metallic/semi-metallic films such as silver, gold or graphene [7,9]. Overall, it is crucial to understand thickness-dependent light absorption by thin solid films for an optimal device performance. This is especially important for photovoltaic devices where optimized film thicknesses are essential for large efficiencies [9–11]. Unfortunately, a careful study of absorption of thin solid films with layer thickness is not available to the best of our knowledge [12]. As such, before layered materials era, it was challenging for researchers to prepare samples suited to measure absorption with varying layer thicknesses.

Intuition suggests that the absorption of light in a material should monotonically increase with increasing layer thickness, and should follow the well-established Beer-Lambert law [12–15]. However, for layer thicknesses in the sub-wavelength regime, which are required for the ultrathin device applications, strong interference effects can lead to drastic deviations from intuitive expectations [16,17]. This can significantly affect device performance if the layer thicknesses are chosen inappropriately.

In the present work, we systematically address this issue both theoretically and experimentally. We measure absorption spectra of thin crystals of MoSe₂ and WS₂ as representative materials, with layer thicknesses up to 200 nm. We notice striking deviations from Beer-Lambert law. These discrepancies are explained using a model based on the generalized transfer-matrix (GTM) method, which addresses interference effects taking place between multiple interfaces of the samples. We extend our results theoretically to various types of solids such as conventional semiconductors (e.g. GaAs, Si, InP, GaN), 2DMMs (e.g. CrSBr, NiPS₃) and (semi)metals (e.g. Au, Al, Ag, graphite) and find universal trends.

The samples used for our experiments are prepared using the standard exfoliation and viscoelastic dry-transfer method [18]. We exfoliate thin layers of MoSe₂ and WS₂ single crystals (obtained from 2D semiconductors, USA) on 0.5 mm thick c-cut sapphire substrate due to its transparency in the spectral measurement region. We measure spatially-resolved (1 μm resolution) reflectance $R(E)$ and transmittance $T(E)$ spectra of the crystals at normal incidence using a home-built confocal spectroscopy setup (see Fig. S1 of the supplementary materials). All measurements are performed at the room temperature ($T = 296 \text{ K}$). Absorption spectra $A(E)$ are deduced from the $R(E)$ and $T(E)$ spectra as $A(E) = 1 - R(E) - T(E)$ [13,16]. Thicknesses of the crystals are initially estimated by comparing their observed colors under a microscope with colors calculated using the transfer matrix method (TMM) (see Fig. S9 and Fig.

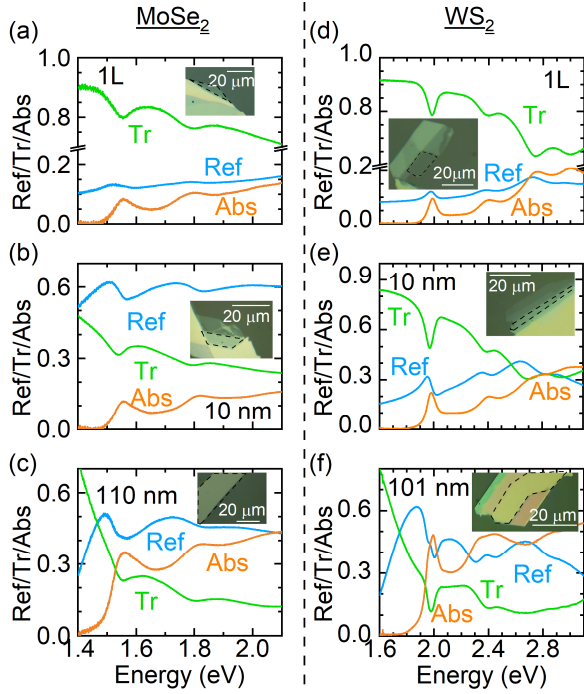


Figure 1. Reflectance (Ref, blue), transmittance (Tr, green), and absorption (Abs, orange) spectra of monolayer (1L), 10 nm, and 110 nm MoSe₂ (left), and 1L, 10 nm and 101 nm thick WS₂ (right). The crystals are exfoliated on 0.5 mm thick sapphire substrate. Insets show optical microscope images of the crystals, where regions enclosed by dashed lines are the flakes for which data are presented.

S10 of the supplementary materials). Crystal thicknesses are accurately measured using atomic force microscopy (AFM). Due to the optically flat nature of the crystals, as confirmed by AFM images, we are able to neglect scattering losses while deducing the absorption spectra. A few examples of AFM images are shown in Fig. S2 of the supplementary material.

In Fig. 1, we show the reflectance, transmittance and absorption spectra of monolayer (1L or 0.65 nm), 10 nm, and 110 nm thick MoSe₂, and 1L, 10 nm and 101 nm thick WS₂ as examples. In all cases, we notice well-known features corresponding to various excitonic resonances in the materials [19,20]. Additionally, after every resonance in the absorption spectrum, there is a step-like feature (Sommerfeld enhancement of the step-like absorption due to 2D joint density of states) [21], which is a classic signature of two dimensionality of these crystals, irrespective of their layer thicknesses. This thickness-independent two-dimensionality has been widely discussed in the literature on TMDCs, which has, for instance, led to the discovery of interlayer excitons in bilayers and multilayers [22]. In the present work, we do not focus on the exciton resonances as such, however, our target is to understand the integrated absorption (area under the absorption curve in the measured spectral region) as a function of layer thickness. As the flake thickness increases from 10 nm to about 100 nm, a notable enhancement in absorption is observed, accompanied by a reduction in transmittance. This is expected since thicker crystals interact much more strongly with the incident light.

Before going into any quantitative description, here we already notice an unusual behavior of reflectance of MoSe₂ where 110 nm thick crystal seems to reflect less strongly (Fig. 1(c)) compared to 10 nm case (Fig. 1(b)). This counterintuitive observation warrants theoretical and experimental investigations of reflectance, transmittance and absorption of TMDCs over a broad region of thicknesses. Let us turn our attention to the absorption spectra first, which are relatively simpler to understand.

Figures 2 (a) and (b) depict measured absorption spectra of MoSe₂ and WS₂ for thicknesses up to 200 nm, respectively. The spectral range for MoSe₂ is between $E_1 = 1.45$ eV to $E_2 = 2.2$ eV (bandwidth ΔE of 0.75 eV), while for WS₂, it is between $E_1 = 1.8$ eV to $E_2 = 3.1$ eV (bandwidth ΔE of 1.3 eV). The absorption approaches 0 below E_1 . We realize that the experimentally measured absorption does not show a monotonic increase with thickness [Figs. 2(a) and 2(b)]. To verify this, we integrate the absorption spectra over experimental spectral

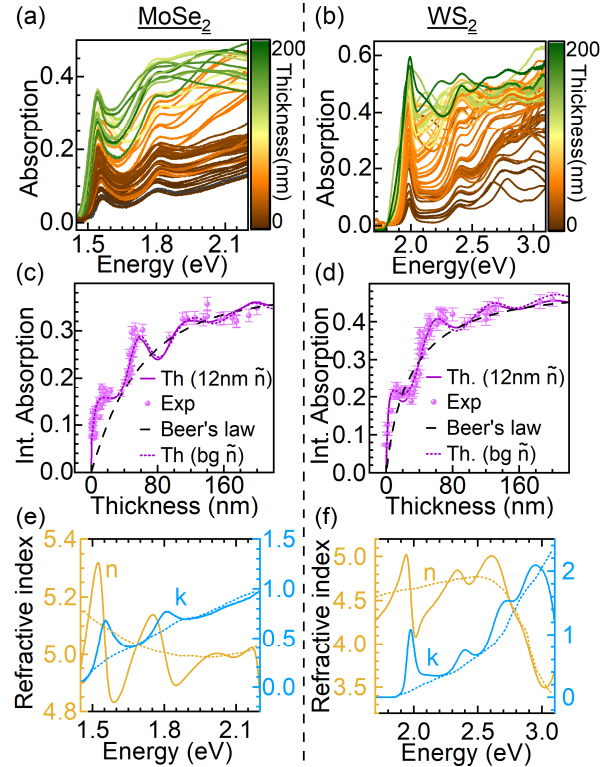


Figure 2. (a) and (b) are the experimental absorption spectra for MoSe₂ and WS₂ with varying thicknesses, ranging from monolayer (1L) to 200 nm for MoSe₂ and WS₂. Spheres in (c) and (d) are the experimental integrated absorption for the two materials with varying thickness. Dashed black lines are obtained using Beer's law. Solid lines are the theoretical curves obtained using complex refractive indices ($\tilde{n}$) of 12 nm thick crystals, while dotted lines are the curves obtained when exciton resonances are removed from $\tilde{n}$. Solid lines in (e) and (f) depict real (n) and imaginary (k) parts of $\tilde{n}$ extracted for 12 nm thick MoSe₂ and WS₂, using the corresponding reflectance and transmittance spectra. Background refractive indices (after removing excitonic contributions) are marked by dotted lines.

regions as a function of layer thickness for both materials. We normalize the integrated absorption A_{int} with the spectral bandwidth i.e. the quantity $A_{\text{int}}/\Delta E$ is plotted as solid spheres in Fig. 2(c) and 2(d) for MoSe₂ and WS₂ respectively, as a function of layer thickness. Notably, the monolayers and bilayers of both materials absorb $\sim 7.5\%$ and $\sim 10\%$ of light, respectively. Strikingly, integrated absorption shows sudden jumps for a certain range of thicknesses (e.g. 0 – 10 nm and 30 – 55 nm in both cases), while plateaus or even dips for other ranges (e.g. a plateau between 10 – 30 nm and a dip between 55 – 85 nm). The integrated absorption tends to saturate after 110 nm accompanied with subtle oscillatory behavior. Overall, this non-monotonic oscillatory behavior strongly deviates from the monotonically rising absorption predicted by the well-known Beer-Lambert law as detailed in the supporting information, and shown as dashed lines in Fig. 2(c) and 2(d). The only experiments available in the literature find a monotonic rise of integrated absorption in V₂O₅ layers with thickness, contrary to our findings [23]. This may be due to the authors of this work measuring absorption at only a few thickness points.

To understand the origin of the oscillatory features in the integrated absorption, we perform calculations based on the GTM method [16,17]. A complete description of the GTM formalism is provided in the supplementary material. Using the GTM approach [24,25], we first calculate the photon-energy-dependent complex refractive indices $\tilde{n} = n + ik$ of MoSe₂ and WS₂ as follows. We use experimentally measured reflectance $R(\lambda)$ and transmittance $T(\lambda)$ spectra of the 12 nm thick crystals of the two materials as inputs, while leaving the photon-energy-dependent dielectric functions $\tilde{\epsilon} = \epsilon_1 + i\epsilon_2 = (n + ik)^2$ as the free parameters. This thickness of 12 nm is chosen for our calculations because it can represent both regimes of ultrathin (1L limit) as well as the bulk reasonably well [26]. The curves obtained for n and k are shown as solid lines in Figs. 2(e) and 2(f) for the two materials, respectively. Using these refractive indices as inputs in the GTM formalism, we compute absorption spectra $A(\lambda) = 1 - R(\lambda) - T(\lambda)$ as a function of thickness for MoSe₂ and WS₂ for the same spectral range as that of the experiment. Thereafter, we integrate area under these curves, normalize it by dividing with the spectral bandwidth ΔE , and plot as solid lines in Figs. 2(c) and 2(d). The theoretical curves agree with the experimental data extremely well within the experimental error. To understand the effect of excitons on our calculations, we manually remove the excitonic features from n and k and obtain the background refractive indices as dotted lines shown in Figs. 2(e) and 2(f). The recalculated integrated absorption curves are depicted as dotted lines in Figs. 2(c) and 2(d) for MoSe₂ and WS₂, respectively. Strikingly, the overall qualitative behavior of the integrated absorption matches exceptionally well with the experimental data. This leads us to conclude that the oscillatory behavior of absorption must be universal to all solids, independent of the excitonic effects.

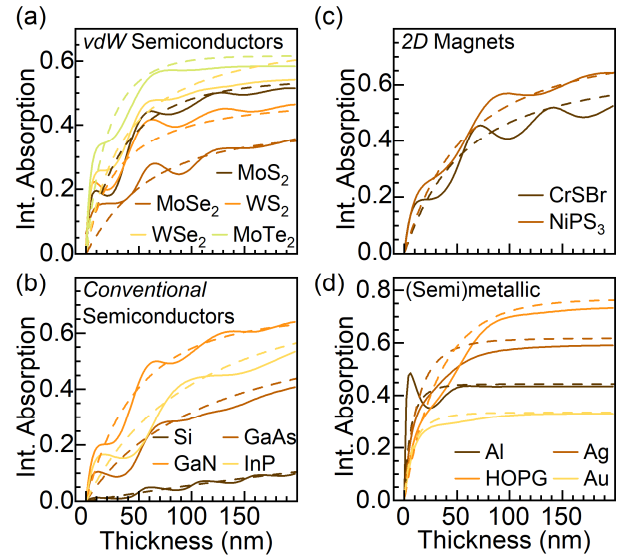


Figure 3. Solid and dashed lines show integrated absorption spectra using GTM method and Beer's law as discussed in supplementary material. (a) vdW semiconductors: MoS₂, MoSe₂, WS₂, WSe₂ and MoTe₂ on 0.5mm thick sapphire; (b) freestanding conventional semiconductors i.e. Si, GaAs, GaN and InP; (c) layered 2DMM: CrSBr and NiPS₃, on 0.5mm thick sapphire, (d) freestanding (semi)metallic materials i.e. Al, Ag, Au and c-HOPG (Graphite on 0.5mm thick sapphire). The curves for WS₂ and MoSe₂ are from the present work, other curves are generated using refractive indices obtained from the literature mentioned in Table 1.

To further establish the universality of the effect, we calculate the integrated absorption of many layered materials using the GTM method, a few 'conventional' semiconductors such as Si, GaAs, GaN and InP, 2DMM such as CrSBr and NiPS₃, and (semi)metals such as Al, Ag, Graphite (c-HOPG) and Au as a function of layer thickness in Fig. 3. The oscillatory behavior of the integrated absorption is obviously clear for all of the materials. Table 1 describes the details about the parameters used in our simulations such as energy range and the references for the refractive indices. The reader is referred to Fig. S8 of the supplementary materials for calculated curves of freestanding layers of layered materials, which show qualitatively the same behavior as Fig. 3.

To gain a deeper understanding of the oscillatory integrated absorption, we take a closer look at the reflectance $R(\lambda)$, transmittance $T(\lambda)$ and absorption spectra $A(\lambda) = 1 - R(\lambda) - T(\lambda)$ as a function of layer thickness for WS₂ as an example (see Fig. S7 of supporting information for the freestanding MoSe₂ and WS₂ layers). Figure 4 (a-c) presents the theoretical (solid lines, calculated using the GTM method) and experimental (circles) R , T and A for WS₂ at some specific wavelengths i.e. 400 nm to 700 nm in steps of 50 nm, as a function of layer thickness. In R and T curves, at a given wavelength, the expected oscillations due to interference taking place between the multiple interfaces of the sample i.e. air-crystal, crystal-substrate and substrate-air interfaces are visible. The extrema appear for certain layer thicknesses where the conditions for interference

Table 1. Details of input parameters i.e. initial and final energies (E_{int} and E_{finl} , respectively in eV) for computation, energy bandwidth (ΔE in eV) and reference used for refractive indices of materials using which integrated absorption has been calculated in Fig. 3.

S. N.	Material	E_{int}	E_{finl}	ΔE	Reference
vdW Semiconductors/2D Magnets (Fig. 3a and 3c)					
1	2H-MoS ₂	1.8	2.7	0.9	[28]
2	2H-MoSe ₂	1.45	2.2	0.75	Present work
3	2H-WS ₂	1.8	3.1	1.3	Present work
4	2H-WSe ₂	1.6	3	1.4	[28]
5	2H-MoTe ₂	1.8	3.1	1.3	[29]
6	CrSBr	1.85	2.4	0.55	[30]
7	NiPS ₃	1.8	2.7	0.9	[31]
Conventional semiconductors (Fig. 3b)					
8	Si	1.4	2.7	1.3	[32]
9	GaAs	1.4	2.7	1.3	[32]
10	GaN	3	5	2	[32]
11	InP	1.4	2.7	1.3	[33]
(Semi)metals (Fig. 3d)					
12	Au	1.8	3.1	1.3	[34]
13	Al	1.8	3	1.2	[35]
14	Ag	3	6	3	[34]
15	c-HOPG	2	3.1	1.1	[36]

are satisfied. For instance, for a freestanding layer (without substrate) of refractive index n , the maxima of reflectance (transmittance) would appear for $d = (2m - 1) \lambda / 4n$ ($d = m\lambda / 2n$) for an integer $m = 1, 2, 3 \dots$ [16]. As expected from intuition, the extrema observed in R and T shift towards higher thicknesses for longer wavelengths. Our experimental results are in excellent agreement with the simulations as seen in Fig. 4. Some deviations in the experimental data from the simulations could be due to the following. Simulations in the entire thickness and wavelength range are performed using a refractive index of a 12 nm thick crystal, whereas experimental data have variations in the excitonic effects as thickness changes. Fig. 4(c) reveals that strikingly, the oscillatory behavior persists even in absorption vs layer thickness curves for all wavelengths. This is counterintuitive since it is usually assumed that the oscillatory effects in reflectance and transmittance cancel each other (energy conservation), being equal and opposite. However, the amplitude of oscillations in absorption is not as pronounced as in reflectance or transmittance. Nonetheless, when integrated over all wavelengths in the spectral range, the oscillations persist in the integrated absorption in Fig. 4(d) which is seen experimentally. Interestingly, another form of ‘quantum’ universality in absorption of 2D layers has been reported earlier in 2D InAs membranes [27]. The authors showed that absorption spectra of InAs layers with thicknesses ranging from 3 – 19 nm display step-like features, with each step measured to be 1.6% of absorption. On the contrary, the present work solely deals within the classical electrodynamics regime and establishes the behavior of integrated absorption in solids with layer thickness at the nanoscales.

In summary, we establish a universal absorption behavior in solids at nanoscale thicknesses. The absorption, integrated

Ref/Trans/Abs of WS₂: Experiment vs Simulation

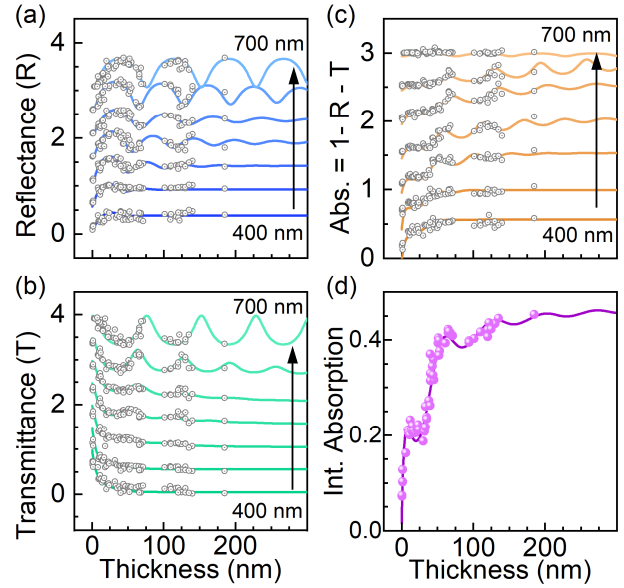


Figure 4. (a) to (c) represent calculated (solid lines) and experimental (circles) reflectance, transmittance and absorption of WS₂ on c-sapphire as a function of layer thickness, at selected wavelengths from 400 nm – 700 nm in steps of 50 nm. Plots are shifted in the y-axis for clarity, by 0.5 units successively. Fig 4(d) depicts the integrated absorption as a function of thickness for WS₂ where solid spheres are experimental data and solid line is theoretical curve calculated using GTM. The oscillatory behaviour from (c) persists in (d) as well.

across the entire spectral range, exhibits oscillatory behavior with varying layer thickness, significantly deviating from the expected Beer-Lambert law. These findings are crucial for advancing the understanding of light-matter interactions in solids and for developing optimized absorption-based next-generation optoelectronic devices, such as photovoltaics.

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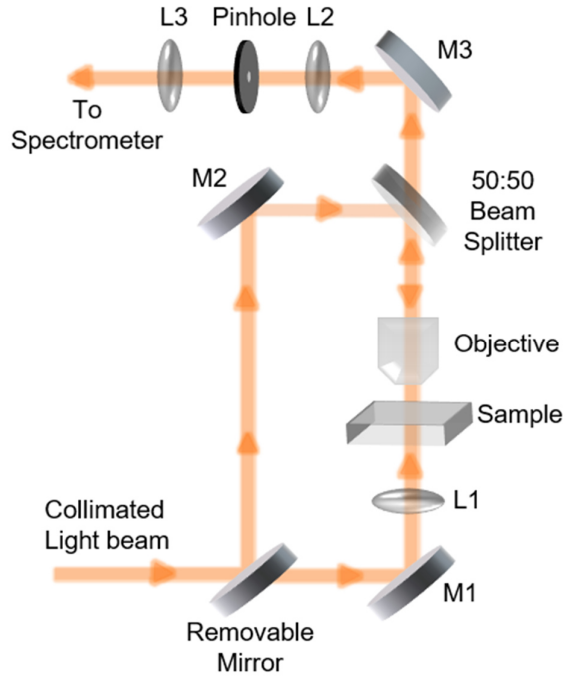


Figure S1. Experimental Setup.

1) *Reflection mode*: Broadband collimated light beam from a Xe arc lamp or tungsten Halogen lamp is reflected from the removable mirror, undergoes reflection from the M2 and 50:50 beam splitter and is focused on the sample using 50x objective lens. The reflected light from the sample passes through the 50:50 beam splitter, reflects from M3, and is focused on pinhole of 20 μm diameter using L2, while L3 collimates it again. The pinhole selects signal from $\sim 1 \mu\text{m}$ region of the sample.

2) *Transmission mode*: Collimated light from the light source is reflected from M1 and is focused on the sample by L1. Afterwards, it is the same as reflection mode. Light after L3 is wavelength dispersed after passing through a 328 mm focal length spectrometer, and is detected using a Peltier-cooled CCD.

Absorption from Beer-Lambert law

Free Standing Layer: Air-crystal-Air system

Consider a solid slab of thickness d , with an absorption coefficient α , bounded by air on both sides. This configuration constitutes an air-solid-air system as illustrated in Figure S3.

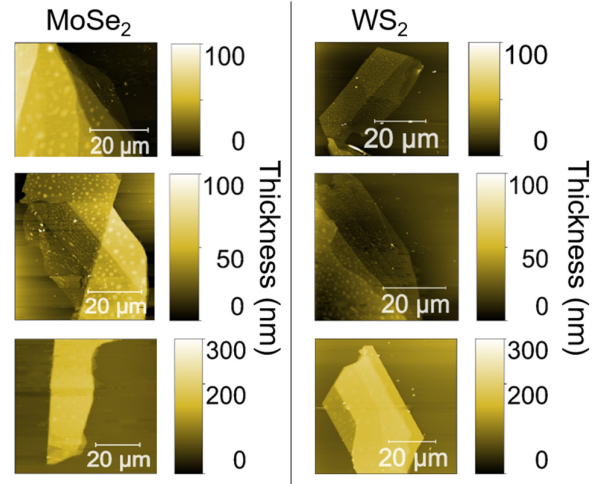


Figure S2. Tapping-mode atomic force microscopy (AFM) height maps of representative mechanically exfoliated MoSe₂ (left) and WS₂ (right) crystals on 0.5mm thick c-sapphire substrate. These images correspond to the same regions shown in the optical micrographs presented in Fig.1. Similar measurements were performed extensively across multiple flakes to ensure reproducibility and consistency of the observed thickness-dependent behavior of absorption reported in this work.

At the *air-solid* interface, consider an infinitesimally thin region of thickness dx . Since absorption within this region is negligible, energy conservation suggests

$$R_o + T_o = 1 \quad (1)$$

where, R_o and T_o are the reflectance and transmittance at the interfaces $x = 0$ and $x = d$, respectively.

Generally, within a solid, the transmitted intensity $I(x)$ at location x can be expressed in terms of the intensity entering the solid I_o at $x = 0$, using Beer-Lambert law as [1]

$$I(x) = I_o e^{-\alpha x} \quad (2)$$

A reflection/transmission (R/T) at every successive interface (including the first interface) leads to a new I_o in Eq. 2, which takes the losses due to R and T at the interface into account. Therefore, the total reflectance of the system, R , can be obtained by considering all internally reflected rays that eventually exit backwards through the $x = 0$ *air-solid* interface as follows [1–3]

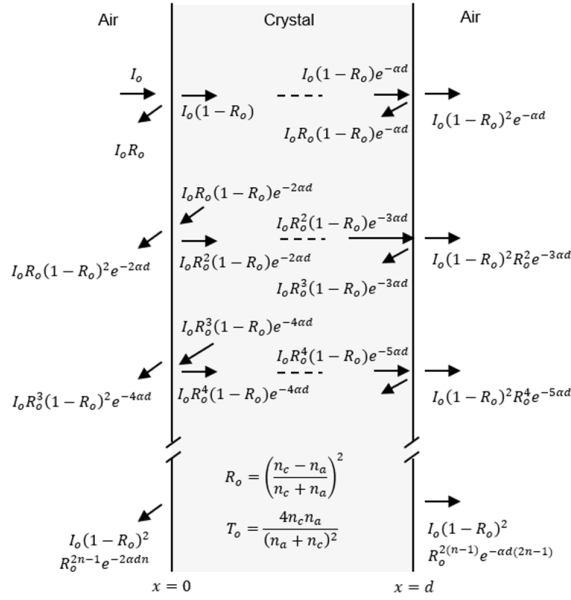


Figure S3. The air-crystal-air (freestanding) system where energy losses of the incident beam due to reflection losses at the interfaces, and attenuation due to Beer-Lambert law in the medium, has been explicitly accounted for. Here, I_o is the intensity of the incident beam, α is the absorption coefficient of the crystal, and R_o is the reflectance at the *air-crystal* interface, calculated using n_a and n_c , which are the refractive indices of air and the crystal respectively.

$$\begin{aligned}
 R &= R_o + (1 - R_o)^2 R_o e^{-2\alpha d} \\
 &\quad + (1 - R_o)^2 R_o^3 e^{-4\alpha d} + \dots \\
 &= R_o + (1 - R_o)^2 e^{-2\alpha d} R_o \sum_{n=0}^{\infty} R_o^{2n} e^{-2\alpha d n} \\
 &= R_o + \frac{(1 - R_o)^2 R_o}{1 - R_o^2 e^{-2\alpha d}} e^{-2\alpha d} \quad (3)
 \end{aligned}$$

Similarly, by considering all rays that eventually exit through $x = d$ *solid-air* interface, the overall transmittance of the system, T , can be written by summing over these contributions as follows [1–3]

$$\begin{aligned}
 T &= (1 - R_o)^2 e^{-\alpha d} + (1 - R_o)^2 R_o^2 e^{-3\alpha d} \\
 &\quad + (1 - R_o)^2 R_o^4 e^{-5\alpha d} \dots \\
 &= (1 - R_o)^2 e^{-\alpha d} \sum_{n=0}^{\infty} e^{-2\alpha d n} R_o^{2n} \\
 &= \frac{(1 - R_o)^2}{1 - R_o^2 e^{-2\alpha d}} e^{-\alpha d} \quad (4)
 \end{aligned}$$

The effective absorption of the system, A , can therefore be computed using Eq. (3) and Eq. (4) as follows [1–3]

$$\begin{aligned}
 A &= 1 - R - T \\
 &= 1 - \left(R_o + \frac{(1 - R_o)^2 R_o}{1 - R_o^2 e^{-2\alpha d}} e^{-2\alpha d} \right) \\
 &\quad - \left(\frac{(1 - R_o)^2}{1 - R_o^2 e^{-2\alpha d}} e^{-\alpha d} \right)
 \end{aligned}$$

$$= \frac{(1 - R_o)(1 - e^{-\alpha d})}{(1 - R_o e^{-\alpha d})} \quad (5)$$

Air-Crystal-Substrate-Air system: Extending the approach described in the preceding section, the effective reflectance and transmittance (and thereby absorption) of an *air-crystal-substrate-air* system can be obtained by accounting for multiple internal reflections at each interface, and attenuation with each medium, as shown in Fig. S4.

Reflectance coefficients at the *air-crystal*, *crystal-substrate*, and *substrate-air* interfaces in Fig. S4 are denoted as R_o^{ac} , R_o^{cs} and, R_o^{sa} , respectively. Absorption coefficients of the crystal and substrate are represented by α_c , and α_s , and their respective thicknesses by d_c and d_s .

We define a parameter, γ ,

$$\gamma = R_{sa} + \frac{(1 - R_{sa})^2 R_{cs} e^{-2\alpha_s d_s}}{1 - R_{sa} R_{cs} e^{-2\alpha_s d_s}} \quad (6)$$

in terms of which, the total reflectance R_{total} , and total transmittance T_{total} , of the system can be written as follows:

$$R_{total} = R_{ac} + \frac{(1 - R_{ac})^2 \gamma e^{-2\alpha_c d_c}}{1 - R_{ac} \gamma e^{-2\alpha_c d_c}} \quad (7)$$

$$T_{total} = \frac{(1 - R_{ac})(1 - R_{cs})(1 - R_{sa})e^{-(\alpha_c d_c + \alpha_s d_s)}}{(1 - R_{ac} \gamma e^{-2\alpha_c d_c})(1 - R_{sa} R_{cs} e^{-2\alpha_s d_s})} \quad (8)$$

Therefore, the total absorption of the system can be computed using equations (7) and (8) as follows:

$$A_{total} = 1 - R_{total} - T_{total} \quad (9)$$

This expression was used to calculate the absorption from Beer-Lambert law for our system, using complex refractive indices (with excitonic features) and plotted in Fig.2 (c) and (d) in the main manuscript.

Generalized Transfer matrix model:

In many cases, the substrate thicknesses are much larger than the coherence length of the light source (e.g. tungsten halogen or xenon arc lamps) used for spectroscopy. To account for the effect of substrates properly, the *conventional* transfer-matrix method such as the one discussed in Ref. [4] fails. It is because, there is no provision for introducing incoherence in this method. A complete transfer-matrix calculation including a substrate results in strong oscillations in absorption such as in Fig. S6 (a and b) for MoSe₂ and WS₂ kept on 0.5 mm thick c-sapphire substrate. Therefore, in the present work, we have performed our calculations using an improved version of the transfer-matrix method, called generalized transfer-matrix method [5,6]. In this method, the substrates can be set as incoherent and the results are much closer to reality

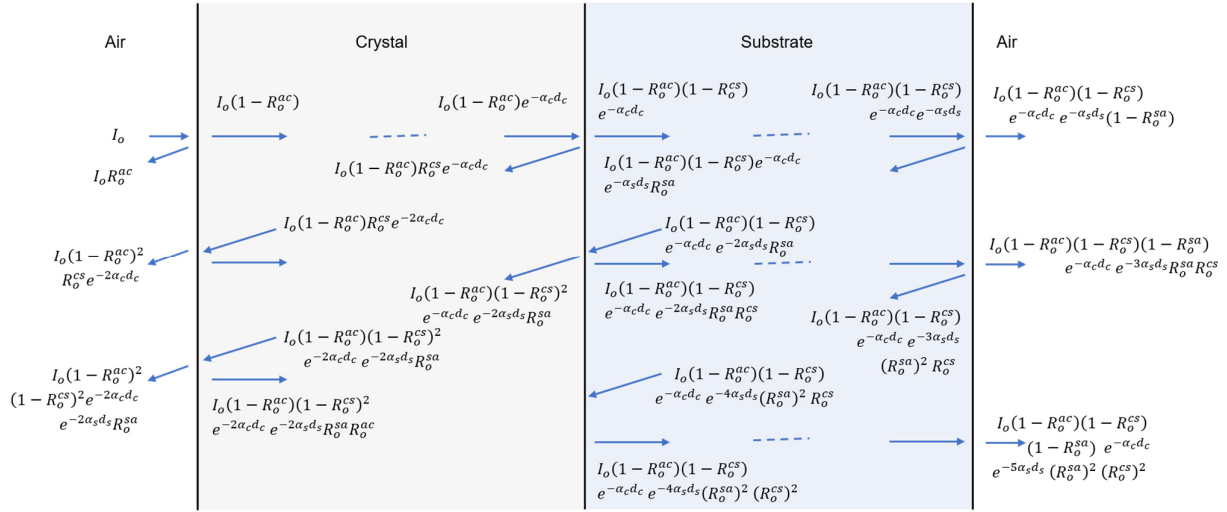


Figure S4: The air-crystal-substrate-air system where energy losses of the incident beam due to reflection losses at each interface, and attenuation due to Beer-Lambert law in the respective media, has been explicitly accounted for. Here, I_o is the intensity of the incident beam. Reflection coefficients at the air-crystal, crystal-substrate, and substrate-air interfaces are denoted as R_o^{ac} , R_o^{cs} and, R_o^{sa} , respectively. Absorption coefficients of the crystal and substrate are represented by α_c , and α_s , and their respective thicknesses by d_c and d_s . Note that only the leading order terms have been shown in the illustration.

(Fig. S6 (c and d)). Here we summarize the method as follows.

Let us consider a multilayer stack of i films which makes $i+1$ interfaces as shown in Fig. S5. The semi-infinite media on the leftmost (0^{th}) and on the rightmost ($i+1^{th}$) sides are considered as air or vacuum. Linearly polarized light passing through this structure can be described with the transfer-matrix formalism:

$$\begin{bmatrix} E_0^+ \\ E_0^- \end{bmatrix} = M \begin{bmatrix} E_{i+1}^+ \\ E_{i+1}^- \end{bmatrix} \quad (10)$$

where E_0^+ , E_{i+1}^+ and E_0^- , E_{i+1}^- are the forward and backward propagating electric field amplitudes at 0^{th} and $i+1^{th}$ interfaces. M is a 2×2 transfer matrix $M = \begin{pmatrix} M_{11} & M_{12} \\ M_{21} & M_{22} \end{pmatrix}$ given by

$$M = I_{01} \cdot L_1 \cdot I_{12} \dots L_i \cdot I_{i(i+1)} \quad (11)$$

where I_{ij} are 2×2 ‘interface’ matrices which connect the fields at the i^{th} and j^{th} boundaries, while L_i are the propagation matrices which describe the change in phase while wave propagates through the i^{th} layer. Both of these matrices are described in detail as follows.

$$I_{ij} = \frac{1}{t_{ij}} \begin{bmatrix} 1 & r_{ij} \\ r_{ij} & 1 \end{bmatrix} \quad (12)$$

where t_{ij} and r_{ij} are the complex Fresnel’s transmission and reflection coefficients at the interface ij for normal incidence (consider s polarization):

$$r_{ij} = \frac{n_i - n_j}{n_i + n_j} \text{ and } t_{ij} = \frac{2n_i}{n_i + n_j} \quad (13)$$

with n_i and n_j as the complex refractive indices of i^{th} and j^{th} films.

The matrix L (propagation matrix) is defined as

$$L_i = \begin{bmatrix} e^{-i\beta_i} & 0 \\ 0 & e^{i\beta_i} \end{bmatrix} \quad (14)$$

where β_i is the phase shift for the wave passing through the film i and is defined as

$$\beta_i = \frac{2\pi n_i d_i}{\lambda} \quad (15)$$

where λ is the wavelength of the incident light, n_i and d_i are the complex refractive index and the thickness of i^{th} film, respectively.

Now we proceed to calculate the final reflectance and transmittance coefficients through the multilayered system. Let us find the reflectance and transmittance amplitudes first. From (10),

$$\begin{bmatrix} E_0^+ \\ E_0^- \end{bmatrix} = \begin{bmatrix} M_{11} & M_{12} \\ M_{21} & M_{22} \end{bmatrix} \begin{bmatrix} E_{i+1}^+ \\ 0 \end{bmatrix} = \begin{bmatrix} M_{11} E_{i+1}^+ \\ M_{21} E_{i+1}^+ \end{bmatrix} \quad (16)$$

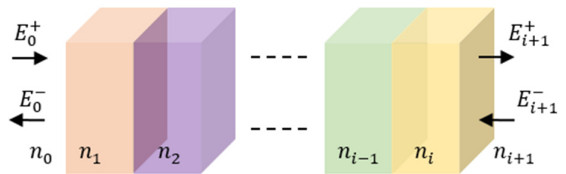


Figure S5. Schematic of the multilayer optical model. A dielectric stack is represented with refractive index n_i and thickness d_i . This model is used to describe how light propagates through thin films and to calculate reflectance, transmittance, and absorption.

with $E_{i+1}^- = 0$ (since there is no light incident back from the right side), reflection and transmission amplitudes are

$$r = \frac{E_0^-}{E_0^+} = \frac{M_{21}}{M_{11}} \text{ and } t = \frac{E_{i+1}^+}{E_0^+} = \frac{1}{M_{11}} \quad (17)$$

The reflectance and transmittance are, therefore, given as:

$$R = r^* r = \left| \frac{M_{21}}{M_{11}} \right|^2 \quad (18)$$

$$T = \left(\frac{n_{i+1}}{n_0} \right) t^* t = \left(\frac{n_{i+1}}{n_0} \right) \left| \frac{1}{M_{11}} \right|^2 \quad (19)$$

where r^* and t^* are the complex conjugates of the reflection and transmission amplitudes.

So far, light passing through every layer in the multilayered structure behaves coherently i.e. light accumulates phase as it passes through the different layers. However, it is problematic since the substrates, which are usually very thick compared to the coherence length of the light source, cannot be treated in this fashion. This is the reason we notice strong oscillations in Fig. S6(a) and (b) riding over the calculated spectra, which are absent in the experiment. For such thick layers, the relative phase between forward and backward-propagating waves inside that layer becomes effectively random. To introduce this *decoherence* effect

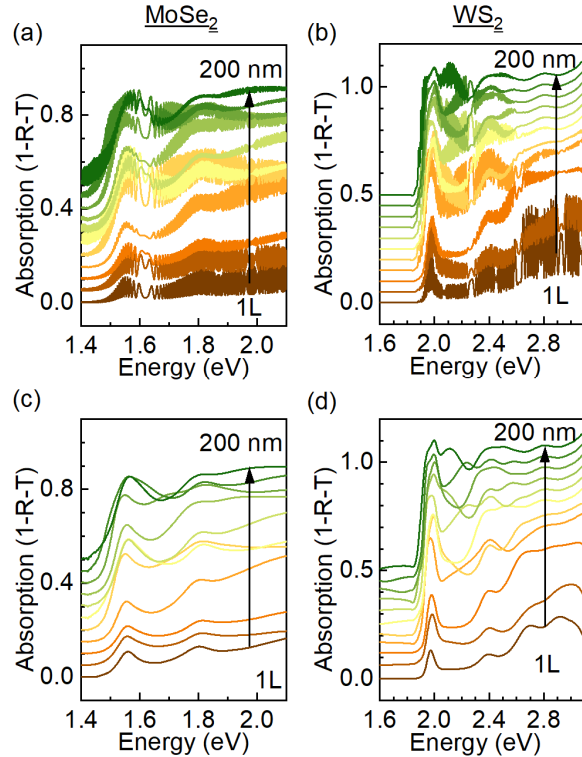


Figure S6. Calculated absorption spectra using *conventional* transfer-matrix method in (a) and (b) for MoSe₂ and WS₂, respectively, and using *generalized* transfer-matrix method in (c) and (d) where the TMDC crystals are kept on 0.5 mm thick c-sapphire substrate. The spectra are calculated for TMDC thicknesses ranging from 1L to 200 nm in steps of 20 nm. The successive spectra are shifted along the y-axes by 0.05 for clarity.

for the substrate, we calculate r and t for several $\beta_i = \frac{2\pi n_i d_i}{\lambda} + \phi_m$ and average those over [6]. Here ϕ_m 's are equidistant phases between 0 and 2π , separated by $2\pi/m$, m being an integer. For instance, we use $m = 8$, where the ϕ_m 's are $0, \frac{\pi}{4}, \frac{\pi}{2}, \dots, 2\pi$. Our calculation reveals that the curves obtained through this method are extremely close to the reality [see Figs. S6(c) and (d)]. Therefore, we use this method everywhere in the present work.

It is important to note that the GTMM approach explicitly

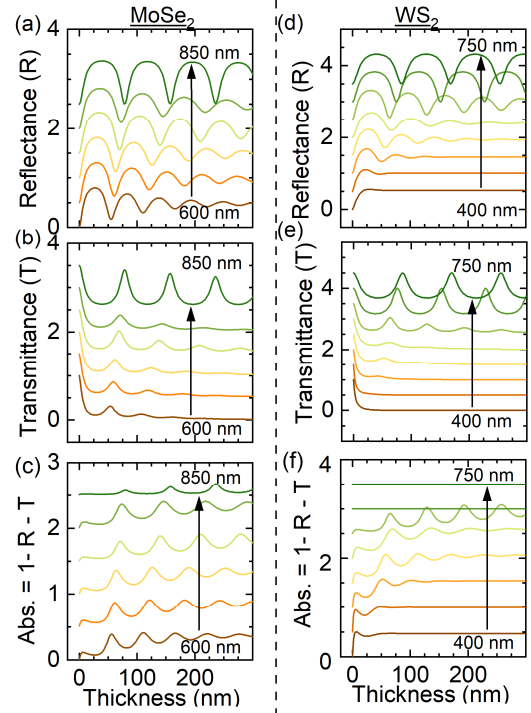


Figure S7. R, T and A calculated as a function of layer thickness for freestanding layers (no substrate) of (a–c) MoSe₂ and (d–f) WS₂ evaluated at selected wavelengths at 50 nm interval. For clarity, successive curves are vertically offset by 0.5 units. The oscillatory features due to interference effects continue to persist in the absorption curves as well. The corresponding integrated absorption curves are given in Fig. S8.

accounts for the interference effects between the light waves in each medium, and, thereby, the formation of standing waves within the system, whereas the Beer-Lambert-based ray tracing method does not.

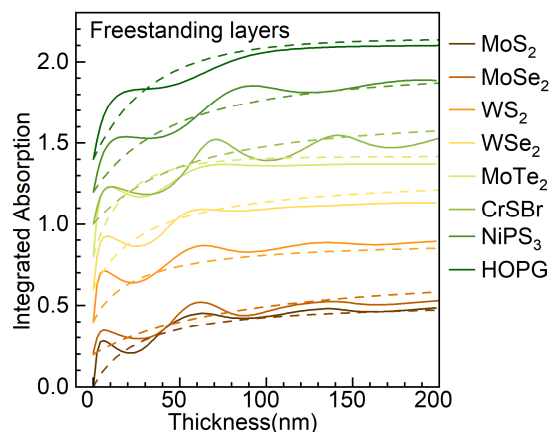


Figure S8. Calculated integrated absorption spectra of the freestanding MoS_2 , MoSe_2 , WS_2 , WSe_2 and MoTe_2 , CrSBr , NiPS_3 , c-HOPG (graphite), using GTMM, as a function of layer thickness with offset of 0.2 on y- axis. Clear oscillatory behavior riding on Beer-Lambert absorption is visible in all the materials.

Color of materials on PDMS

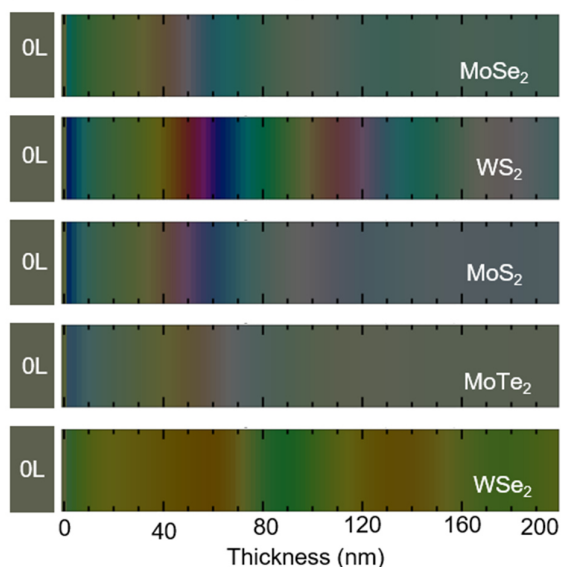


Figure S9. Perceived optical colors of MoSe_2 , WS_2 , MoS_2 , MoTe_2 , and WSe_2 on a polydimethylsiloxane (PDMS) as a function of layer thickness, calculated using transfer-matrix method. The leftmost panel ('OL') shows the calculated color of bare PDMS. The calculations are performed assuming illumination with a broadband white-light LED. With increasing thickness, each TMDC exhibits a characteristic progression of interference-induced colors. These distinct color sequences provide a rapid optical means for identifying and distinguishing thickness variations in TMD layers on PDMS.

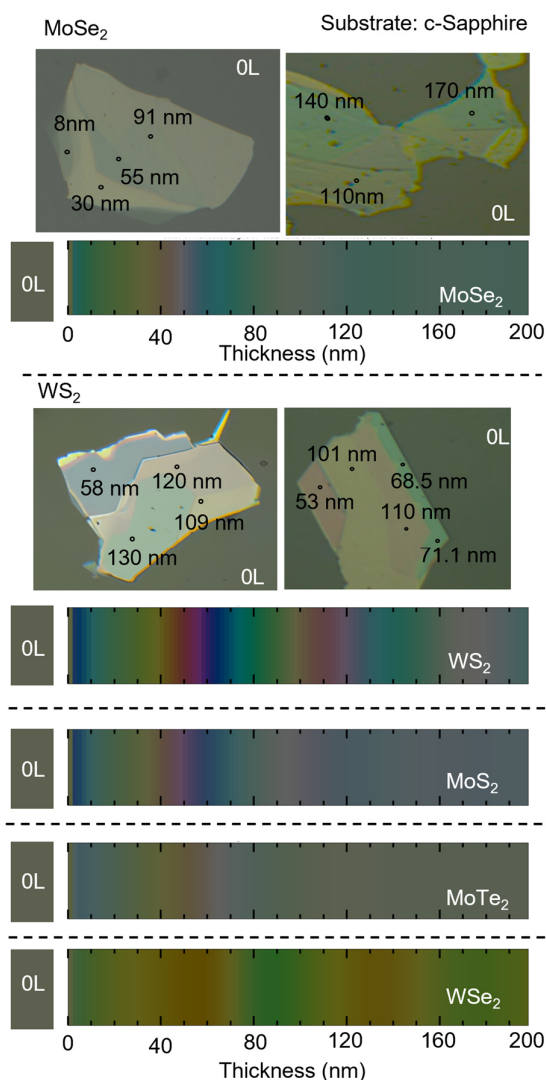


Figure S10. Optical images of exfoliated MoSe_2 (top) and WS_2 (middle) flakes on a 0.5 mm thick sapphire substrate. Thickness values marked on the flakes are obtained from atomic force microscopy (AFM) measurements. At the bottom of the optical images, a color bar is shown, which displays the expected color of the flakes of certain thicknesses kept on c-sapphire calculated using transfer-matrix method. The calculations are performed assuming illumination with a broadband white-light LED. The regions labeled 'OL' show the calculated color of bare substrate, providing the reference contrast for identifying the TMDC layers. The colors predicted by the calculations agree excellently with the colors in optical images. Simulated optical color maps of MoS_2 , MoTe_2 , and WSe_2 on the same substrate are also shown, highlighting the distinct thickness-dependent color evolution across different TMDCs.

Color of materials on SiO₂/Si (300 nm)

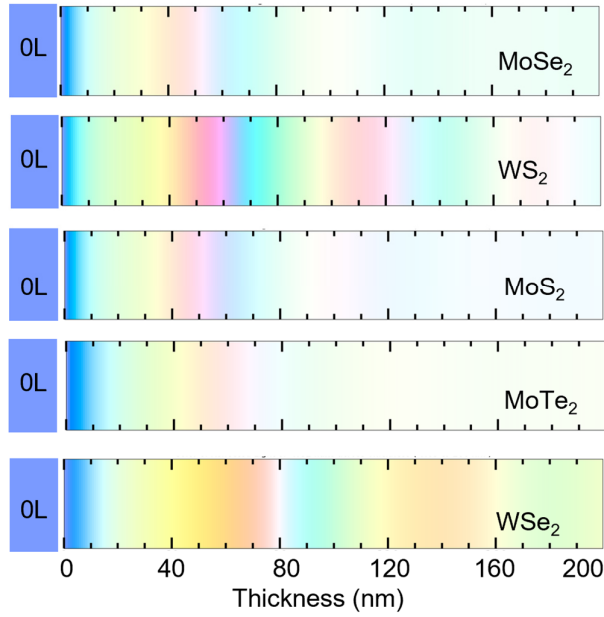


Figure S11. Perceived optical colors of MoSe₂, WS₂, MoS₂, MoTe₂, and WSe₂ on a 300nm SiO₂/Si substrate as a function of layer thickness calculated using transfer-matrix method. The leftmost panel ('OL') shows the calculated color of bare 300nm SiO₂/Si. The calculations are performed assuming illumination with a broadband white-light LED.

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Color of materials on SiO₂/Si (80 nm)

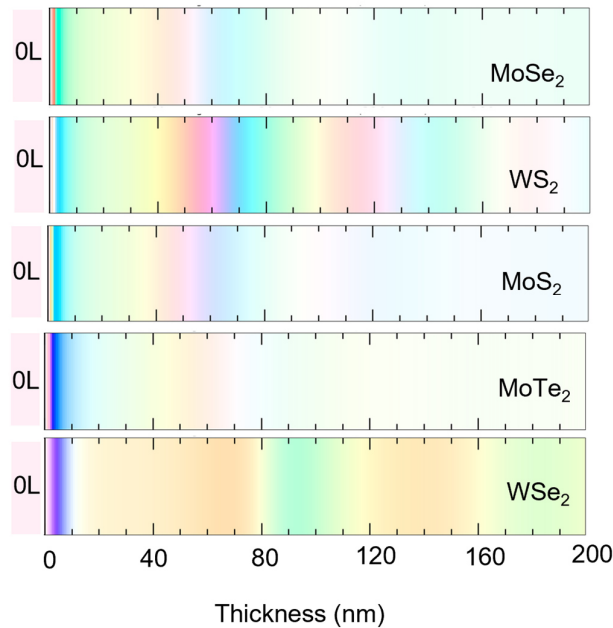


Figure S12. Perceived optical colors of MoSe₂, WS₂, MoS₂, MoTe₂, and WSe₂ on a 80nm (SiO₂/Si) substrate as a function of layer thickness calculated using transfer-matrix method. The leftmost panel ('OL') shows the calculated color of bare 80nm SiO₂/Si. The calculations are performed assuming illumination with a broadband white-light LED.