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Deep Learning Assisted Raman Spectroscopy for Rapid Identification of 2D Materials

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Abstract

Two-dimensional (2D) materials are gaining significant attention for their unique properties and potential applications. Raman spectroscopy is a rapid, non-destructive tool for characterizing these materials, but traditional analysis methods are often time-consuming and subjective. ***In this study, we leverage deep learning, including classificatory and generative models, to enhance Raman spectra analysis for 2D materials.*** To address the challenges of limited and unevenly distributed data, ***we use Denoising Diffusion Probabilistic Models (DDPM) for data augmentation and develop a four-layer Convolutional Neural Network (CNN) for classification. Our CNN model achieves an accuracy of 98.8%, with the DDPM-CNN approach reaching 100% classification accuracy,*** demonstrating the method's effectiveness and reliability in automated material analysis. This work highlights the potential of deep learning-assisted Raman spectroscopy for precise and rapid 2D material characterization.

Introduction and objectives

This study addresses the challenges of traditional Raman spectroscopy analysis for 2D materials by integrating a Denoising Diffusion Probabilistic Model (DDPM) with a 1D CNN classifier. ***By using advanced data augmentation techniques,*** we expand the training dataset, significantly improving classification accuracy. Our approach not only enhances the identification of 2D materials but also streamlines the analysis process, reducing manual intervention and enabling efficient, automated characterization, particularly in complex and industrial applications.

Materials

The task of identifying different 2D materials is framed as a multi-class classification problem. We used a dataset of 594 Raman spectra representing seven distinct 2D materials and three stacked combinations: Black phosphorus (BP), Graphene, MoS_2, ReS_2, Tellurium (Te), WSe_2, WTe_2, BP–WSe_2 (S1), Te-ReS_2-WSe_2-Graphene (S2), and Te-WSe_2-WTe_2 (S3). The spectra vary due to differences in substrates, with detailed dataset composition shown in Table 1.

Materials	Quantity of spectra
BP	35
Graphene	209
MoS_2	8
ReS_2	15
Te	270
WSe_2	6
WTe_2	28
S_1	8
S_2	7
S_3	8
Total:	594

Table 1. Statistics of Raman spectral dataset of 2D materials studied in this work. S_1 to S_3 refer to various heterostructure stacks (see text for details).

Method

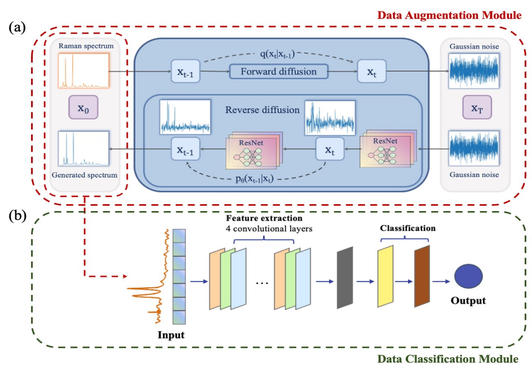

Figure 1. Illustration of the DDPM-based data augmentation for Raman Spectroscopy of 2D materials classification framework. (a) Data augmentation module based on DDPM. (b) Spectral classification module based on 1D CNN.

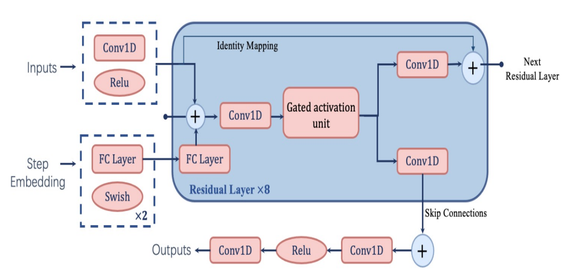

Figure 2. An illustration of the ResNets architecture. In this schematic, "FC" denotes the fully connected layer. "Relu" represents the rectified linear unit (Relu) activation function. The "gated activation unit" consists of the hyperbolic tangent (tanh) activation function and the sigmoid activation function, it can be denoted as $tanh(W_{f,k} * x) \odot \sigma(W_{g,k} * x)$, where W represents a convolutional filter, f and g represent the filter and gate, respectively, and k represents the layer index.

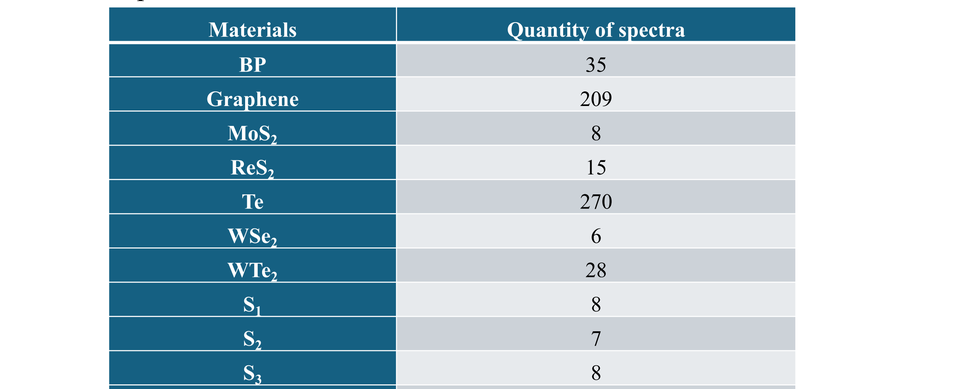

Figure 3. The architecture of the four-layer CNN for Raman spectroscopy classification.

Conclusion

This study successfully applies deep learning with DDPM-based data augmentation to improve the classification of 2D materials from Raman spectra, achieving up to 100% accuracy. While effective, the method relies on the initial dataset's quality and requires significant computational resources. Future research should explore expanding this approach to other spectroscopic techniques and integrating advanced methods like transfer learning to enhance material characterization further.

Results

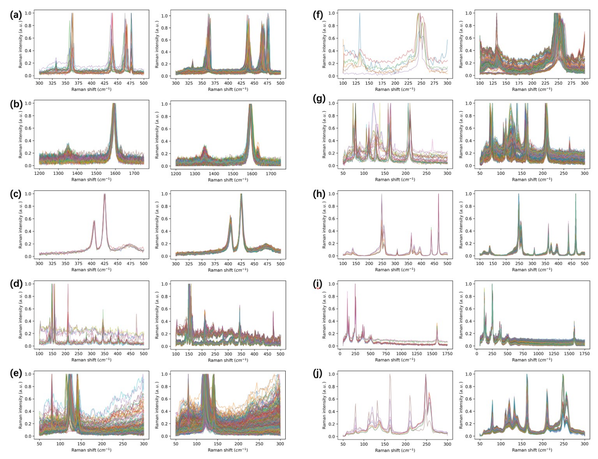

Figure 4. The Raman spectra of various 2D materials before and after data augmentation using DDPM: (a) BP, (b) Graphene, (c) MoS_2, (d) ReS_2, (e) Te, (f) WSe_2, (g) WTe_2, (h) BP–WSe_2 stack (S_1), (i) Te-ReS_2-WSe_2-Graphene stack (S_2), and (j) Te-WSe_2-WTe_2 stack (S_3). The left side represents the original Raman spectra dataset, while the right represents the augmented Raman spectra dataset.

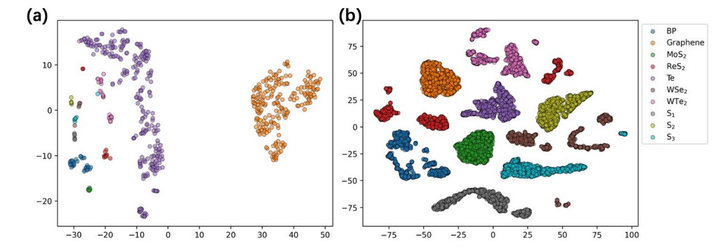

Figure 5. t-SNE plots for (a) the original and (b) the augmented dataset (including original spectral data) of different 2D materials. [S_1: BP–WSe_2 stack, S_2: Te-ReS_2-WSe_2-Graphene stack, S_3: Te-WSe_2-WTe_2 stack.]

Method	Accuracy	Precision	Recall
CNN	0.988	0.945	0.937
DDPM-CNN	1.000	1.000	1.000
ANN	0.946	0.658	0.646
DDPM-ANN	1.000	1.000	1.000
RF	0.906	0.566	0.574
DDPM-RF	1.000	1.000	1.000
SVM	0.966	0.829	0.786
DDPM-SVM	1.000	1.000	1.000
KNN	0.953	0.826	0.770
DDPM-KNN	0.988	0.989	0.988
LR	0.960	0.731	0.711
DDPM-LR	1.000	1.000	1.000

Table 2: The average performance of ten-fold cross-validation comparisons between the proposed methods (with DDPM) vs. baselines.

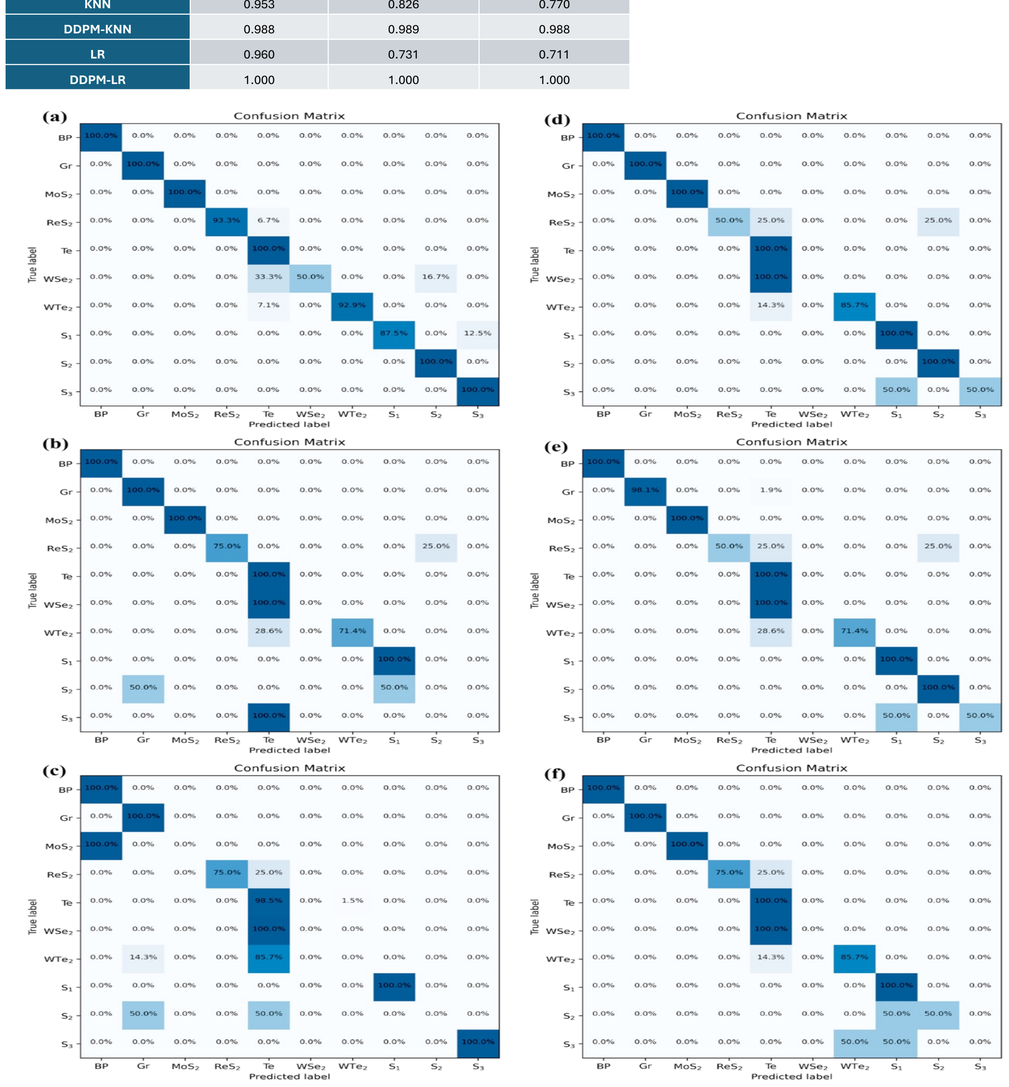

Figure 6. Confusion matrices depicting the average accuracy of ten-fold cross-validation in the classification of each category by different algorithms: (a) CNN, (b) ANN, (c) RF, (d) SVM, (e) KNN, and (f) LR. The diagonal elements represent the percentage of true positives, which is a key indicator of the algorithm's ability to correctly identify each category. The off-diagonal elements represent misclassification rates.

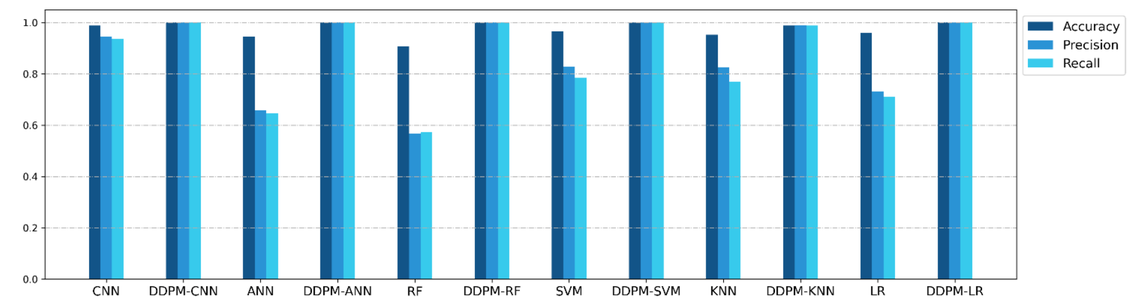

Figure 7: Bar chart of the average performance of ten-fold cross-validation between the proposed methods (with DDPM) vs. baselines.

Acknowledgment

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