

Identification and Structural Characterization of Twisted Atomically Thin Bilayer Materials by Deep Learning

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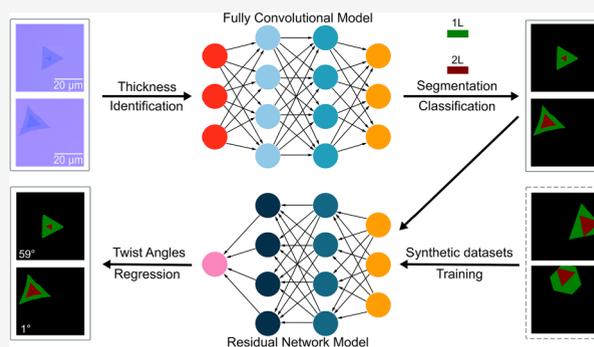
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ABSTRACT: Two-dimensional materials are expected to play an important role in next-generation electronics and optoelectronic devices. Recently, twisted bilayer graphene and transition metal dichalcogenides have attracted significant attention due to their unique physical properties and potential applications. In this study, we describe the use of optical microscopy to collect the color space of chemical vapor deposition (CVD) of molybdenum disulfide (MoS_2) and the application of a semantic segmentation convolutional neural network (CNN) to accurately and rapidly identify thicknesses of MoS_2 flakes. A second CNN model is trained to provide precise predictions on the twist angle of CVD-grown bilayer flakes. This model harnessed a data set comprising over 10,000 synthetic images, encompassing geometries spanning from hexagonal to triangular shapes. Subsequent validation of the deep learning predictions on twist angles was executed through the



second harmonic generation and Raman spectroscopy. Our results introduce a scalable methodology for automated inspection of twisted atomically thin CVD-grown bilayers.

KEYWORDS: *Twist angles, Transition metal dichalcogenides (TMDs), Deep learning, OpenCV, Raman*

Inspired by magic-angle graphene,^{1,2} twisted bilayer graphene and transition metal dichalcogenides (TMDs) have emerged as a promising platform for the study of Moiré physics, encompassing a range of phenomena such as Hubbard physics,^{3,4} superconductivity,⁵ or valley polarization.⁶ The twist angle in TMD bilayers can significantly alter their correlated electronic phases and their optical properties.⁷ For example, in the twist angle ($2^\circ \leq \theta < 6^\circ$), low-frequency interlayer shear and layer breathing modes exhibit rapid change with the twist angle θ .⁸ Additionally, the formation of the moiré Brillouin zone introduces new energy subbands in twisted MoS_2 bilayers with twist angles close to 0° or 60° ⁹ or high-lying excitons in bilayer WSe_2 , which can be tuned over 235 meV by enforcing different twist angles in the range 0° to 60° .¹⁰ The electric field control of the 2H bilayer MoS_2 interlayer exciton at room temperature is possible due to the out-of-plane electric dipole.¹¹ Chemical vapor deposition (CVD) can be used to fabricate bilayer graphene¹² and bilayer TMDs with different twist angles,^{13–17} i.e., different stacking arrangements between the two layers. Typically, only 0° (AA stacking, or 3R) or 60° (AB stacking, or 2H) arrangements are possible for the second layer on bilayer TMDs, as these are energetically more favorable.^{16,18} Reflectivity spectra have shown an A and B exciton energy difference of 49 meV between the 2H and 3R bilayer MoS_2 .¹⁵ Reverse-flow chemical vapor epitaxy provided

a way to controllably grow high-quality bilayer TMD single crystals with different growth temperatures.¹⁶

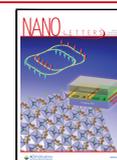
Second harmonic generation (SHG) spectroscopy is frequently employed to characterize the twist angles of TMDs.^{19,20} This reliable method determines the orientation of exfoliated flakes and subsequently enables the stacking of homo- or heterolayers using a dry transfer technique.^{21–23} In addition to SHG, differential reflectance spectroscopy can also be used for characterizing TMDs, specifically by identifying twist angles through the transition of interlayer excitons.^{15,24} Raman spectroscopy, particularly low wavenumber Raman spectroscopy,^{25,26} is another common method used for this purpose.^{8,16,27–29} For cases where atomic precision is required, transmission electron microscopy (TEM) can be employed to determine the twist angles of graphene.³⁰ Further augmenting the atomic precision of characterization techniques, scanning tunneling microscopy (STM) offers local probing of twist angles with atomic level resolution.³¹ Although accurate and reliable, these experimental techniques are costly, require

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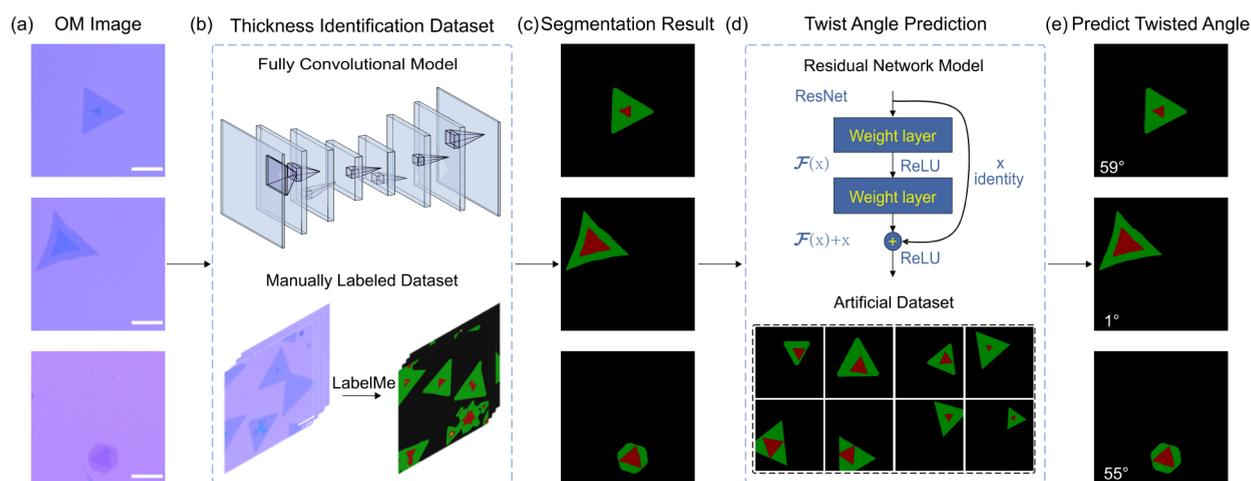


Figure 1. Identification and analysis of optical micrographs of CVD-grown bilayer atomically thin materials (e.g., TMDs) using deep learning. Images of the TMDs are shown as an example. (a) Original optical micrographs (OMs) captured by an optical microscope. (b) Images processed and labeled using LabelMe, followed by training via a convolutional neural network (CNN) employing various classification methods. (c) Typical outcomes of the TMD thickness derived from the processing in step (b). (d) The regression CNN model trained by the artificially generated data set for twist-angle prediction. (e) Twist angles predicted by using the CNN model from (d) are shown at the left corner of each image. Scale bar: 10 μm .

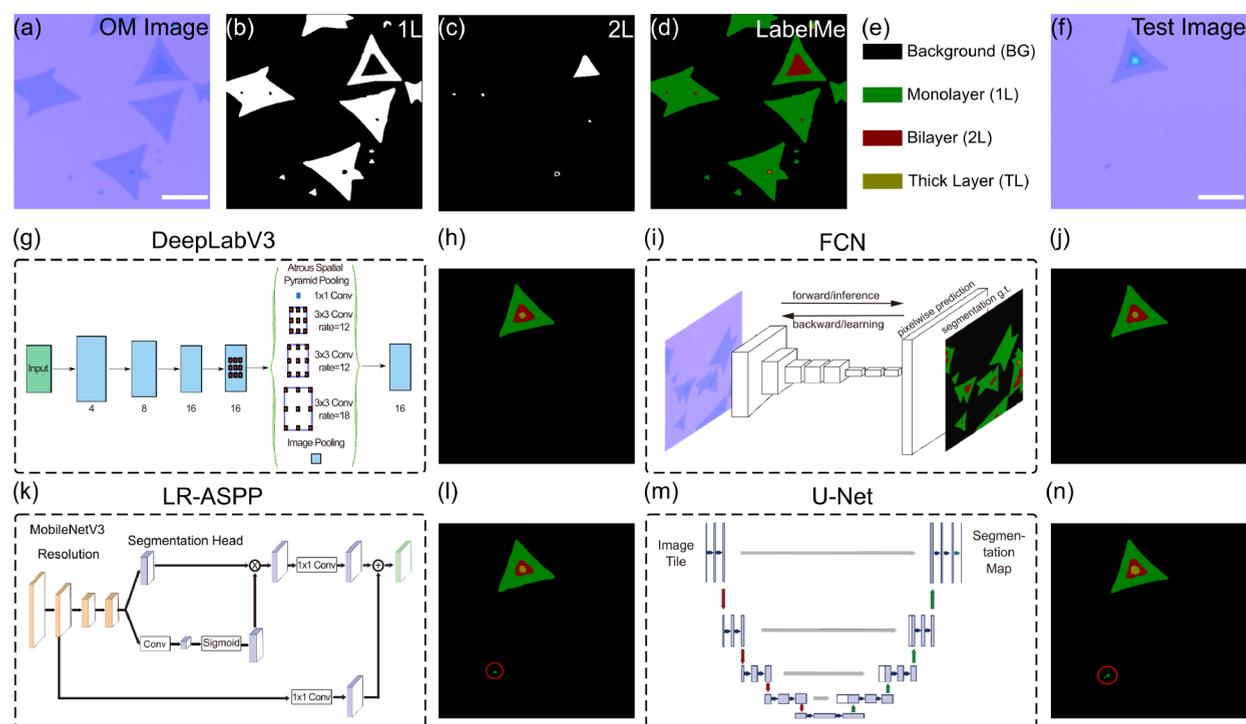


Figure 2. Segmentation techniques for classifying thickness in atomically thin CVD-grown bilayer flakes. (a) Optical micrographs of a bilayer MoS_2 flake. (b,c) Detail of the labeling for single and bilayer regions. (d) Final labeled result identifying thickness variations. (e) Explains the color labels, correlating colors to specific thickness levels. (f) Optical micrographs of another bilayer flake used as a test image. (g–n) Four CNN models and segmentation results employing DeepLabV3, FCN, LR-ASPP, and U-Net models, respectively. Notably, the U-Net model excels in recognizing the contours of imperfect triangular flakes. Scale bar: 10 μm .

specialized equipment, and are time consuming. It is therefore desirable to develop alternative structural characterization techniques that are cost-effective, fast, and easily implemented without compromising accuracy and reliability.

Although the thickness of two-dimensional (2D) materials is normally confirmed by atomic force microscopy (AFM), Raman, etc.,³² optical contrast is frequently adopted by experienced researchers due to its speed and simplicity.³³ Recent developments in artificial intelligence (AI) have led to

the adoption of new techniques for processing microscopy image data sets of layer thicknesses, edges, dimensions, etc.^{34–36} For example, an autonomous robotic search and stacking of graphene flakes was proposed to detect up to 400 monolayer graphene samples in 1 h.^{34,35} Unsupervised Machine Learning (ML) and Deep Learning (DL) techniques have been used to classify 2D materials into different categories.^{37–40} In combination with an automatic optical microscope stage, the desired 2D materials can be searched

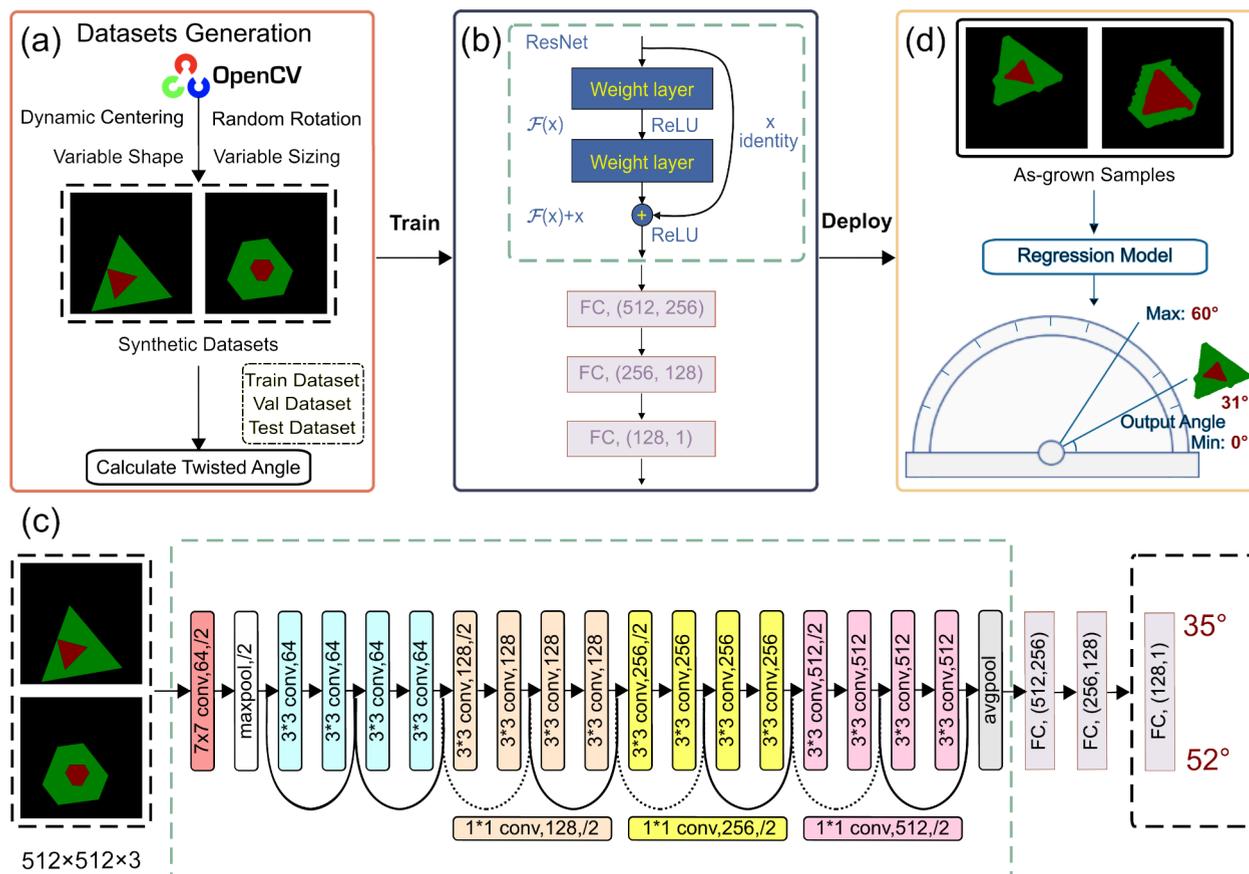


Figure 3. Deep learning approach for recognizing twist angles in atomically thin bilayer flakes. (a) Synthetic data sets illustrating varying twist angles in uniformly colored MoS_2 flakes postsegmentation. (b) ResNet CNN model training using the linear regression approach on the data sets from (a). (c) The detailed structure of ResNet with input training synthetic data sets and output twist angles of the data sets. (d) Prediction of twist angles for actual as-grown MoS_2 bilayer samples using the trained CNN model.

automatically.³⁹ However, to the best of our knowledge, no automatic procedure for determining the twist angle in bilayer atomically thin materials (e.g., TMDs and graphene) has been described up to now. Presumably, the DL model may lack sufficient accuracy due to the absence of adequate experimental data for training.

In this work, a systematic methodology is reported that demonstrates the potential of DL and image processing tools for determining the twist angles in CVD-grown bilayer TMDs and graphene. Specifically, we trained four different DL algorithms to identify the thickness of the CVD-grown flakes. The twist angle in individual bilayer TMD flakes can be estimated by means of image processing tools, such as implemented in OpenCV.⁴¹ This procedure, illustrated below, is nevertheless slow and not effective for large-scale sample analysis. To circumvent this problem, we developed a second DL model to characterize the twist angle of individual flakes in an efficient way. All codes and data sets are open access and freely available, provided with user-friendly instructions. Our work aims to provide new tools designed to facilitate and make more effective structural characterizations of CVD-grown twisted TMD samples, with extended applicability to CVD-grown graphene and hexagonal boron nitride (h-BN) and other CVD-grown 2D materials.

In our study, optical micrographs of CVD-grown bilayer atomically thin materials are captured as shown in Figure 1 (for MoS_2) and supplementary for graphene (Figure S15).¹² These

images are then processed and utilized to train a convolutional neural network (CNN) for the identification of flake thickness. Subsequently, a different CNN model, developed using a synthetic data set, is used to predict the twist angles of the flakes, with these predictions displayed on the corresponding images. The workflow diagram of the process to identify the twisted bilayer of TMDs is visually shown in Supporting Information Figure S1.

We utilized deep learning techniques to determine the thickness of atomically thin flakes, harnessing a supervised neural network trained on manually labeled images. These atomically thin flakes were initially distinguished by optical contrast and subsequently verified through AFM and Raman spectroscopy,^{13,14} as detailed in Supporting Information Section 1.1.

Figure 2 showcases the adeptness of our segmentation DL models in classifying flakes into monolayer (1L), bilayers (2L), and thicker layers (TL) with remarkable precision, as further elaborated in Figure 2 and Figure S7. Figure 2(a) shows an original unprocessed microscopy image. Figures 2(b–e) illustrates the manual labeling of background, monolayer, bilayer, and thick layers.

In our evaluation, four DL models were rigorously tested: DeepLabV3,⁴² Fully Convolutional Network (FCN),⁴³ MobileNetV3 (LR-ASPP),⁴⁴ and U-Net,⁴⁵ each employing distinct segmentation strategies and architectures. For instance, DeepLabV3 integrates a backbone architecture for feature

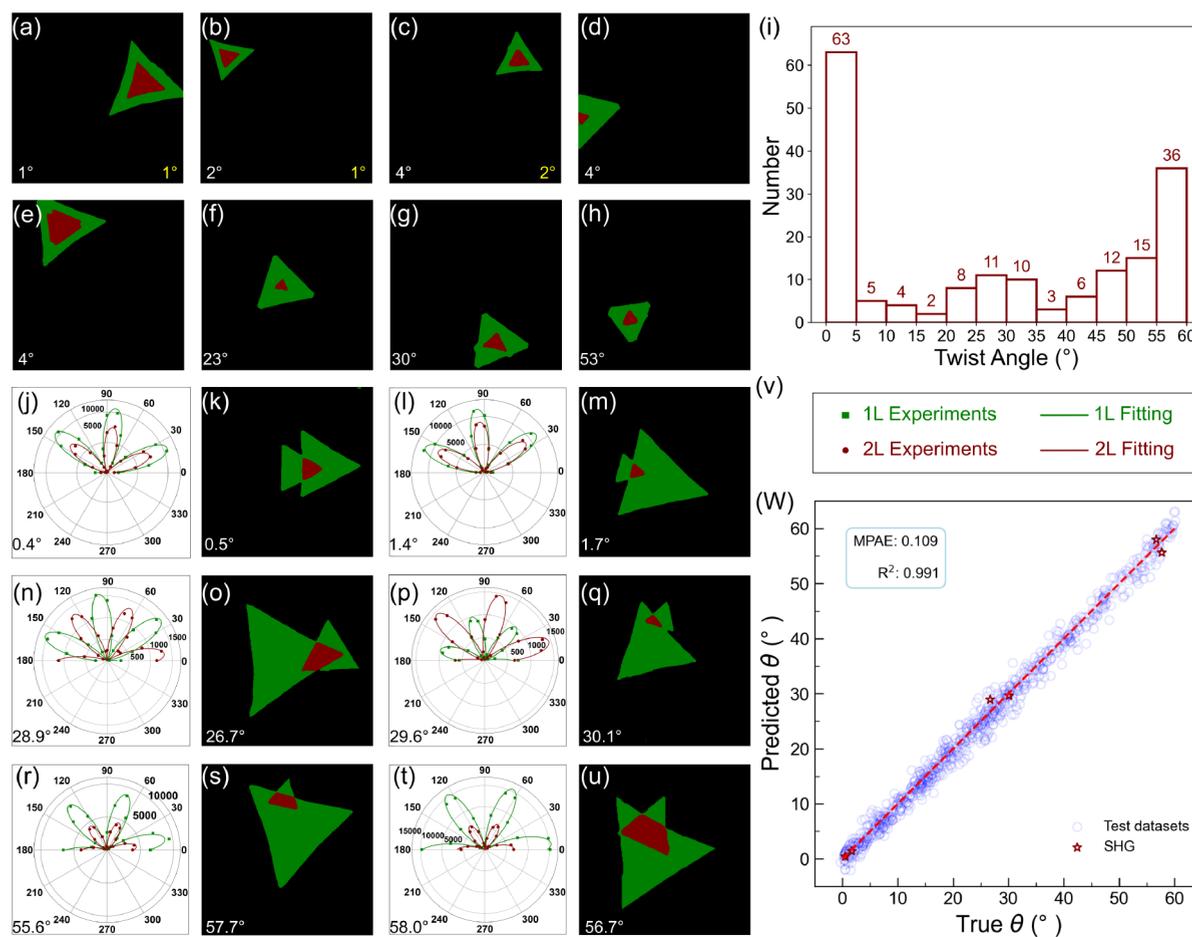


Figure 4. Performance evaluation of the twisted bilayer MoS₂ identification by the CNN model. (a)–(h) Predicted angles for different flakes, showcasing the efficacy of the CNN-based model (indicated in the bottom-left corner of each subfigure) in comparison to angles obtained via OpenCV (displayed in the bottom-right corner). It is noteworthy that in numerous instances OpenCV was unable to identify the angles, resulting in absent data in the bottom-right corner of some subfigures. (i) Histogram of the quantity of bilayer MoS₂ at various twist angles. (j)–(u) Second harmonic generation (SHG) and thickness identification for the corresponding samples with (v) serving as the legend. (w) Comparison between true and predicted twist angles θ using the artificially generated test data sets and SHG data set in (j)–(u), respectively.

extraction with advanced techniques, such as dilated convolutions and spatial pyramid pooling. The FCN model, tailored for semantic segmentation, ensures that the output size matches the input. Meanwhile, LR-ASPP, a variant of DeepLabV3's ASPP, aims for efficiency in mobile and edge computing. Notably, U-Net, recognized for its effectiveness in biomedical image segmentation, features a distinctive U-shaped architecture. Our implementation utilized Python 3.8 and Pytorch 1.10,⁴⁶ with U-Net emerging as the most adept in capturing the nuances of shape, particularly evident in cases of distorted triangular overlays. Compared to the original micrographs, U-Net's segmentation reveals enhanced detail in both monolayer and bilayer configurations (Figure 2(n)), affirming its selection as the primary model for our study. For a comprehensive understanding of the labeling process and additional model comparisons, readers can refer to the Supporting Information (Section 2.1).

Once the pixels in an experimental image have been classified, either by a human or by any of the trained DL models described in Sec. I of SI, and the resulting flakes color-coded according to their thickness, it is possible to determine the twist angle formed between an underlying single-layer and an overlying second layer using appropriate image analysis software, such as OpenCV.⁴¹ It contains useful functionalities

to e.g. determine contour lines enclosing a particular flake and measure the corresponding enclosed area, fit an approximate polygonal shape, or find the minimum desired polygonal shape that encloses a given set of pixels in the image. Given that TMDs typically exhibit a triangular shape, we employ triangles as the chosen polygonal shape, leading to an estimation of the corner positions. By carrying out this process for each of the layers in a flake, it is possible to estimate the twist angles between each pair of layers from their corner positions by simple trigonometric calculation.

Nevertheless, the use of image analysis software as described above has several disadvantages. First, it is not easily automated, requiring human intervention and thus resulting in a slow and tedious process that cannot be applied efficiently on a large scale. A better alternative is to train a DL model to directly predict the twist angles from the appropriately cropped experimental image of a twisted bilayer [see Figure 1(d,e)]. Training such a model would require a large database of preprocessed experimental images, for which the twist angles had been previously determined. However, there is a more practical and expedient procedure, which consists of employing a synthetically generated image database. As can be seen in Figure 1 and Figure 2, experimental samples of MoS₂ bilayer flakes typically consist of two roughly triangular shapes, with

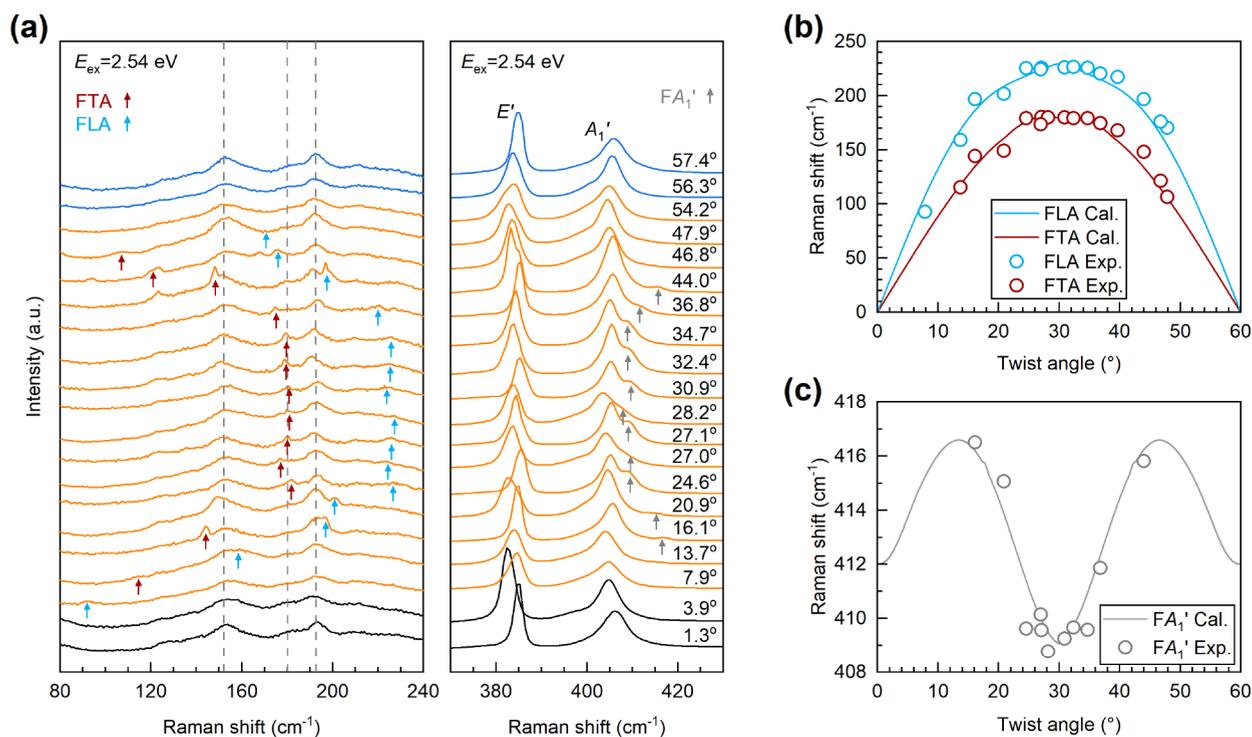


Figure 5. Moiré phonons in twisted CVD-grown bilayer MoS₂. (a) Raman spectra of CVD-grown bilayer MoS₂ with the excitation energy (E_{ex}) of 2.54 eV. Peaks assigned to Moiré phonons modes²⁸ including FTA, FLA, and FA₁' modes are marked by red, blue, and gray arrows, respectively. The dashed lines correspond to the peak position of second-order Raman modes, and the E' and A_1' modes are also marked out. (b, c) Comparison between the calculated (Cal, lines) and experimental (Exp, circles) frequencies of twist-angle-dependent Moiré phonons including (b) FTA, FLA, and (c) FA₁' phonons.

the rotation angles in the range of 0 to 60°. It is trivial to generate large numbers of synthetic images of rotated pairs of superimposed triangles. This adds the advantage that such images can be made to sample homogeneously all possible twist angles, while training on labeled experimental images would result in a bias toward the experimentally observed twist angles. It is even possible to generate synthetic images that mimic more faithfully the experimental ones, by, e.g., cropping corners of the triangles or introducing random noise to their edges.

In the process of generating the data sets for training the second neural network, a sequence of images is created, including double-layered polygons that morph sequentially from hexagons to truncated triangles and finally into triangles (as illustrated in Figure 3(a)). Formation of the outer polygon begins with the careful selection of random variables such as central coordinates, side lengths, and rotational angles, promoting visual diversity. Once the outer polygon is defined, an inner polygon is positioned within it. The inner polygon's location, rotation, and side length are determined through random selection, while ensuring its side length is equal to or less than the outer polygon's, preserving their hierarchical nesting and maintaining distinct spatial separation between them.

The angle formed by a vertex of each polygon and its respective center is calculated, providing a measure to assess the angular difference between them. This angular variation is then incorporated into the filename of the stored image, serving as a perceptible geometric attribute label. Consequently, the generated data sets, enriched with clear geometric annotations and offering a wide variety of forms, become a

valuable resource for training data sets in the subsequent recognition of twist angles in CVD-grown samples.

After that, a *Residual Network* (ResNet)⁴⁷ Convolutional Neural Network (see Figure 3) is trained to predict the twist angle by regression to a synthetically generated database of more than 10,000 images. The input images are RGB images with a resolution of 512 × 512 pixels obtained from Figure 3(a). In model design, the choice between using a single-layer or multilayer approach for regression tasks hinges on the complexity of the data relationship. A direct 512 to 1 dimension reduction in the network might be suitable for a simpler, linear problem, while a more gradual decrease, such as 512–256–128–1, tends to perform better for capturing complex, nonlinear relationships in the data (as shown in Figure 3(b) and (c)). In the latter approach, additional layers aid the model in learning nuanced data patterns, offering a potential boost in the predictive accuracy for intricate problems. Upon being input into the ResNet network, the final angles are obtained, as illustrated in Figure 3(b) and (c). After training the CNN regression model, the real as-grown bilayer MoS₂ micrographs after the first deep learning model are used as input, and the twist angle of the bilayer MoS₂ is obtained through this second neural network. The capabilities of the model are illustrated in Figure 4. The identification of twist bilayer MoS₂ using OpenCV is also demonstrated in Figure S8 and shown in Figure 4 as a comparison for the DL methodology. It can be concluded that the CNN regression model we used here can identify the twist angle in bilayer MoS₂ while being tolerant of shape irregularities. For bilayer graphene synthesized through CVD, it is evident that different edges can correspond to varying twist angles in the secondary layer¹² (Figure S15). Demonstrably, our methodology

proficiently enables the identification of such twist angles within hexagonally shaped, CVD-grown bilayer graphene, offering insightful exploration into its structural intricacies, as depicted in Figure S15.

To verify the identification of the twist angles of CVD-grown bilayer MoS₂, SHG measurements are performed on these samples with various twist angles as shown in Figure 4(j)–(v). The true and predicted twist angle θ using the artificially generated test data sets and SHG data set is summarized in Figure 4(w), showing the reasonable accuracy of our model.

One possible application of our methodology could be to select more samples for probing their optical property correlations. The strain, defects, and doping of the inhomogeneity of the individual bilayer CVD-grown flakes could influence the Raman spectra, making the interpretation of Raman signal challenging.^{48–54} Figure 5(a) shows the Raman spectra of the CVD-grown bilayer MoS₂ samples with twist angles ranging from 0 to 60°. The three vertical dashed lines correspond to the peak positions of the second-order Raman modes $E'(M)^{LO_2}$ -LA(M) (~ 150 cm⁻¹), $A_1'(M)$ -LA(M) (~ 178 cm⁻¹), and TA(K) (~ 190 cm⁻¹).⁵⁵ In addition, several branches of Moiré phonons are observed in the Raman spectra. Moiré phonons refer to zone-center phonons in twisted bilayer MoS₂, which are folded from the off-center phonons in monolayer MoS₂ due to the periodic Moiré potential and thus exhibit a shift in peak frequency with the change of twist angle.²⁸ The arrows indicate the assigned Moiré phonon modes, including folded TA (FTA), folded LA (FLA), and folded A_1' (FA_1') modes, which exhibit high-frequency sensitivity to the twist angle, similar to the previous results.²⁸ Figure 5(b),(c) summarizes the experimental and calculated frequencies of the three Moiré phonon modes for comparison. The experimental peak positions of Moiré phonons agree well with the theoretical ones²⁸ based on the twist angle of twisted bilayer MoS₂ and the phonon dispersion of monolayer MoS₂. Note that the frequencies of the FA_1' mode on the CVD-grown bilayer MoS₂ diverging relatively from the theoretical curve in the range of 24.6° to 34.7° may arise for different reasons, where strain is very likely as E' and A_1' modes also show behaviors of strain-induced shifts.⁵¹ We did not discuss the results of the twist angles at 0–5° and 55–60° in detail because the case is very complicated due to the phonon renormalization induced by lattice relaxation.⁸

Another possible application of our methodology could be the optimization of the large-scale growth for bilayer atomic materials (as shown in Figure S17). In addition, the exfoliated flakes with regular shape could also be the next possible applications of our model (as shown in Figure S18).^{56,57} This thorough workflow acts as a clear guide, detailing the methodology utilized in the study for the precise identification and analysis of twisted bilayers in TMDs grown via CVD. This method can also be adapted to include graphene (as shown in Figure S15) and hBN, as well as their heterostructures.

In conclusion, we have demonstrated a robust and efficient methodology for the identification and analysis of twisted bilayers in TMDs by using deep learning and OpenCV techniques. Our approach, which combines optical micrographs, deep learning models, and OpenCV, provides accurate and comprehensive predictions of the thickness properties and twist angles of bilayer TMDs. The comparison of various deep learning models revealed that the U-Net model exhibits superior performance in terms of global accuracy, mean intersection over union, and processing speed.

Our methodology can be extended to other two-dimensional materials grown by CVD, including both homostructures and heterostructures, highlighting its versatility and broad applicability in the field of 2D materials analysis. Our data sets and codes are made freely available as a service to the community. We hope that, by facilitating and automating the structural analysis of TMDs, this work will contribute to further advancements in the field of TMDs and 2D materials in general and thus also the rapid growing Autonomous lab using AI.⁵⁸

METHODS

A description of the data pipeline, data preparation, deep learning training of the semantic segmentation model, and twist angle identification by OpenCV can be found in the SI.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.nanolett.3c04815>.

Data set preparation, the detailed results of the semantic segmentation, the synthetic data sets, the verification of the twisted bilayer graphene, and the Raman spectra of CVD-grown twisted bilayer MoS₂. All scripts used in this study are available on GitHub⁵⁹ (PDF)

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[†]H.Y., R.H., H.W., and X.H. contributed equally to this work. Y.X. supervised the project; Y.X. and E.R.H. conceived the idea. H.Y. performed the coding with the help of Y.X. and R.H.; H.Y. and Y.Z.X. prepared the data sets with assistance from H.C. and M.G.; H.Y., X.H., K.H., and Y.Z. prepared the figures with the help of Y.X., R.H., and E.R.H.; H.W., Y.Z., and X.Z. measured the Raman spectra and SHG and analyzed the data under the supervision of P.T.; Y.X., R.H., and E.R.H. wrote the manuscript.

Notes

The authors declare no competing financial interest.

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