

Deep-learning-assisted robotic assembly of van der Waals superlattices

Satoru Masubuchi¹, Eisuke Watanabe¹, Yuta Seo¹, Momoko Onodera¹, Shota Okazaki², Takao Sasagawa², Kenji Watanabe³, Takashi Taniguchi^{1,4}, and Tomoki Machida¹

¹ Institute of Industrial Science, University of Tokyo

Komaba 4-6-1, Meguro-ku, Tokyo 153-8505, Japan

Phone: +81-3-5452-6158 E-mail: msatoru@iis.u-tokyo.ac.jp

² Laboratory for Materials and Structures, Tokyo Institute of Technology,

4259 Nagatsuta, Midori-ku, Yokohama, 226-8503, Japan

³ Research Center for Functional Materials,

National Institute for Materials Science, 1-1 Namiki, Tsukuba 305-0044, Japan

⁴ International Center for Materials Nanoarchitectonics,

National Institute for Materials Science, 1-1 Namiki, Tsukuba 305-0044, Japan

Abstract

We develop a deep-learning-assisted robotic assembly system of van der Waals (vdW) heterostructures. The system can automatically search atomically thin 2D crystals on SiO₂/Si substrate and stack four cycles of designated 2D crystals per hour with a few minutes of human intervention for each stack cycle. The system enabled the fabrication of the superlattice consisting of 29 alternating layers of the graphene and the hexagonal boron nitride. This capacity provides access to experiments on complex vdW heterostructures.

1. Introduction

Van der Waals heterostructures are comprised of stacked atomically thin two-dimensional crystals and serve as novel materials providing unique properties. However, since the advent of the mechanical exfoliation technique of atomically thin two-dimensional crystals and vdW heterostructures, random nature in positions and shapes of exfoliated two-dimensional crystals have required the repetitive manual tasks of optical microscopy-based searching and mechanical transferring, thereby severely limiting the complexity of heterostructures. To solve the problem, we develop a robotic system that automatically searches exfoliated 2D crystals and assembles them into vdW superlattices. The system is comprised of (i) automated microscope to search atomically thin 2D crystals on SiO₂/Si substrate, (ii) computer-assisted design system of vdW heterostructures, and (iii) robotic stacking system of 2D crystals (Fig. 1).

2. Automated searching system of atomically thin 2D flakes

The motorized optical microscope scans the surface of SiO₂/Si substrate with exfoliated 2D crystals, and the optical images are processed through an algorithm that extracts the region of targeted two-dimensional materials. By using the conventional rule-based image recognition algorithms [1], this system enabled searching 10000 graphene monolayers in 8 hours with a small error rate (<7%). As a step forward to the rule-based image processing, one can utilize unsupervised machine learning to classify graphene flakes [2]. A large number of optical microscope images of SiO₂/Si sur-

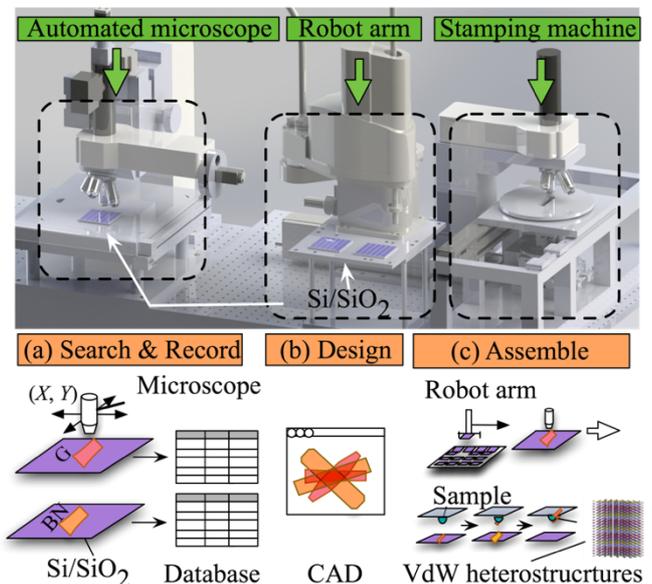


Fig. 1. (top panel) Computer assisted design schematics of automated assembly system. (middle panel) Schematics for fabrication process. (a) Atomically thin two-dimensional crystals on SiO₂/Si are searched by automated optical microscope and their information are stored in database. (b) The combination of 2D crystals for vdW heterostructures are designed by CAD software. (c) The designed vdW heterostructures are assembled by stamping machine.

face were analyzed to extract feature values of Hue, Saturation, and Value. The feature values formed discrete clusters in (H, S, V) space, which corresponds to silicon substrate and graphene flakes with 1, 2, 3, ... layers thick. Unsupervised machine-learning identified each class with an accuracy of ~90%. Further forward, deep learning image recognition algorithms can segment various 2D materials in optical microscope images (Fig. 2(a)) [3]. Through training with many flake images, the deep-learning algorithms acquire a skill to identify atomic layers on SiO₂/Si substrates. The detection process is robust against changes in the microscopy conditions, such as illumination and color balance, which obviates the parameter-tuning process required for the

rule-based detection. Integrating the algorithm with a motorized optical microscope enables the automated searching of 2D materials such as graphene, hBN, MoS₂, and WTe₂ (Figs. 2(b)-(i)). These developments allowed us to utilize a large number of 2D materials simply by exfoliating and running the automated searching process.

3. Robotic assembly of vdW heterostructures

After searching, the big catalog of available 2D crystals are developed in a relational database. vdW heterostructures are designed by selecting 2D crystals from database. Designated 2D flakes are stacked by the pick-up method using robotic arms and a movable stage, which are autonomously controlled on a computer screen. The targeted 2D crystals are automatically aligned to the center of optical microscope by utilizing the template matching. The automated alignment and high-yield transfer process allowed us to fabricate vdW superlattices of ~30 layers in less than one day (Fig. 3). Besides, the system produces various vdW superlattices: trilayer graphene, a graphene/hBN Moiré superlattice, an hBN tunnel device, monolayer tungsten disulfide contacted by graphene, and twisted monolayer-bilayer graphene (Fig. 3). The devices exhibited the highest transport and optical properties, demonstrating that the proposed system provides a practical approach for fabricating vdW superlattices. The high complexity of the heterostructures enabled by this system creates access to a variety of experiments on complex vdW heterostructures.

4. Conclusions

We developed the autonomous robotic assembly system of vdW heterostructures. Because this system is contained in a glovebox, the system can handle oxygen- and humidity-sensitive 2D crystals, such as black phosphorous and niobium diselenide. By this development, we can reduce the human intervention involved in the vdW heterostructure fabrication by orders. The wider material design freedom enabled by our system offers unprecedented opportunities for exploring the full potential of vdW heterostructures.

Acknowledgements

This work is supported by the Core Research for Evolutional Science and Technology (JPMJCR15F3 and JPMJCR16F2).

References

- [1] S. Masubuchi *et al.*, Nature Communications **9** (2018) 1413.
- [2] S. Masubuchi and T. Machida, npj 2D Materials and Applications **3**, (2019) 4.
- [3] S. Masubuchi *et al.*, npj 2D Materials and Applications **4**, (2020) 3.

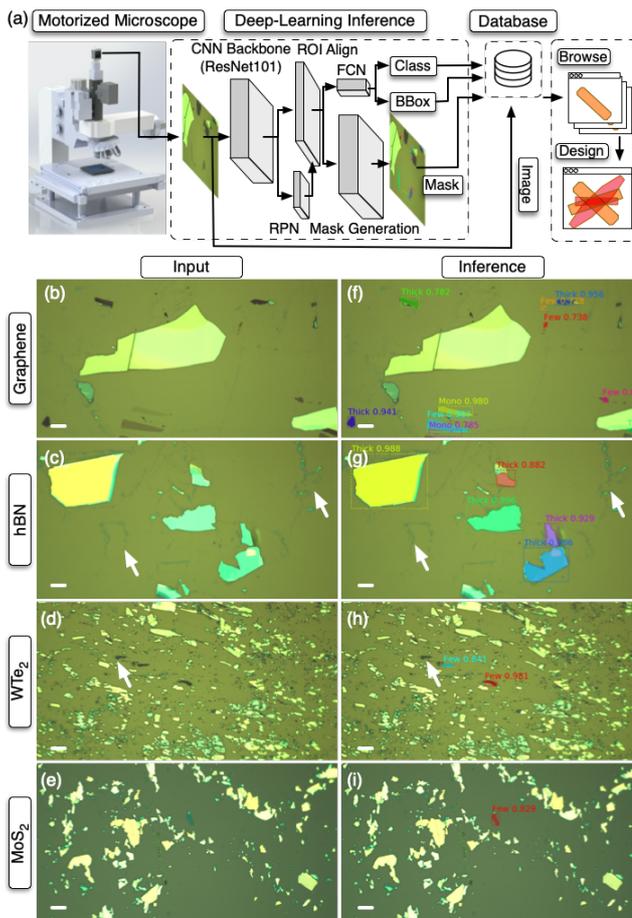


Fig. 2. Schematic of the deep-learning-assisted optical microscope system to search for exfoliated 2D crystals. The optical microscope images are input into the trained deep-learning inference algorithm. The inference results and images are stored in a database. (d)-(i) Segmentation of 2D crystals. Optical microscope images of (d) graphene, (e) hBN, (f) WTe₂, and (g) MoS₂, and (h)-(k) inference results for (b)-(e), respectively.

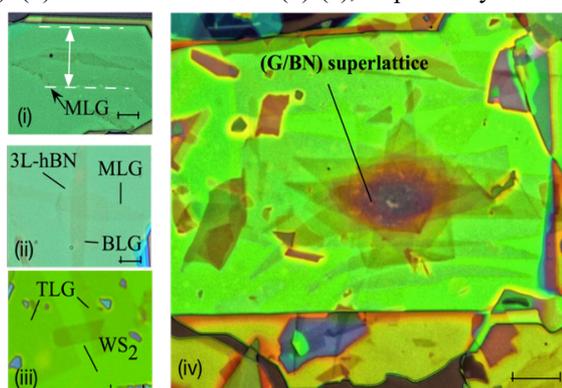


Fig. 3. Optical microscopy images of fabricated vdW heterostructures. (i) monolayer graphene encapsulated in h-BN with crystallographic alignment. (ii) monolayer graphene/3L-hBN/bilayer graphene tunnel device. (iii) WS₂ contacted by trilayer graphene. (iv) (graphene/hBN)₁₄ superlattice structure. Scale bar corresponds to 5 μ m.