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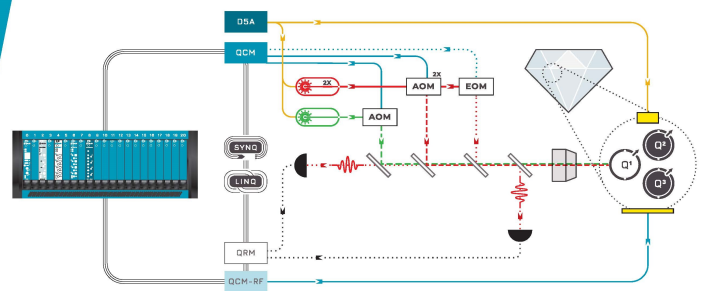
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ABSTRACT

We report the design and fabrication of a vertical structure using a distributed Bragg reflector and dielectric material layer to achieve optimized optical absorption enhancement for a stack of monolayer WS₂ and MoS₂, namely, a tenfold increase in absorption over a 100 nm spectral range. Our research indicates that we can approach over 50% absorption by finely tuning the thickness of the spacer layer. Our theoretical model shows that the dependence of the absorption coefficient on the spacer thickness can be understood as a solution of a non-Hermitian Schrödinger equation. These results advance the development of broadband optical devices, including solar energy conversion and sensitive optical sensors, by using two-dimensional excitonic materials.

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Modifying light absorption while diminishing its reflection is one of the most fundamental processes in light-matter interactions and is critical for designing modern optoelectronic devices, for example, signal conversion and energy harvesting. Controlled absorption of light has been used in optical sensors,¹ photodetectors,² photodiodes,³ solar cells,^{4–6} photocatalysis,^{7,8} and luminous energy storage.^{9,10} Using monolayers aids the general requirement for on-chip and integrated devices. One method is to use the destructive interference of light,^{11,12} which often involves complicated designs and complex structures.^{13,14} In contrast, in this paper, we consider a dissipative optical system that is able to localize a photonic state to enhance the intrinsic optical

absorption of two-dimensional materials. By customizing the optical resonances to match the absorption energy bands in the materials, the optical absorption of the devices can thus be strongly modified. This involves taking into account the real and imaginary terms of the complex susceptibility. As shown below, this theoretical model maps to the case of a Schrödinger equation with a non-Hermitian Hamiltonian.

Two-dimensional transition metal dichalcogenides (2D TMDCs) are promising candidates as building blocks for next-generation optoelectronic devices not only for their thinness but also for their ultra-large exciton binding energy¹⁵ and generally excellent optical properties.^{16,17} The layers can be stacked together by interlayer van

der Waals force in many ways, like building with “Lego” blocks.^{18,19} When two TMDC monolayers are used to make a bilayer, exotic optical phenomena can appear that depend on the relative angle between the crystal axes of the two layers.^{20,21}

Although these monolayers have a relatively strong light absorption in a visible light range, as high as 5%–10%,²² this absorption efficiency is not sufficient for practical application in optoelectronic devices. Therefore, it is a design goal to extract the greatest possible energy using these atomic layer materials.^{23,24} In recent years, studies have been performed on enhancing the absorption rate of TMDCs, especially using metal layers to enhance absorption.^{25–28} Although the preparation of structures with metal layers is convenient, the reflectivity of the thin metal layer is not high, and the reflectivity cannot be flexibly adjusted to different wavelengths, resulting in a poor absorption enhancement effect. Some other groups reported methods that exploit grating and photonic crystal structures to increase optical absorptions.^{29,30} However, those structures are often more complex designed and need more sophisticated fabrications.

Here, we propose to approach the absorption of the monolayers using a distributed Bragg reflector (DBR). The absorption can be tuned by the variation of the thickness of the dielectric material layer. In addition to offering a strongly increased absorption in a monolayer, we find that our devices offer an attractive route toward the development of broadband light absorption enhancement by taking advantage of the easy stacking methods to form van der Waals heterostructures. This strategy presented in this work offers high universality and, in particular, can be required in solar energy conversion and sensitive optical sensors applications.

Our system structure is designed based on the theory of non-Hermitian optics.³¹ Mathematically, the reflection of normally incident light by layered dissipative media is equivalent to the one-dimensional scattering problem of a non-Hermitian Schrödinger equation with complex potential V_c . Assuming the input light is propagating along the x -direction, we have $V_c(x) = k_0^2[1 - n(x)]$ with $k_0 = 2\pi/\lambda$ the wavenumber in vacuum and $n(x)$ is the local (complex) refractive index. Varying the thickness of different layers, such as the spacer and the hexagonal boron nitride (hBN), is equivalent to changing the width of the potential wells in $V_c(x)$, which can be used

to adjust the optical reflectivity of the system. Via fine-tuning of the thickness of specific layers, the optical absorption of the light in this structure can be enhanced well beyond 50%, which is known to be the upper bound of single-pass light by a thin absorbing layer,^{32,33} and even achieves perfect absorption if a non-Hermitian bound state is tuned to be resonant to the incoming light.³⁴ Furthermore, since both the complex potential V_c and the effective energy of the input light are wavelength-dependent, the optical reflectivity for different wavelengths can be changed separately, which aids the design of devices with broadband absorption.

The multi-layer structure was obtained by alternately growing two dielectric materials for multiple periods (here, the samples have eight periods) to get high light reflectivity in a particular wavelength band called the stop band. By changing the thickness of the grown materials, we can modify the stop band. First, a DBR with a stop band width of ~ 100 nm and a center wavelength of ~ 630 nm was grown. Then, monolayers of WS_2 ($\text{ex}_A \sim 610$ nm) and MoS_2 ($\text{ex}_A \sim 660$ nm) were used as the absorbing materials since the two materials possess similar band structures, with optical transitions within this stop band. Finally, between the WS_2 and DBR, and between the MoS_2 and the WS_2 , a spacer layer of silicon dioxide (SiO_2) and a thin flake of hBN were inserted, respectively.

Figure 1(a) illustrates the schematic of the full structure design on the order of DBR/spacer/ WS_2 /hBN/ MoS_2 from the bottom up. Figure 1(b) shows the transfer-matrix simulation results of the reflection of the full structure. The red line is the reflectance of the bare DBR. It indicates that the stop band is from about 560 to 690 nm. The blue line is the calculated reflectance of the full structure. We can see two deep absorption valleys where the monolayer absorption occurs, at 610 and 660 nm, within the stop band of DBR. When thermal broadening of these lines at room temperature is taken into account, they will merge, resulting in broadband absorption.

To tune the thickness of the spacer, we transferred the monolayer WS_2 onto the top of the DBRs with different spacer thicknesses. The DBR consisted of alternating layers of SiO_2 and silicon nitride and was grown on a silicon substrate by the Plasma Enhanced Chemical Vapor Deposition (PECVD). With the same system, the different spacer layers were deposited on each DBR, with bare silicon exposed

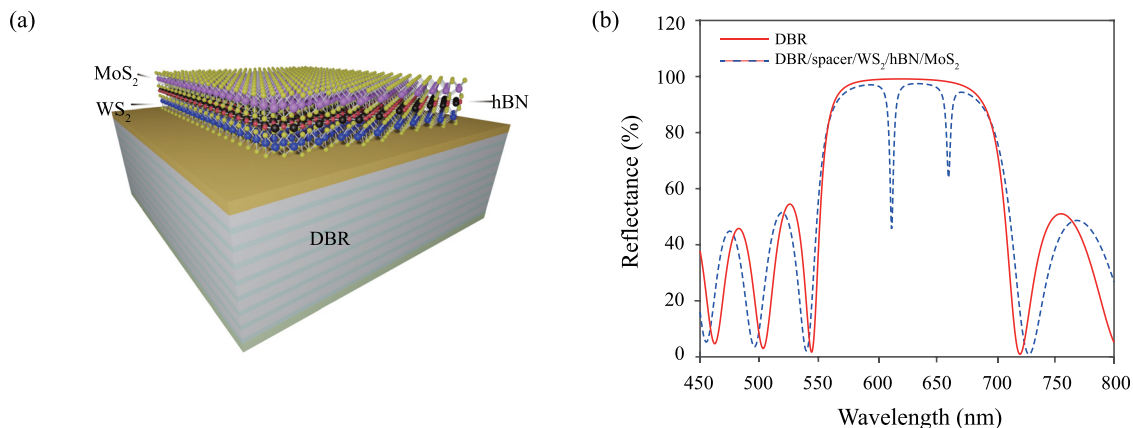


FIG. 1. (a) Schematic diagram of the designed structure with the order of DBR/Spacer/ WS_2 /hBN/ MoS_2 . (b) The simulated reflectance by transfer matrix method. The red line is the reflectance of a bare eight periods DBR. The blue dotted line is the reflectivity of the entire structure with a 100 nm SiO_2 spacer layer and 10 nm hBN flake accordingly.

alongside as a control. The thickness of the spacer layer was determined by a Filmmetrics system. Isolated monolayers of WS₂ with the typical size of $10 \times 10 \mu\text{m}^2$, as shown in Fig. 2(a), were mechanically exfoliated from bulk crystals. The whole samples were fabricated by the standard dry transfer method to the surface of the DBR substrates with the different spacer layers of SiO₂ coating. An Argon laser (532 nm) with a spot size of $1 \mu\text{m}$ at normal incidence, focused through a $100\times$ microscope objective, was employed as the pump source. The PL spectra were collected by the same microscope objective and directed toward a charged couple device (CCD) equipped spectrometer to help prescreen the monolayers. The differential reflectance spectra were carried out in the same configurations but with a mirror in the middle of the light path that could be flipped to block the laser while letting the white light go through. The white light size was cut down to $2 \mu\text{m}$ by an aperture and thus can precisely illuminate the area of interest. Six sets of samples were made with the thickness of the spacer layers ranging from 60 to 160 nm. A maximum of 50.6% absorbance was obtained in the sample with the spacer layer thickness of 100 nm. To quantify whether or not this absorbance is significantly sensitive to the spacer thickness, finite-element modeling using COMSOL Multiphysics 6.0 was performed. We set the center

wavelength of the stop band at 630 nm and varied the thickness of the spacer layers. A plane wave was employed in the model. The simulation results and the experimental data were calculated using the following equation:

$$\frac{\Delta R}{R} = \frac{I_{\text{sub}} - I_{\text{samp}}}{I_{\text{sub}}} \quad (1)$$

Here, I_{sub} and I_{samp} stand for the reflected light intensity when the incident light sheds on the substrate and the sample, respectively. We plot the differential reflectivity as a function of the spacer thickness for wavelength ~ 610 nm (monolayer WS₂ ex_A) in Fig. 2(d). Both show the same behavior: the absorbance increases when the spacer layer is thin, reaches a maximum when the thickness is about 100 nm, and then decreases at greater thickness. A control sample was made of a monolayer WS₂ on a piece of bare silicon. As shown in Fig. 2(b), the absorbance of the WS₂ monolayer on the Si substrate is only 6.6%, which agrees with that of TMDC monolayers given in the other literature.²² We have achieved enhancement of the absorbance by a factor of 8 using the DBR. (The absorbance spectra of the WS₂ monolayer for the different spacer layer thicknesses are shown in supplementary Fig. S2.)

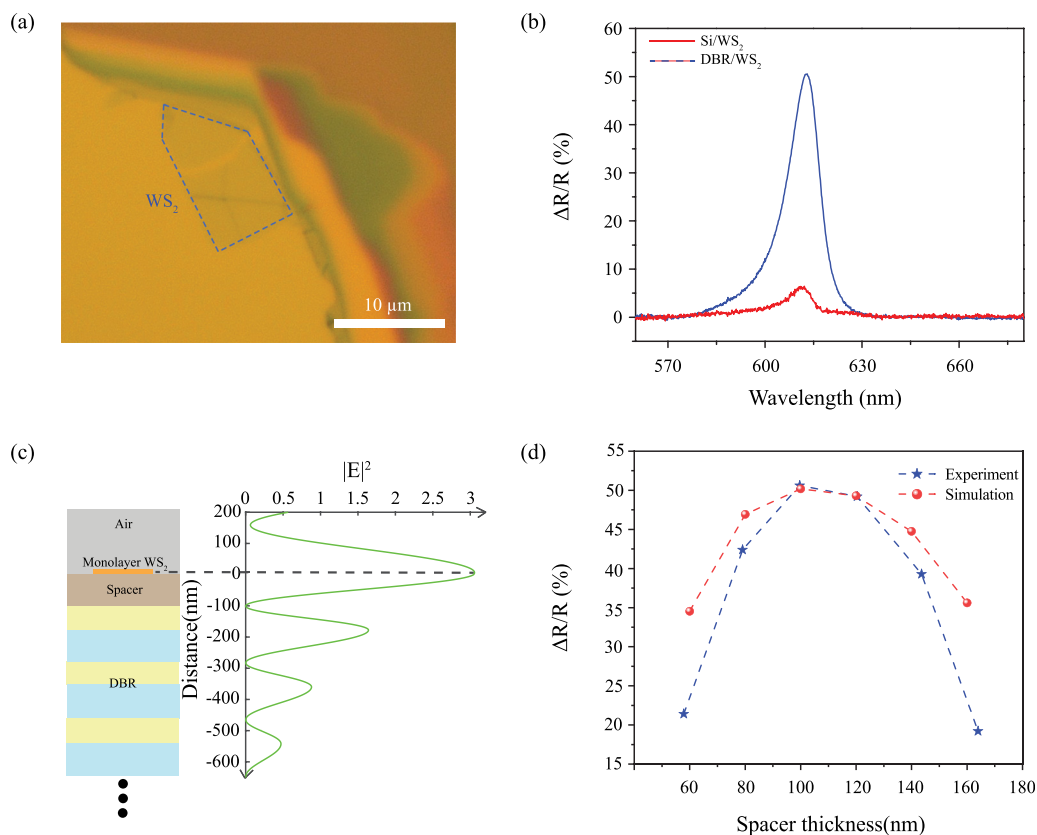


FIG. 2. (a) An optical microscope image of a WS₂ monolayer on a DBR with a SiO₂ spacer layer. (b) The differential reflectance spectrum of the monolayer WS₂ transferred on top of a DBR with a 100 nm SiO₂ spacer (in blue) compared to the same type of monolayer on a piece of bare silicon (in red). (c) Left: schematic of the sample, with the distances shown on the vertical axis of the plot on the right. Right: calculated electric field magnitude (assuming the $|E|^2$ of incident field is 1) as a function of distance. An anti-node of the field occurs at the location of the monolayer. (d) Spacer layer thickness-dependent differential reflectivity for wavelength ~ 610 nm. The blue dots are the experimental results for six different thicknesses. The red dots are from our simulation. Dotted lines connect the data points.

Figure 2(c) shows the simulated distribution of the electric field along in sample, which clearly shows that the maximum E-field intensity is at the position of the WS₂ monolayer. The light passing through the WS₂ monolayer is affected by the reflection of the DBR and the spacer layer. Theoretically, this structure can lead to photonic bound states located near the position of the WS₂ monolayer.

As a next step, a thin flake hBN was selected and used as another spacer layer, and then a monolayer MoS₂ was deposited. The hBN here plays two roles, both as a protector for the monolayer WS₂ beneath and as a dielectric medium to compensate for the spacer thickness mismatch. An atomic force microscope was used to determine the thicknesses of the hBN, giving the step profile in the inset of Fig. 3(a). According to our calculations, about 10 nm hBN was the best for realizing the broadband absorption enhancement. In Fig. 3(a), the monolayers of WS₂ and MoS₂ are highlighted with the blue and the red dotted lines, respectively, while the hBN is shown by the region below the black dotted line. The effective area (green highlighted) where the three materials overlap is about $5 \times 10 \mu\text{m}$. Thus, the interesting site can completely cover the white light spot with a diameter $\sim 2 \mu\text{m}$.

Figure 3(b) exhibits the differential reflectivity spectra of the full stack on the DBR compared to monolayer WS₂ and monolayer MoS₂ control samples on a bare Si substrate. As expected, the overlaid area possesses two dominant peaks, and they are the characteristic absorption peaks of the WS₂ and the MoS₂. Compared to the monolayers on the Si substrate, the absorptances increased from 6.6% to 32.2% and 2.2% to 26.9%, respectively. However, as seen by comparison to Fig. 2(b), the enhanced absorption of a single monolayer of WS₂ was reduced from 50.6% down to 32%, presumably because the monolayer is vulnerable to being affected by its environment. As shown in Fig. 3(b), the absorption peak of the WS₂ monolayer was at 616 nm, which is about 6 nm red-shifted compared to the WS₂ monolayer without capping; this can be attributed to the stress-induced deformation of the lattice, which can shift bandgap of the material due to the deformation potential effect.³⁵

We, thus, see that the absorption peaks of the two materials can be superimposed, with at least an absorption rate of 10% throughout a broadband spectral region.

To further illustrate the physical mechanism of the optical absorbing process in our system, especially the dependence of the absorption coefficient on the system structure, we have developed a theoretical model that relates our optical system to the non-Hermitian physics of a one-dimensional Schrödinger equation. As we know, non-Hermitian quantum mechanical systems have recently attracted great attention in a variety of subfields in physics including condensed matter, atomic gases, and optics as well.³⁶ The non-Hermitian optics described by our theoretical approach, thus, not only helps in understanding the absorption process in the systems from a different perspective but also provides a novel approach for designing and simulating various non-Hermitian quantum physics in two-dimensional monolayer systems.

For generality, we consider systems with one-dimensional spatial-dependent (absolute) permittivity $\epsilon(x)$ and assume a constant permeability $\mu \equiv \mu_0$, where μ_0 is the permeability of vacuum, as all materials involved in this work are non-magnetic. It is then straightforward to show that the electric field E satisfies

$$\frac{\partial^2 E}{\partial t^2} - \frac{1}{\mu\epsilon} (\nabla^2 E - \nabla(\nabla\epsilon^{-1} \cdot E)) = 0. \quad (2)$$

For an incoming plane wave propagating along the x -direction, we may write its electric field as $E = E(x)\hat{e}_z e^{-i\omega t}$ with ω being its frequency and $\hat{e}_z$ being the unit vector along the z -direction. Equation (2) can then be written in the form of a time-independent Schrödinger equation

$$-\frac{\partial^2}{\partial x^2} E + V_c(x)E = k_0^2 E, \quad (3)$$

where $k_0 = \omega/c = \omega\sqrt{\mu_0\epsilon_0}$ is the wavenumber in vacuum and the effective complex potential V_c is related to the local refractive index through $V_c(x) = \mu_0(\epsilon_0 - \epsilon(x))\omega^2 = k_0^2(1 - n(x))$.

The problem of calculating the absorbance in the experiment setup can then be mapped to a non-Hermitian scattering problem as illustrated in Fig. 4(a) (see the [supplementary material](#) for more details). One can see that the effective potential is complex because of

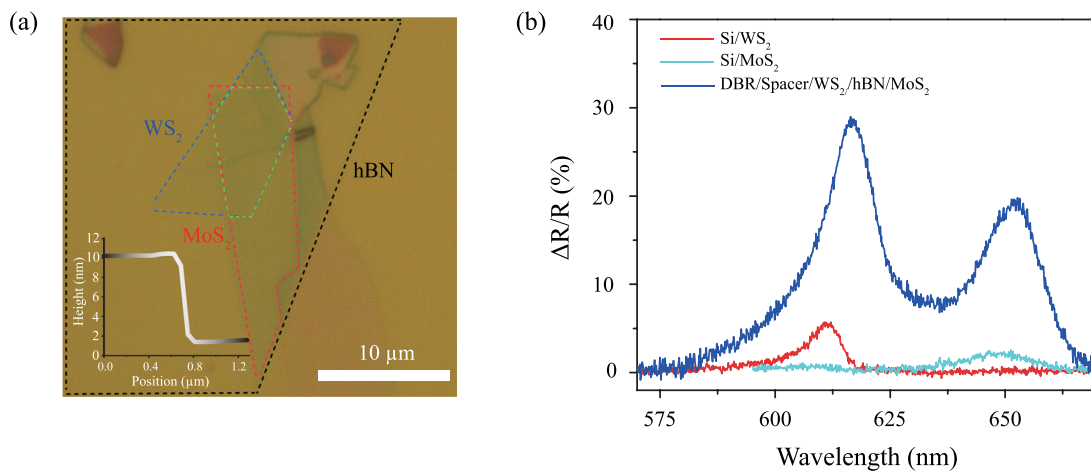


FIG. 3. (a) Image of the structure with the layers labeled in order of deposition. Inset is a clear step profile showing the thickness of 10 nm of the selected hBN measured by AFM. (b) Differential reflectance spectra for the entire structure and the control samples.

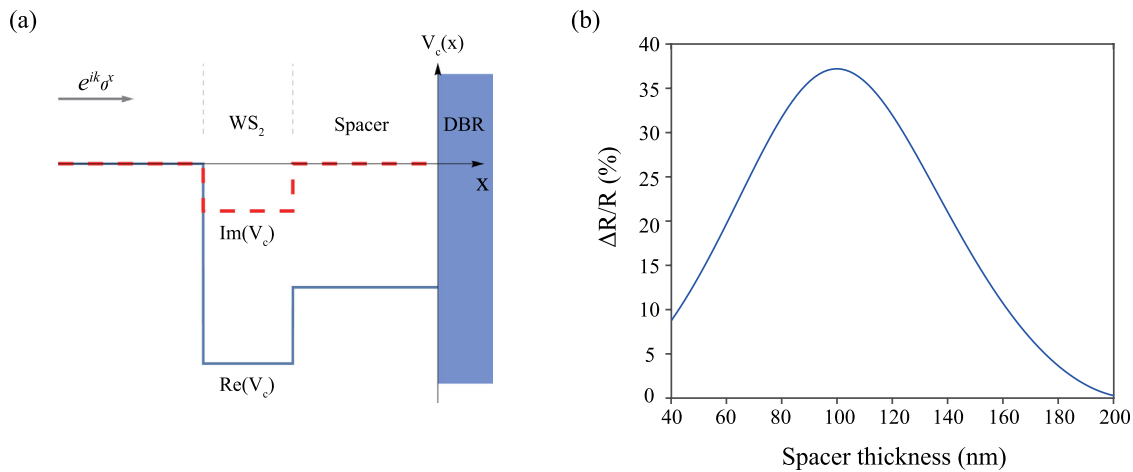


FIG. 4. (a) A schematic plot of the effective potential $V_e(x)$ for the non-Hermitian scattering problem for the WS₂-spacer-DBR structure. The potential has a nonvanishing imaginary part at the monolayer WS₂ because it is a dissipative material. (b) The calculated absorbance for 610 nm light as a function of the spacer thickness for the WS₂-spacer-DBR structure. The refractive index of WS₂ is obtained from Ref. 37.

the complex refractive index of monolayer WS₂. For further illustration, we theoretically calculate the absorbance of light with 610 nm wavelength in the WS₂-spacer-DBR structure and plot it as a function of the spacer thickness. The result clearly shows that the absorbance has a peak when the spacer thickness is around 100 nm, confirming the experimental measurements. It is worth mentioning that the theoretical curve presented in Fig. 4(b) does not exactly match the measured results in Fig. 2(d). The main reason that caused this deviation is that we have simplified the non-Hermitian scattering process by assuming that the DBR can be treated as a perfect mirror (an infinite hard wall in the language of the quantum scattering process). While in the experiment, the real DBR is never perfect, which necessarily causes some difference between the measured results and the simplified model. As a matter of fact, it would be quite easy to get rid of this deviation by choosing a “best fitted” reflection coefficient r in our theoretical calculation. The comparison of the experimental data and the theoretical calculation with various reflection coefficients can be found in the [supplementary material](#). Nevertheless, as shown in Fig. 4(b), the simplified theoretical model does not affect the optimal spacer thickness but only slightly changes the absorbance peak value. Furthermore, the calculation of the corresponding non-Hermitian scattering process through the DBR/spacer/WS₂/hBN/MoS₂ also helps verify the optimal experimental setup of the hBN thickness for achieving broadband absorbance.

The theoretical approach we used here is equivalent to the traditional method used in optics, such as the transfer matrix method. However, mapping the problem to a non-Hermitian scattering problem allows us to inspect the optical problem from a new perspective. For example, suppose we assume that all the mediums are non-dissipative. In that case, i.e., if the refractive index $n(x)$ is real everywhere, the Schrödinger equation (3) becomes Hermitian, and the optical theorem from Hermitian scattering theory³⁸ naturally implies that the absorbance must vanish. For non-Hermitian scatterings, it can be shown that a system with complex potential V_c can support bound states of energies with positive real parts.³⁹ Thus, through a proper design of the optical structure, one can fine-tune the energy of

a non-Hermitian bound state to be precisely in resonance with the effective scattering energy [i.e., k_0^2 as shown in Eq. (3)]. The “wavefunction” of the bound state then has the asymptotic form $E(x) \simeq e^{ik_0x}$ for $x \rightarrow -\infty$. Optically, this corresponds to a pure incoming plane wave without any reflection, and the system reaches critical coupling at this point.³⁴

In conclusion, we have explored the effect of the thickness of each spacer layer on the absorption of WS₂ and MoS₂ on a high-quality reflector. We have demonstrated that tuning the thickness of the spacer layer to form photonic bound states at the WS₂ monolayer and MoS₂ monolayer positions can induce strong absorption in two-dimensional materials, which may be useful for various applications, such as photon energy harvesting and sensitive sensing. Theoretically, we have shown that the optical system is equivalent to a non-Hermitian Schrödinger equation. By calculating the one-dimensional scattering problem of this Schrödinger equation, we obtain the absorption coefficient as a function of spacer thickness. The results agree with our experimental measurements. Our research shows that TMDC materials can have high optical absorption efficiency, atomically thin thickness, and scalability in applications, making them promising in studying modern optoelectronic devices.

See the [supplementary material](#) for the complete study of the spacer layer thickness-dependent differential reflectivity spectra and the theoretical details on non-Hermitian scattering.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Xingzhou Chen: Data curation (equal); Formal analysis (equal); Writing – original draft (equal). **Jian Wu:** Resources (supporting); Writing – review & editing (supporting). **Zheng Sun:** Conceptualization (lead); Project administration (lead); Supervision (lead); Writing – review & editing (lead). **Min Zhang:** Data curation (supporting); Formal analysis (supporting). **Ming Li:** Methodology (supporting). **Zhigao Hu:** Resources (supporting). **Kenji Watanabe:** Resources (supporting). **Takashi Taniguchi:** Resources (supporting). **David W. Snoke:** Writing – review & editing (equal). **Zheyu Shi:** Data curation (equal); Formal analysis (equal); Writing – review & editing (equal).

DATA AVAILABILITY

The data that support the findings of this study are available within the article and its [supplementary material](#).

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