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Intrinsic setting of the exciton state in MoS₂ monolayers *via* tailoring the Moiré correlation with a sapphire substrate†

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This study aims to elucidate the specific Moiré correlation and associated exciton properties within MoS₂ monolayers grown randomly oriented on a c-cut single-crystalline sapphire (Al₂O₃) substrate, which facilitates a distinct Moiré correlation. Notably, the exciton state in MoS₂ monolayers appeared periodically linked to the stacking geometry with the sapphire substrate. Specifically, the observed stacking configuration of MoS₂[112̄0]/Al₂O₃[112̄0] induced a red-shifted exciton state, while a 30-degree-misaligned stacking, such as MoS₂[112̄0]/Al₂O₃[011̄0], increased the exciton energy. The variation in the exciton state due to changes in the stacking geometry between MoS₂ and sapphire thus exhibited a 6-fold periodicity, reflecting the combination of hexagonal MoS₂ and the trigonal sapphire substrate. This transition in the exciton state of the MoS₂ monolayer was attributed to stacking-induced strain: the MoS₂[112̄0]/Al₂O₃[112̄0] stack resulted in a closely packed nature with induced tensile strain in the film plane, whereas the MoS₂[112̄0]/Al₂O₃[011̄0] stack appeared lightly packed, thus rendering the MoS₂ structurally relaxed. These findings regarding the stacking-induced strain issues are consistent with the results of Raman spectra. This work underscores the potential for manipulating the crystallographic stacking between single-crystalline MoS₂ monolayers and sapphire substrates to serve as a versatile platform for investigating photonics in MoS₂-based heterostructures.

1. Introduction

Transition-metal dichalcogenides (TMDs) in the form of monolayers have sparked significant interest due to their

unique physical properties.^{1–3} These properties include optics,^{4–6} electrical transport,^{2,7} piezoelectricity,^{8,9} and valley spintronics,^{10,11} driving advancements in two-dimensional (2D) semiconductor technologies. Photonics is a crucial area of studying TMD monolayers, given their direct bandgap,⁹ which expands the possibilities for semiconductor applications in visible-light (VL)-based photonics.^{12,13} Moreover, the sensitivity of TMD monolayers to VL makes them suitable for characterization using VL-based spectroscopic techniques such as Raman and photoluminescence (PL) spectroscopy using lasers of 633 nm or 532 nm. In the emerging field of twistrionics, it has been demonstrated that combining two TMD monolayers with specific crystallographic geometry can produce diverse Moiré patterns based on bilayer stacking, leading to significant changes in the properties of TMDs.^{14–16} Twistrionics explores how the packing of two TMD monolayers influences the formation of Moiré patterns, offering insights into *ex situ* treatments for enhancing or modifying TMD properties. It also provides a new dimension to tailor TMDs to fit various facets of application. However, an important yet overlooked aspect of twistrionics is the growth of TMD monolayers on a single-crystalline substrate with specific stacking geometry. Understanding the pseudo-Moiré correlation in TMD monolayers on single-crystalline substrates is crucial for establishing a strong interaction between physical properties and stacking geometry, mirroring the current focus on twistrionics but presenting an unresolved challenge.

This study attempts to explore the effect of the stacking geometry, as well as the Moiré correlation, of an MoS₂ monolayer grown on a c-cut single-crystalline sapphire (Al₂O₃) substrate using a chemical vapor deposition technique. Without the pre-treatments utilizing annealing for establishing atomic steps on the sapphire surface before deposition,^{17,18} the synthesized MoS₂ monolayers appeared to be randomly orientated on the sapphire surface, providing a unique playground to study the variation of the photonics properties of MoS₂ monolayers arising from the specific Moiré pattern *via* layer stacking. This work would give another insight into the current twistrionics and may pave an avenue toward structurally tailorable 2D device technologies.

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2. Experimental

MoS₂ monolayers were grown by using a chemical vapor deposition (CVD) technique in a 3-temperature-zone furnace, in which the sulfur powders (Alfa Aesar, 99.5%) of 0.3 g were placed at the edge of zone 1, MoO₃ powders (Alfa Aesar, 99.95%) of 2 mg were placed at zone 2, and the sapphire substrates with c-cut surface were positioned 7 cm downstream from the MoO₃ source at zone 3. Before CVD, the furnace was purged using an Ar flow for three consecutive cycles to maintain a contamination-free environment in the beginning. After the Ar purging, the temperatures at zones 1, 2, and 3 were ramped up to 550, 750, and 850 °C, respectively, in 90 minutes. The sulfur powders were subsequently heated up to 200 °C to supply the sulfur vapors interacting with MoO₃ vapors at zone 2. During the experiment, an Ar carrier gas of

100 sccm was applied throughout the process to carry the hybrid S/MoO₃ atmosphere to zone 3 and then the deposition on the surface of sapphire substrates was carried on for 15 minutes. Finally, the samples were cooled down to room temperature in the furnace for approximately 2 hours. The morphology of MoS₂ monolayers was pictured using an optical microscope to examine the domain orientations on a sapphire substrate as shown in Fig. 1(a). The Raman and photoluminescence spectra were acquired using a Raman spectroscopy system with an argon laser source of 533 nm at room temperature. The laser beam was focused onto the sample's surface and the spot size was approximately 1 μm. The continuous-wave (CW) laser power was 75 mW, and a 1200 line per mm grating was employed for an energy resolution of 2 meV. The integration time for collecting Raman and PL spectra was 30 and 5 seconds to yield distinguishable signals,

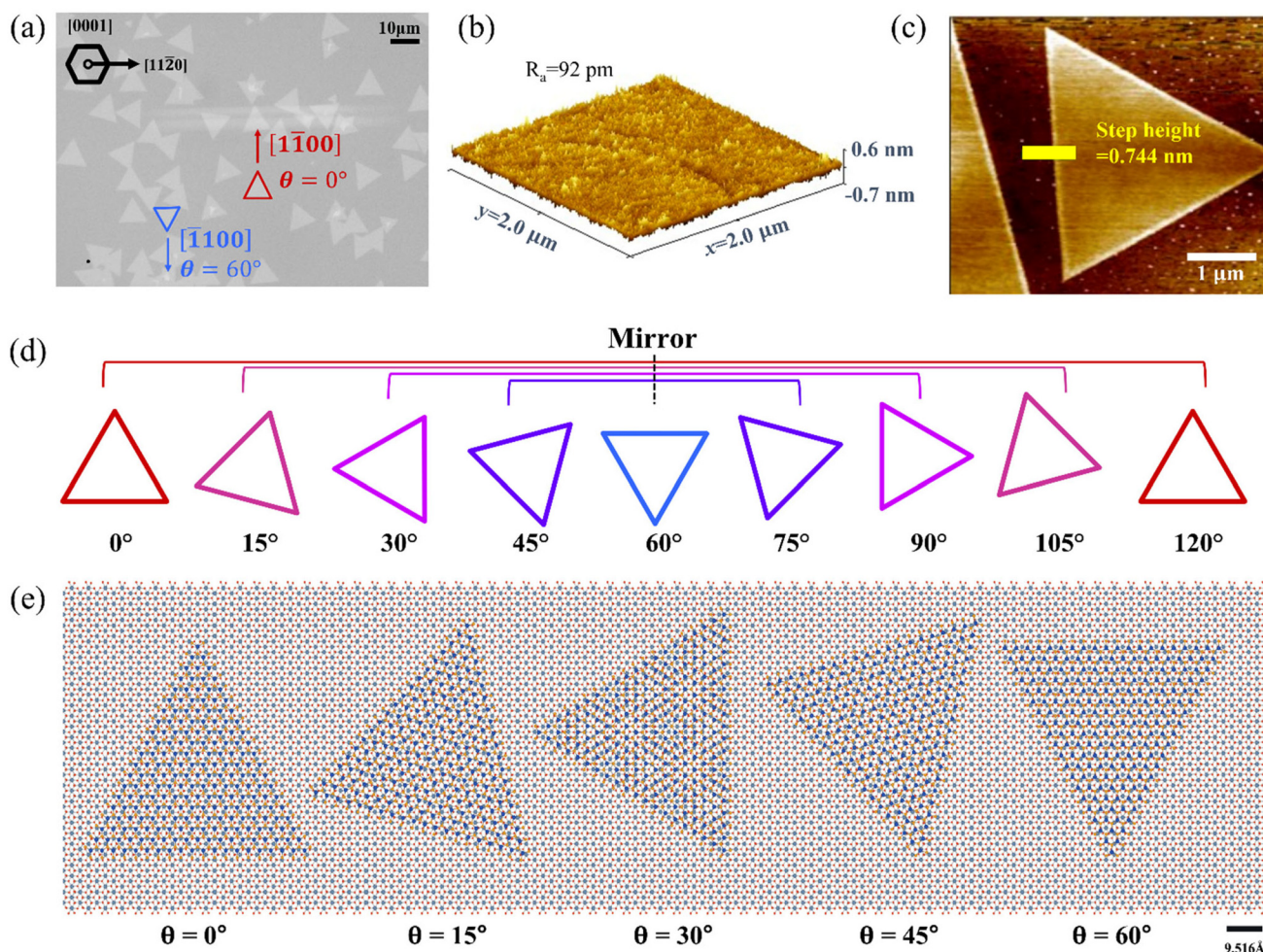


Fig. 1 (a) Optical microscopy image of MoS₂ monolayers distributed on the sapphire substrate with well-defined crystallographic orientations. (b) Atomic force microscopy image showing the surface roughness of the bare Al₂O₃ substrate employed in this study. The roughness (R_a) is approximately 92 pm as an atomically flat condition for studying the Moiré correlation in the MoS₂/Al₂O₃ stack. (c) Atomic force microscopy image showing the MoS₂/Al₂O₃ stack, in which the step height of MoS₂ is 0.744 nm, corresponding to the thickness of the monolayer. (d) θ -Dependence and the periodicity of MoS₂ monolayers defined via the distribution in (a), in which $\theta = 0^\circ$ stands for aligning MoS₂[1 $\bar{1}$ 00]/Al₂O₃[1 $\bar{1}$ 00]. $\theta = 15^\circ$ and 45° are crystallographically identical to $\theta = 75^\circ$ and 105° , respectively, due to their mirror stacking geometry in both hexagonal lattices. (e) The simulated Moiré patterns upon growing MoS₂ monolayers on sapphire with various θ . The simulated patterns are obtained using Vesta software.



respectively. All the PL and Raman spectra were acquired from the samples at 298 K under the atmosphere.

3. Results

The MoS₂ monolayers (space group: $P\bar{6}m2$, $a = b = 3.19214$ Å and $c = 13.37829$ Å)¹⁹ grown on a c-cut single crystalline sapphire (Al₂O₃, space group: $R\bar{3}C$, $a = b = 4.75815$ Å and $c = 12.98970$ Å)²⁰ substrate investigated in this study are randomly orientated, as shown in Fig. 1(a). Fig. 1(a) also defines the crystallographic correlation between MoS₂ monolayers and the sapphire substrate, in which we assign the stacking angle (θ) = 0° for the alignment through MoS₂[$\bar{1}100$]/Al₂O₃[$\bar{1}100$], as labeled in Fig. 1(a). The sapphire substrate utilized in this work was atomically flat with a surface roughness of ~ 92 pm as shown in Fig. 1(b). Fig. 1(c) shows the atomic force microscopy image of the sharp edge of the MoS₂ domain, verifying the step height of the monolayer of ~ 0.744 nm. In

Fig. 1(a), the MoS₂ monolayers with varying θ rotating clockwise can be observed on the atomically flat sapphire substrate, which may be due to the growth mechanisms lacking preferred nucleation sites during the chemical vapor deposition.^{21,22}

Fig. 1(d) demonstrates the correlation of the varying θ in reference to the crystallography of the sapphire substrate. It should be noted that the period of the varied θ shows a 6-fold symmetry based on the crystallographic correlation between MoS₂ monolayers and the sapphire substrate. Therefore, the resulting stacking patterns are demonstrated in Fig. 1(e), in which the pattern appears to be in a 6-fold periodicity, providing a model system to use spectroscopy to resolve the effect of stacking geometry.

Fig. 2(a) and (b) display the θ -dependent PL spectra of MoS₂ monolayers on the Al₂O₃ and SiO₂ substrates, respectively. Note that the SiO₂ utilized is an amorphous substrate and herein MoS₂[$\bar{1}100$]/Si[100] is set as $\theta = 0^\circ$ for a reference to verify the θ -dependence in the MoS₂/Al₂O₃ stack. Consequently, both systems feature the exciton state in MoS₂,

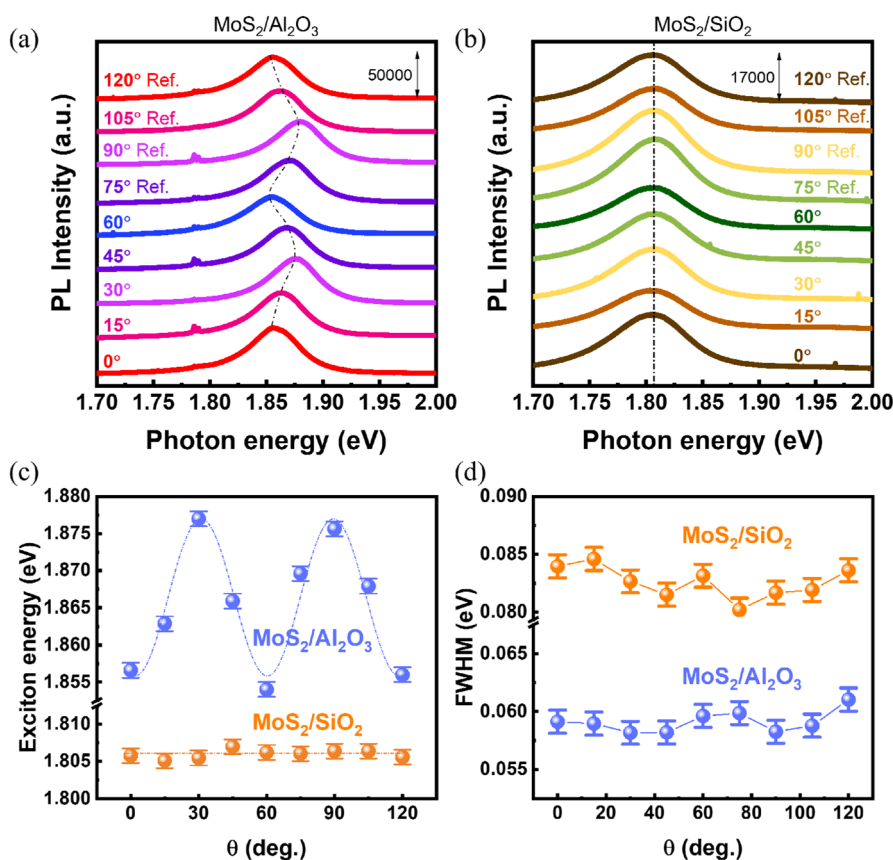


Fig. 2 θ -Dependent PL spectra acquired from the (a) MoS₂/Al₂O₃ and (b) MoS₂/SiO₂ stacks. Note: the spectra in (a) and (b) taken for $\theta = 75, 90$, and 105 degrees are arbitrarily selected to extend the 6-fold correlation for a clear comparison, and signals at approximately 1.785 eV in (a) correspond to the Raman response of the Al₂O₃ substrate and are independent of varying θ . (c) Changes of exciton energy as a function of θ acquired from the MoS₂/Al₂O₃ and MoS₂/SiO₂ stacks as the representatives for the well-defined crystallographic correlation and the absent structural correlation, respectively. Herein, the $\theta = 0^\circ$ of the MoS₂/SiO₂ stack is defined by aligning MoS₂[$\bar{1}100$]/Si[100]. (d) Full width at half maximum (FWHM) of PL spectra as a function of θ acquired from the MoS₂/Al₂O₃ and MoS₂/SiO₂ stacks. The error bars in (c) and (d) were determined by the energy resolution of PL spectroscopy ~ 2 meV. The PL spectra were acquired using a Raman spectroscopy system with an argon CW laser source of 533 nm of 75 mW at room temperature under the atmosphere.



which serves as a spectroscopic signature of the monolayer. However, the θ -dependent PL spectra acquired from the MoS₂/Al₂O₃ stack show significant 6-fold periodicity as indicated by the dashed line as a guide to the eye. In contrast, this 6-fold periodicity does not appear in the MoS₂/SiO₂ heterostructure. Note that all the PL spectra were acquired at the center of MoS₂ domains to eliminate the effects of structural imperfection in the vicinity of the edge (see ESI †). In order to clearly demonstrate the 6-fold periodicity in the MoS₂/Al₂O₃ stack, Fig. 2(c) displays the plots of the exciton energy *versus* the change of θ for the MoS₂/Al₂O₃ stack alongside the results taken from the MoS₂/SiO₂ stack, and their full width at half maximum (FWHM) is shown in Fig. 2(d). The former follows an ideal 6-fold periodicity as expected from the schematic diagram in Fig. 1(e) and the 6-fold periodicity vanishing in the latter case should be attributed to the amorphous nature of SiO₂, giving rise to the absence of crystallographic correlation while stacking. It could be noticed that the FWHM is less sensitive to the θ dependence and is nearly constant for both cases; therefore, the observed PL shifting may be attributed to the band structural modification rather than the n-doping associated transition, *i.e.* exciton to trion.^{23,24} In addition to the vanished 6-fold periodicity, the redshifted exciton of MoS₂ on SiO₂ (to ~ 1.805 eV) was observed, which might be attributed to the transition from exciton to trion as a result of the

extra n-doping effect as observed in the CVD-grown MoS₂ on the SiO₂ substrate relative to sapphire.^{25,26} Besides, the broadening PL FWHM taken from the MoS₂/SiO₂ stack may also reveal the exciton to trion transition and superimposition accompanying the n-doping effect.^{25,26} Again, the data of $\theta > 60^\circ$ were selected for arbitrary references because those are crystallographically repeated as conceptually demonstrated in Fig. 1(d) and (e).

Based on the results in Fig. 2(a) and (b), it has been shown that the exciton state in MoS₂ monolayers was highly correlated with θ in the MoS₂/Al₂O₃ stack, namely the Moiré correlation between MoS₂ monolayers and the Al₂O₃ substrate. While revisiting the θ -dependent Raman spectra of the MoS₂/Al₂O₃ and MoS₂/SiO₂ stacks shown in Fig. 3(a) and (b), respectively, a similar 6-fold periodicity can be observed in MoS₂/Al₂O₃ and is also absent in MoS₂/SiO₂. Fig. 3(c) and (d) present the plots of the A_{1g} and E_{2g}¹ state of MoS₂ for the cases of MoS₂/Al₂O₃ and MoS₂/SiO₂, respectively, in which the wave number differences (Δk) between E_{2g}¹ and A_{1g} are all approximately 20 cm⁻¹, which stands for the fingerprint of the MoS₂ monolayer.^{25,26} However, it can be seen that both the E_{2g}¹ and A_{1g} states were stiffened while the θ deviates from the 0°/60° correlation in the MoS₂/Al₂O₃ stack, and reach the maximum wave number value while θ reaches the 30°-associated counterparts, as shown in Fig. 3(c) and (d). According to the previous

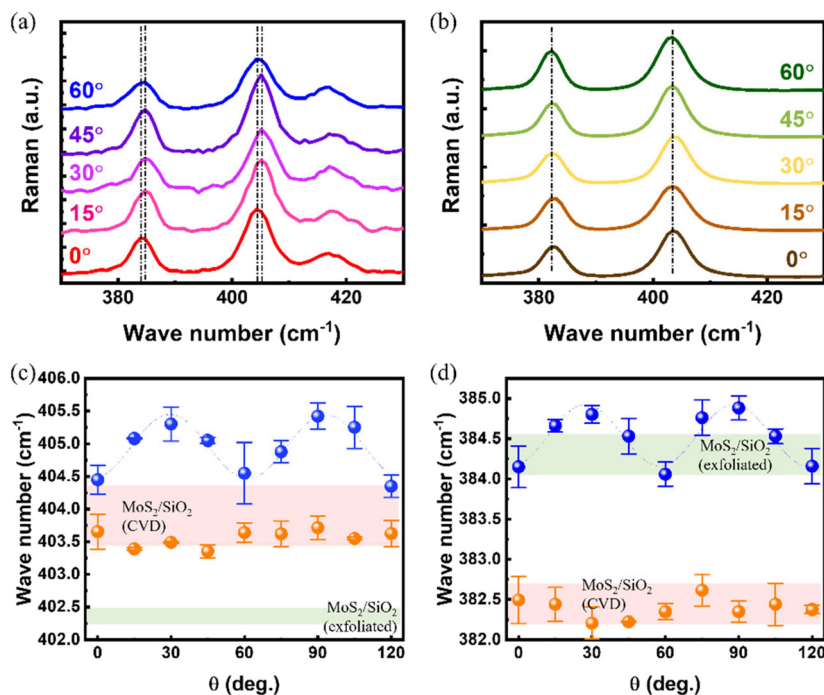


Fig. 3 θ -Dependent Raman spectra acquired from the (a) MoS₂/Al₂O₃ and (b) MoS₂/SiO₂ stacks. The wave number difference (Δk) between the E_{2g}¹ and A_{1g} states is approximately 20 cm⁻¹, suggesting the characteristics of MoS₂ monolayers. Plots of the (c) A_{1g} and (d) E_{2g}¹ states acquired from the MoS₂/Al₂O₃ and MoS₂/SiO₂ stacks with the θ -dependence. The Raman spectra were acquired using a Raman spectroscopy system with an argon CW laser source of 533 nm of 75 mW at room temperature under the atmosphere. The green and pink shadows in (c) and (d) highlight the E_{2g}¹ and A_{1g} states acquired from the MoS₂/SiO₂ stack fabricated using CVD and exfoliation methods for identifying the tensile strained/n-doped and strain-free/electrically neutral conditions of MoS₂ monolayers, respectively ref. 26. Error bars were defined by the difference between the peak positions of the E_{2g}¹ and A_{1g} states and the peaks fitted *via* the Gaussian–Lorentzian function.



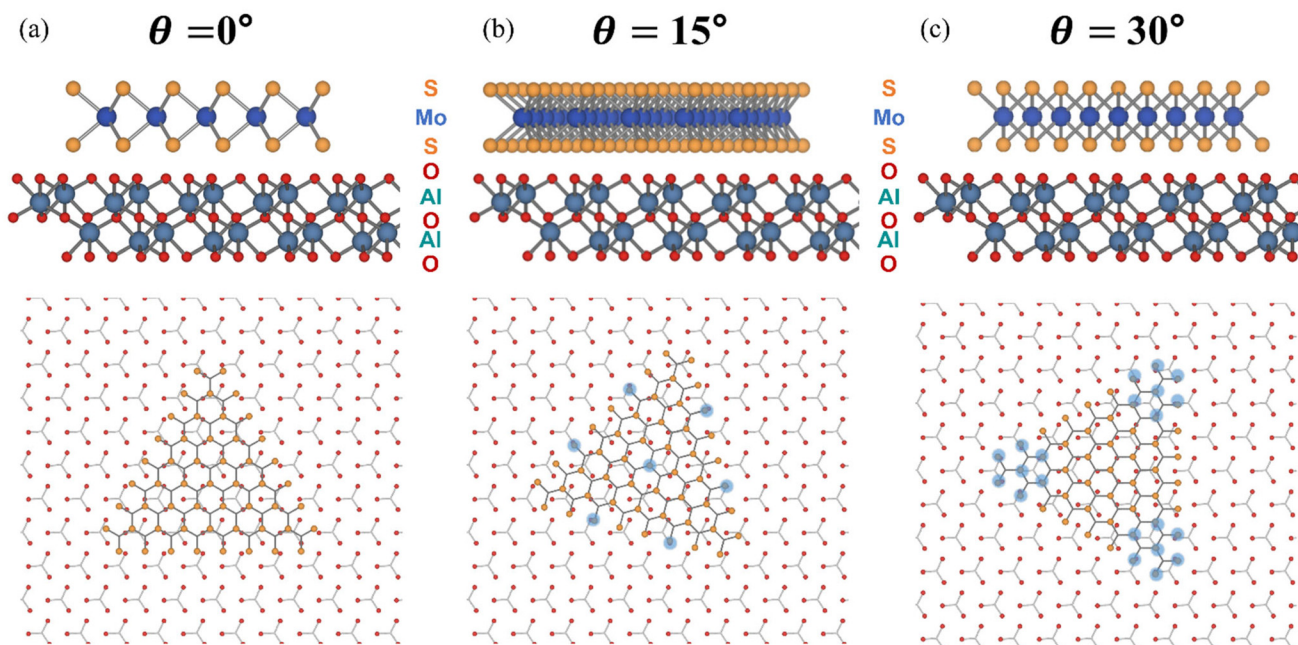


Fig. 4 Simulated $\text{MoS}_2/\text{Al}_2\text{O}_3$ stack images for (a) $\theta = 0$, (b) $\theta = 15$, and (c) $\theta = 30$ degrees via side-viewing (top panel) and top-viewing (bottom panel). The orange, blue, red, and green atoms represent S, Mo, O, and Al, respectively. The blue spots in the top-view images of each bottom panel highlight the site where the S and O atoms are superimposed, namely, the S atoms populate directly or partially on top of the O atoms. Note: for the top-view image, only the S and O atoms are left to demonstrate the superimposition issue because the S and O atoms are in the first contact layer.

literature, the spontaneous Raman shifting on the A_{1g} state is correlated with the n-doping effect led by charge-transfer from the substrates^{26,27} or the gating,²⁸ which echoes the exciton to trion transition on the PL results in Fig. 2(c). On the counterpart, Raman softening on the E_{2g}^1 state may be attributed to the isotropic tensile stress of the MoS_2 monolayers in the plane,²⁹ which may structurally result from the preferred atomic population during the growth of MoS_2 on the sapphire substrate, as reported in the epitaxially grown heterostructures, such as at the pyramid top sites on the substrate.^{30,31} The softened E_{2g}^1 state of MoS_2 on SiO_2 may be attributed to the isotropic tensile strain embedded during CVD growth, which is absent in the exfoliated MoS_2 on SiO_2 as demonstrated in the supporting information of A. Michail *et al.*'s study.²⁶ Fig. 4 series demonstrates the simulated $\text{MoS}_2/\text{Al}_2\text{O}_3$ stack for examining the θ dependence, in which the $0^\circ/60^\circ$ correlation (Fig. 4(a)) can always promote the S atoms at the bottom surface of MoS_2 to populate at the valley site among O atoms, suggesting the closely packed configuration at the $\text{MoS}_2/\text{Al}_2\text{O}_3$ interface. Therefore, the closely packed nature may give rise to the isotropic tensile stress and reduce the van der Waals gap distance accordingly. In contrast, for the misaligned cases such as the $\theta = 15^\circ$ (Fig. 4(b)) or $\theta = 30^\circ$ correlation (Fig. 4(c)), it can be seen that the S atoms are partially superimposed with O atoms from the top-view images, especially for $\theta = 30^\circ$, as highlighted by the blue spots. The result suggests that partial S atoms may populate directly on top of O atoms, hence generally giving rise to the longer van der Waals gap distance and reducing the horizontal tensile strain, conceptually demon-

strated in ESI 2.† Both results echo the Raman stiffening at the E_{2g}^1 and A_{1g} states while the θ deviates from the $0^\circ/60^\circ$ correlation.

4. Discussion

This study demonstrates a strong 6-fold correlation between the exciton state and the stacking geometry of MoS_2 monolayers on a sapphire substrate. Based on the previous literature, the variation in exciton energy in the $\text{MoS}_2/\text{Al}_2\text{O}_3$ stack may arise from three possibilities: (1) dielectric screening effects,^{32,33} (2) tensile strain-induced band structure modification,^{34,35} and (3) the doping effect.²⁶ The former two can be revealed through changes in the E_{2g}^1 state, and the latter one can be revealed through the A_{1g} state. Yuxuan Lin *et al.* reported that the dielectric screening effect can be tuned by varying the dielectric environment of the MoS_2 monolayer. Their study observed a blueshift of ~ 20 meV on PL upon increasing the solvent dielectric constant from 2 to ~ 33 .³³ In our case, the energy shift in the exciton state is approximately 30 meV from $\theta = 0^\circ$ to $\theta = 30^\circ$. Therefore, we can reasonably exclude the dielectric screening and doping effect as a striking factor in driving the exciton change, since the stacking geometry should only give rise to limited changes in the dielectric environment and the extra doping. Another scenario involves the issues of strain and stress. Zhiwei Li *et al.* reported that a tensile stress can be applied to the MoS_2 monolayer using a flexible membrane, PVA.³⁵ In this scenario, an energy redshift



of ~ 30 meV at the exciton state on PL together with an E_{2g}^1 shift of ~ 1 cm $^{-1}$ was observed when applying a tensile strain of $\sim 1.5\%$.³⁵ These results are very close to our observations in the θ -dependent PL and Raman spectra of the MoS₂/Al₂O₃ stack. It could be noticed that the exciton state of the strain-free MoS₂ in the study is approximately at 1.88 eV,³⁵ coinciding with the model as proposed in Fig. 4c for the case of $\theta = 30^\circ$. Besides, it has been reported that bandgap reduction³⁶ and the S vacancy may be attributed to the tensile strain,³⁷ which electronically serves as the n-doping source in the $\theta = 0^\circ/60^\circ$ stack, thus leading to the observation of A_{1g} softening. Although the thermally induced strain might also be considerable for observing the transition of the exciton state of the MoS₂ monolayer,³⁸ that should be still limited in this study because all the MoS₂ monolayers investigated were distributed on the same sapphire substrate with the nearly identical morphology. It should be noted that the effects of electron cloud redistribution due to changes in the van der Waals gap distance, which accompany variations in θ , cannot be entirely excluded. However, we still adopt a conservative stance on this matter due to the lack of direct evidence demonstrating the occurrence of electron cloud redistribution. Therefore, we believe, at this stage, that the strain/stress issue arising from the atomic stacking at the MoS₂/Al₂O₃ interface should play a major role in mediating the exciton state of the MoS₂ monolayer, which can be tailored through the Moiré correlation with the sapphire substrate.

5. Conclusions

In summary, this study explores the intrinsic tuning of the exciton state in MoS₂ monolayers grown on a c-cut single-crystalline sapphire substrate by manipulating different Moiré correlations, which has been overlooked in the past but may open another research branch in twistronics. By employing a chemical vapor deposition technique, the MoS₂ monolayers were observed to exhibit random orientation on the sapphire substrate, resulting in a wide distribution of stacking morphology. This variability provided an adaptable platform to investigate how specific Moiré correlations influence the exciton state. Interestingly, the exciton state reflected in the photoluminescence spectra appeared to be sensitive to changes in θ , exhibiting a 6-fold correlation during θ -dependent probing. A blueshift transition in the exciton state was observed as the structural stacking between MoS₂ monolayers and the sapphire substrate weakened, achievable by misaligning both $[\bar{1}100]$ and sapphire $[\bar{1}100]$. The stacking geometry would give rise to various degrees of packing density, hence leading to the variable tensile stress in the MoS₂ plane as a result of its lattice mismatch with Al₂O₃. Based on spectroscopic characterization, the intrinsic tuning of the exciton state seems to be adjustable based on specific Moiré correlations *via* altering the local contact configuration established in the sapphire/MoS₂ heterostructure, providing a versatile platform to explore associated physical properties such as charge-density-wave or band structural engineering.

Author contributions

CWC and FMC co-conceived and conducted the experiments under the supervision of CYY. LCC assisted in MoS₂ syntheses under the supervision of SKC and took care of the spectroscopic characterization. FMC aided in measurements of Raman and PL spectra. CWC performed the analyses and drafted the manuscript. CYY revised the manuscript and structured the logic of the study. All authors commented on the manuscript to improve the quality and readability.

Data availability

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Conflicts of interest

No potential conflict of interest was reported by the authors.

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