Abstract: Defects introduced during the growth process greatly affect the device performance of two-dimensional (2D) materials. Here we demonstrate the applicability of employing machine-learning-based analysis to distinguish the monolayer continuous film and defect areas of molybdenum disulfide (MoS$_2$) using position-dependent information extracted from its Raman spectra. The random forest method can analyze multiple Raman features to identify samples, making up for the problem of not being able to effectively identify by using just one certain variable with high recognition accuracy. Even some dispersed nucleation site defects can be predicted, which would commonly be ignored under an optical microscope because of the lower optical contrast. The successful application for classification and analysis highlights the potential for implementing machine learning to tap the depth of classical methods in 2D materials research.

Keywords: 2D materials; machine learning; random forest algorithm; Raman spectrum

1. Introduction

Transition-metal dichalcogenides (TMDCs) are a class of layered materials analogous to graphene, which have aroused immense interest in the last decade as a promising platform for electronic and optoelectronic applications in the post-Moore era [1,2]. The adjacent layers in these materials are held together by weak van der Waals forces but strong covalent bonding forces inside the layer, making it possible to cleave or synthesize to the limit of a monolayer. Even though TMDCs have been studied for decades in their bulk form, the properties of monolayers and few-layers with ultrathin thickness differ dramatically from the macroscopic material characteristics, being the biggest reason for renewed interest in this material class. Compared to their bulk counterpart, monolayers of many TMDCs (such as MoS$_2$, WS$_2$, WSe$_2$ and MoSe$_2$) are especially exciting since they are direct band gap semiconductors, making them ideal candidates to replace silicon for device applications, such as light-emitting diodes, photodetectors and photodiodes [3]. The chemical vapor deposition (CVD) method provides a convenient and controllable way to grow high-quality and large area 2D materials at a reasonable cost, which has been earmarked as the process that will deliver scalable production [4,5]. Nowadays, continuous films of 2D materials compatible with current silicon-based microfabrication...
processes are greatly needed for industrial electronic and optoelectronics applications [6–8]. However, the present uniformity of as-grown monolayer films is still inadequate, such as structural differences and poor controllable layer distribution, etc. It is a remarkable fact that the introduced cracks that appear in synthesis processes can adversely affect the device performance and directly increase the chip failure risk [9]. These factors greatly limit the further applications of these materials. As a consequence, it is essential to check the uniformity of the obtained 2D materials. It is necessary to locate and distinguish the monolayer continuous films and the random crack areas, as well as bilayer areas prone to be introduced in the growth process, in order to better understand and improve the growth process [10].

To identify these areas with thickness differences, the most typical methods are atomic force microscopy (AFM) [11,12], optical microscopy (OM) [13–15], differential reflectance spectra [16,17] and Raman spectra [18,19]. AFM is a versatile method used to measure the thickness of 2D materials; however, the materials are easily destroyed due to the sliding of the tip on the sample surface when working on contact mode, while the tapping mode takes a relatively long time to measure even a small area [20,21]. Time-domain terahertz spectroscopy is an emerging technique for imaging 2D materials, but the comparatively large spot size hinders the identification of laterally small flakes [22]. In such a situation, an accurate, versatile and nondestructive method is highly desirable not only in fundamental research but also for practical applications. Recently, machine learning approaches have attracted considerable attention for solving various problems in materials science and optical engineering. OM using red–green–blue (RGB)-based optical contrast combined with machine learning has shown emerging potential in identifying and determining the thicknesses of 2D materials. Lin et al. first used the support vector machine (SVM) method to learn the contrast information of optical images to determine the layer numbers of graphene and MoS2 [23]. Later, clustering analysis [24] and the convolutional neural network (CNN) [25–27] also joined in this stage play, expanding the identification types and application scenarios of 2D materials. However, because the accuracy of using optical images to determine the layer number based on machine learning is not too high, the previous researchers mainly regarded it as an initial screening to reduce manual work [25]. The attempt to use photoluminescence (PL) imaging and computer vision techniques to analyze monolayer TMDCs also enlightened us to combine the research methods of intrinsic material properties with new technologies to obtain information about molecular structure and layer number simultaneously [28]. Nowadays, based on the relation between the Raman frequency shifts of the $E_{2g}^1$ and $A_{1g}$ Peaks (TMDCs) and those of the 2D and G peaks (graphene), Raman spectroscopic mapping has been widely used to identify the thickness and confirm the uniformity of 2D materials [29–32]. As a high-resolution imaging technique, it does give us more information to study the nature of matter. However, it is difficult to identify these defects in the selected area and at the same time simply using one-side Raman frequency shifts. Hence, we consider solving this problem by introducing a machine learning approach to use more Raman features for the simultaneous identification.

To our knowledge, we are the first to present a recognition method to distinguish the monolayer continuous film and random defect areas of 2D semiconductors using the machine learning method with Raman signals. Compared to other unsupervised techniques, the supervised machine learning represented by random forest can not only reduce the computational expense and time but also achieves high accuracy. In the introduction process for the random forest algorithm, we use several Raman characteristics extracted from spatial mapping results as the input variables and the sample thickness type as the output variable for generating the decision trees. The successful application of a machine learning approach to the classification and analysis of the CVD-prepared MoS2 highlights the potential of this method for 2D materials research.
2. Materials and Methods

2.1. Synthesis of Monolayer MoS$_2$ Continuous Film

The monolayer MoS$_2$ continuous film mentioned in this paper was synthesized via the CVD method similar to our previous work [33]. The film was grown using MoO$_3$ powders (99.97% Sigma Aldrich, St. Louis, MO, USA) as the molybdenum source and sulfur powders (99.98%, Sigma-Aldrich, St. Louis, MO, USA). First, MoO$_3$ and sulfur powders were loaded into two separate Al$_2$O$_3$ crucibles, which were located at the center and the upstream of a dual-temperature-zone tube furnace with a diameter of 100 mm. A piece of Si substrate with thermally grown 300-nm-thick SiO$_2$ was loaded at the downstream. Before the film growing, the quartz tube was evacuated to 4000 Pa at room temperature. Then, the temperature of the MoO$_3$ was increased to 670 °C and the temperature of the sulfur was increased to 190 °C with 50 sccm of argon gas, and maintained for 10 min. Finally, the furnace was naturally cooled down to room temperature.

2.2. Characterization and Measurements

We carried out PL, AFM and Raman measurements. The PL signals were collected by a confocal microscopy setup (LabRAM HR Evolution, Horiba Co., Kyoto, Japan) with a 532 nm continuous-wave (CW) laser of a frequency-doubled Nd:YAG laser. The height profiles were measured using AFM taken by an FM-Nanoview6800 (FSM-Precision Co., Suzhou, China) in tapping mode. Raman spectra were obtained using the same confocal microscopy system equipped with a programmable scanning stage with a 532 nm CW laser (Changchun New Industries Optoelectronics Tech. Co., Changchun, China) as the excitation source. We chose the 100× objective lens (MPLFLN 100×, NA = 0.9) and set the laser power below 1 mW to avoid local heating and undesirable oxidation of the sample. The scanning step was set as 0.2 µm and the integration time was also carefully optimized to obtain an adequate spectrum resolution and a satisfactory signal-to-noise ratio, while maintaining acceptable data acquisition duration and avoiding drift.

3. Results and Discussion

As shown in Figure 1a, the MoS$_2$ monolayer continuous film has a relatively smooth surface. The thickness of the monolayer region is around 0.88 nm as confirmed by AFM, which corresponds to an interlayer S–Mo–S layer [19]. The heights of the undertint line and dark triangle areas were found to be crack and bilayer defects, respectively. The PL spectra in Figure 1b show two peaks at 670 nm and 620 nm corresponding to A (1.9 eV) and B (2.0 eV) direct excitonic transitions with the energy split from the valence-band spin–orbital coupling, respectively. The bilayer shows a decline in PL intensity compared with the monolayer [34]. As shown in Figure 1c, two Raman-active modes, E$_{2g}^1$ and A$_{1g}$, exhibit significant differences, and the Raman frequency difference of the monolayer sample is ~17.9 cm$^{-1}$, while that of the bilayer sample is ~21.1 cm$^{-1}$. This is consistent with previous results for mechanical exfoliation (ME)-prepared and CVD-prepared samples, implying this difference is universal between samples obtained from different preparation methods [35–37]. The cracks nucleated at sulfur vacancies propagate along the energy-favored zigzag directions upon the relatively fast temperature-drop-induced thermal strain, which results in an orientation-specific fracture behavior [38]. In addition, the appearance of an E$_{2g}^1$ mode proves that all the monolayer and bilayer samples are 2H-MoS$_2$ [39]. In the completely exposed internal area of the crack, the Raman signal of the Si substrate is mainly collected. The peak around 520 cm$^{-1}$ is attributed to the Si mode. Similar to the case of multilayer graphene, the Raman signals from the Si substrate can be absorbed by the MoS$_2$ flakes, which makes the intensity of the Si mode monotonously decrease from the bare substrate to the monolayer and bilayer MoS$_2$ flakes [40,41]. In Figure 1c, the intensity of the Si mode is normalized to display the spectral information more intuitively like the previous work [42].
AFM data to ensure effective classification. Nowadays, spectroscopy is the backbone of research in such diverse fields, ranging from physics to engineering, chemistry and biology [45]. Compared to relying on AFM to determine thickness, the more convenient way is to use the potentiality of classical spectroscopy research methods to obtain information about molecular structure, chemical composition and even layer number simultaneously.

The pixels of the optical image in Figure 1a were reduced to be consistent with the collection points of the spectra by bicubic sharper process. Then, the reserved pixels could be clustered using the k-means algorithm [43], which can partition all pixels into three clusters with each cluster having a mean value, and pixels in one cluster are closest to the corresponding mean value among the cluster means [44]. Combined with the AFM heights, three regions with crack, monolayer and triangle bilayers can be identified as shown in Figure 2a. When using k-means clustering to classify different regions of the optical image, in addition to ensuring sufficient optical contrast, the clustering also depends on the AFM data to ensure effective classification. Nowadays, spectroscopy is the backbone of research in such diverse fields, ranging from physics to engineering, chemistry and biology [45]. Compared to relying on AFM to determine thickness, the more convenient way is to use the potentiality of classical spectroscopy research methods to obtain information about molecular structure, chemical composition and even layer number simultaneously.

In the research methods for 2D material properties, Raman spectroscopy is the most commonly used technique in many fields, since it allows the essential characteristics of matter that are invisible by standard OM and AFM to be viewed [46]. Ultralow-frequency Raman spectroscopy has been used to reliably determine layer numbers of TMDC flakes, but this technique requires expensive adapters and nonstandard equipment setup [47]. Therefore, it is of vital importance to look for a suitable technique using the standard Raman system. Generally, high-frequency Raman peaks of lattice vibrations (i.e., phonons) in TMDCs exhibit several prominent features, including frequency, intensity and full width at half maximum, which contain useful information in characterizing the physical and chemical properties of the materials. Figure 2b,c show the Raman spectral mapping of the $E_{2g}^1$ and the $A_{1g}$ peaks. From these two mappings, we can find the differences between the monolayer and bilayer areas. The frequency of the $E_{2g}^1$ peak decreases, while that of the $A_{1g}$ peak increases with increasing layer number. The observed blueshift of the $A_{1g}$ peak originates from the constraint of
atom vibration by the interlayer van der Waals force in MoS$_2$, whereas the stacking-induced structure changes or long-range Coulomb interlayer interactions account for the redshift of the E$_{2g}^1$ peak [18]. Therefore, as shown in Figure 2d, the Raman frequency difference between the E$_{2g}^1$ and A$_{1g}$ peaks can be used as a fingerprint feature to identify the monolayer and bilayer MoS$_2$ regions of the flakes [19]. However, the crack area cannot be identified straightforwardly by only using the frequency shifts of the E$_{2g}^1$ or A$_{1g}$ peaks.

![Figure 2](image)

**Figure 2.** (a) K-means algorithm clustered image for the selected sample with monolayer (cyan), bilayer (dark cyan), and crack (light cyan) regions. Raman spectral mapping of the (b) Pos(E$_{2g}^1$) and the (c) Pos(A$_{1g}$). (d) Raman spectral mapping of the frequency difference between the Pos(E$_{2g}^1$) and the Pos(A$_{1g}$).

The great discrepancies in Raman intensity from crack to monolayer inspired us to consider more features extracted from Raman spectra to realize the simultaneous identification of these three regions. It is worth noting that when the detection position controlled by the scanning stage moves to the boundary of two regions, i.e., the junction of the crack and monolayer, it will inevitably collect signals both from the crack and monolayer MoS$_2$ at the same time, which makes it difficult to classify different areas based on Raman intensity by setting the threshold manually. We chose the average value of the E$_{2g}^1$ peak intensity of a large monolayer MoS$_2$ area as the reference intensity, and all thresholds (a fixed percentage) were set based on this value. The Raman spectral intensity mappings of the E$_{2g}^1$ peak are shown in Figure 3a–c and the thresholds are 68, 70 and 72%, respectively. It is not difficult to see that when the threshold changed slightly, the predicted information of the crack area and the monolayer area also changed. This means that an effective evaluation tool is lacked for judging the rationality of the artificially set thresholds. In addition, the monolayer sample generally has two Raman peaks, E$_{2g}^1$ and A$_{1g}$, under the excitation of green or blue lasers with proper power. When using the same intensity threshold of the A$_{1g}$ peak to make judgments, there are also some differences in the conclusions drawn compared to the E$_{2g}^1$ peak, as shown in Figure 3d–f, which further increase the difficulty of manual processing. The rising machine learning approaches can help to successfully extract and analyze the multiple Raman characteristics among many samples to address this problem.
we define $Z$ variables, which are acquired from the k-means algorithm results. The data that are not used in the training process for each decision tree can be used to estimate the skill and variance by combining diverse trees, hence yielding a better model overall. Moreover, the out-of-bag decision tree typically exhibits high variance and tends to overfit, random forest can achieve reduced gain, the greater the “purity improvement” obtained using this feature variable. Since one individual tree has multiple nodes, and the threshold values of the variables at each node are computationally

Random forest is an ensemble learning algorithm that uses a group of decision trees built by the subtraining sets as weak individual learners of randomly sampled training data. Each decision tree has multiple nodes, and the threshold values of the variables at each node are computationally determined to yield the largest information gain. Generally speaking, the greater the information gain, the greater the “purity improvement” obtained using this feature variable. Since one individual decision tree typically exhibits high variance and tends to overfit, random forest can achieve reduced variance by combining diverse trees, hence yielding a better model overall. Moreover, the out-of-bag data that are not used in the training process for each decision tree can be used to estimate the skill and effectiveness of the trained random forest model. The whole algorithm is conducted in Python using the open-source pandas and scikit-learn machine learning libraries [52].
Figure 4. Basic architecture of the learning procedure in the random forest method. Each small square represents a spatial measurement point carrying Raman characteristic information. The subtraining sets from 1 to n are acquired by a bootstrap sampling process, and then decision trees based on these subtraining sets can be built. The out-of-bag data of each tree can be used to estimate the effectiveness of the trained random forest model.

Figure 5a shows the prediction procedure for the random forest method. After generating the whole forest, new samples from different positions carrying the input Raman characteristic information will be judged through the formed decision trees one by one. Since the training sets are different from each other, they may give different judgments for one sample by different trees. The final output result is generated by the democratic majority voting which can acquire a dramatically reduced variance. After the optimal parameter combination is fixed, it can immediately give a classification result for the new input data. The accurate measurement of Raman spectra and the use of a sufficient number of training sets enable the constructed random forest to obtain a relatively high accuracy rate. As shown in Figure 5b–e, we choose different sample areas on several pieces of substrate to test our model, and the classification results via the random forest method are successful in distinguishing the monolayer, crack and bilayer areas. In the central areas of these three regions, random forest can provide reliable classification results via the random forest method are successful in distinguishing the monolayer, crack and bilayer areas. Benefitting from the high-speed computing power of the computer, the random forest algorithm can be easily and continuously strengthened by increasing the amount of data. As the number of learning data of insufficient sample types are increased, the recognition accuracy is expected to be further improved [48].

Furthermore, some dispersed dots in Figure 5b are easily predicted, which would be commonly ignored under optical microscope because of the lower optical contrast. As reported in the previous work [53], several stages were observed during the MoS$_2$ atomic layer growth. Initially, some small domains were nucleated at random locations on the substrate. Then, the nucleation sites continued to grow and formed boundaries when two or more domains met, resulting in partially continuous films. The as-grown films are predominantly monolayer, with small areas consisting of two or more layers at the preferred nucleation sites [42,53], which explains the manifested bilayer Raman characteristics of these areas located on the monolayer continuous film. This suggests that prediction via the random forest possesses the application potential to view subtle differences through the material’s basic features. By using a shorter wavelength laser and a larger numerical aperture objective, it is expected that...
the spatial resolution will be further improved, which makes it possible to identify defects that are
difficult to find and/or determine only by optical images. A smaller laser spot is ideal for analyzing
the microscopic characteristics of the sample. In theory, the higher the spatial resolution, the more
precise the micro-area spectral information that can be obtained. The random forest algorithm is not a
conservative and fixed tool and it can be flexibly adjusted according to our actual needs to adapt to
different scenarios. During the Raman spectra acquisition process, it is better to ensure the stability of
the experimental conditions, so as to ensure the accuracy to the greatest extent.

As shown in Figure 6a, when using only the Raman frequency difference between the $E_{2g}$
and $A_{1g}$ peaks, the AUC is 0.9891 for bilayer/others but is only 0.7543 for crack/others, which means that
the Raman frequency difference can recognize bilayer samples extremely well but is not good for the
cracks. Uniformly, the PR curves in Figure 6b also prove this point, in which the average precision (AP)
values are 0.9895 and 0.7167 for bilayer/others and crack/others, respectively. However, when taking all
the input variables ($\alpha, \beta, \gamma, \delta$ and $\epsilon$) into consideration, the numerical values of AUC and AP are 0.9852
and 0.9867 for crack, and 0.9902 and 0.9914 for bilayer, which means that this technique based on the
random forest method can successfully characterize and confirm the monolayer, crack and bilayer
areas at the same time.
In this study, we demonstrated an effective method based on a random forest algorithm to classify monolayer MoS$_2$ continuous film, random crack and bilayer areas from the variables extracted from Raman spectra. The random forest method was used to analyze multiple Raman features to identify samples, solving the problem of ineffective identification of samples by just one specific variable. It can
successfully determine the importance of a certain characteristic variable and some dispersed defect dots can also be predicted, which would commonly be ignored under an optical microscope. By taking peak intensity and frequency information into consideration at the same time, a high accuracy rate is obtained. The method developed in this work can also be used for other 2D materials and can provide a valuable reference for material characterization in several fields.

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