Optical Nano-science Research Section

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1. Introduction

We are engaged in fundamental and applied research of nano-materials from a viewpoint of optics and material science. Our research aims to explore new physical and chemical phenomena leading to the applications of novel nano-materials including carbon nanotubes, layered transition metal dichalcogenides, perovskites for the efficient utilization of light energy and the development of future optoelectronic devices with ultra-low energy consumption. The followings are main the research achievements in the year of 2024.

1. Robotic mechanical exfoliation of two-dimensional semiconductors combined with Bayesian optimization

Recently, atomically thin two-dimensional (2D) materials including graphene, and monolayer transition metal dichalcogenides have attracted much attention in a variety of disciplines due to their electronic and optical properties that do not appear in their bulk crystals. Several methods for fabricating monolayer 2D semiconductors with thicknesses of only a few nanometers are mechanical exfoliation, chemical exfoliation, chemical vapor deposition, and so on. Among these methods, the mechanical exfoliation from bulk single crystals has been widely and frequently employed to fabricate the high quality graphene and monolayer 2D semiconductors.

Nevertheless, the mechanical exfoliation method itself is recognized as a simple process; however, it encounters significant bottlenecks due to the substantial manpower requirements. The process is composed of numerous steps, such as substrate cleaning, exfoliation of bulk single crystals, transfer of small flakes to the substrate, and monolayer detection, which make it challenging to efficiently prepare the large-area monolayer 2D materials over 100 μm². Moreover, the experimental conditions must be carefully selected from a vast number of potential parameters such as types of tape, folding time, peeling velocity, and so on, and the detailed microscopic mechanism of mechanical exfoliation itself has yet to be elucidated. At the present stage, the only ways are to wait for serendipity by repeating the many experimental trials, or to rely on experienced and skilled researchers to find large graphene and monolayer 2D materials. Recently, several advanced methods have been proposed to support the

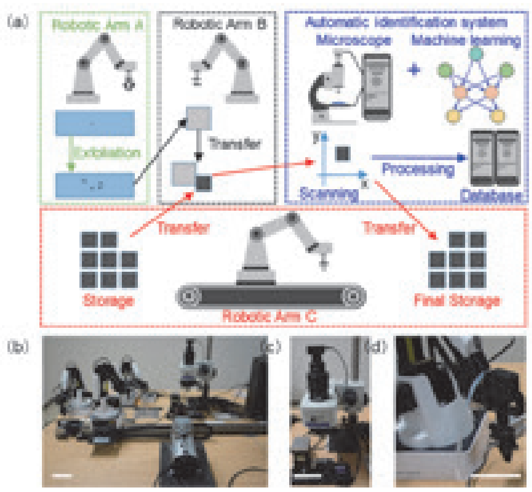

Figure 1 Schematics, functionalities, and photographs of the developed robotic system. (a) Schematic representation of the 2D monolayer exfoliation and searching process. Initially, the single crystals of 2D materials on the blue-tape are repetitively folded by the robotic arm-A. Subsequently, the small flakes of 2D materials are transferred to the Si substrate by the designed stamp of the robotic arm-B. An automatic detection system based on machine learning is employed to identify the 2D monolayers and catalogue their position in a database. Subsequently, the Si substrate with monolayer 2D materials is storaged. (b-d) Photographs of (b) the entire system, (c) the optical automatic detection and identification platform, and (d) the blue-tape folding apparatus. The scale bars in the photographs correspond to 10 cm.

fabrication process by using machine learning to identify monolayer 2D materials and robots to automatically stack exfoliated 2D materials. These efforts represent the next generation of research aimed at combining robotic systems and machine learning to efficiently advance all research in the field of 2D materials on a large scale. In this context, it is strongly required to develop the efficient and highly reproducible strategy to fabricate the large-area and high-quality monolayer 2D materials by mechanical exfoliation, which would provide a significant impact on the wide range of research area in 2D materials fundamental research and development.

Figure 1a illustrates the schematics of the developed robotic fabrication and searching system for high-speed and reproducible mechanical exfoliation and detection of monolayer 2D semiconductors. The system is composed of robotic mechanical exfoliation, transfer from the mechanical exfoliation to detection, and automatic searching and identification of monolayer 2D semiconductors equipped with a microscope. Figures 1b-d show the optical images of the whole robotic system we have developed, the optical automatic identification platform, and the folding device, respectively.

We attempted to efficiently explore the optimized experimental conditions for mechanical exfoliation by combining the developed robotic system with Bayesian optimization (BO), because the mechanical exfoliation is a simple process; however, it involves a huge number of experimental conditions. In BO, it is necessary to set parameters that have a significant impact on the results in order to efficiently explore a large parameter space, and the parameter space was set based on experiments conducted with the developed robotic system. Moreover, we systematically varied and selected the parameters that had a greater effect on the results of mechanical exfoliation. The type of blue-tape, number of blue-tape folding, peeling velocity, and number of transfer onto the blue-tape were selected, resulting in a total of 12,000 experimental conditions. Such a huge number of experimental conditions makes the tasks difficult for optimization without the support of data science approaches such as BO.

Several initial conditions should be provided to construct an appropriate model function for the BO algorithm. The initial ten experimental conditions are selected from all the experimental conditions (12,000 experimental conditions) using the D-optimization criterion to ensure that the characteristics among the parameters are not constant. Considering the results of these initial conditions, the appropriate kernel function45 is used to construct the model function. Moreover, the acquired number and integrated total area of monolayer flakes were treated as indicators. However, these are not suitable for the BO algorithm due to their high variance even under the same experimental conditions. A new index that indicates the quality of mechanical exfoliation results needs to be designed according to the target, i.e., efficiently acquiring many monolayer flakes larger than 100 μm^2 suitable for the fabrication of devices and vdW heterostructures.

We introduced the new evaluation index as a large exfoliated area performance (LEAP). The LEAP is an index that indicates how large and many monolayer WSe_2 can be efficiently obtained with a threshold areal size of 100 μm^2. The LEAP is composed of the value of monolayer appearance probability (MAP: the percentage of monolayers produced that exceed 100 μm^2) and large exfoliated area score (LEAS: the average size of monolayers that can be fabricated), corresponding to indices for the number and area of monolayer flakes, as described beow,

$$\mathrm{LEAP} = a_1 \frac{n}{N} + a_2 \frac{S}{N \times 100}$$

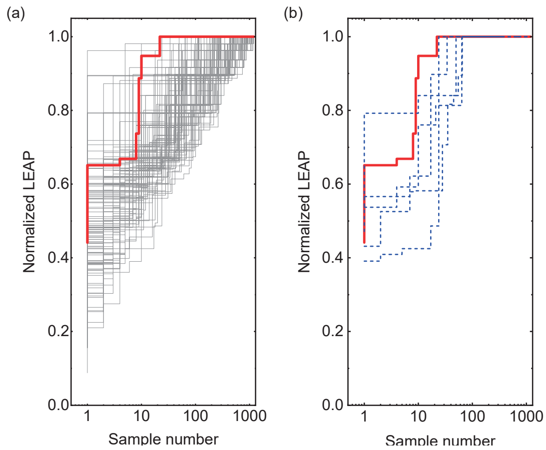

Figure 2 Convergence performances in Baysian optimization and random simulation. (a) Comparison of convergence performance between Bayesian optimization and random simulation. Grey lines and red line show the simulated traces by the random simulation, and the Baysian-optimization, respectively. (b) Blue lines show the fastest five traces that reached to convergence in the random simulations. The score of LEAP are normalized to compare.

where the first term and second term correspond to MAP and LEAS, and $a_{1(2)}$ are coefficient of MAP and LEAS, respectively, and N and n indicate the total number of WSe_2 monolayers larger than 10 μm² and 100 μm² , respectively. Also, S indicates the total area of WSe_2 monolayers larger than 10 μm².

We simultaneously compared the results of BO with predicted random simulation in order to show the effectiveness of BO in the mechanical exfoliation process. Figure 4a shows a simulated benchmark of calculated LEAP by selecting the parameters in the framework of random simulation and BO. In the random simulation, 1,200 randomly selected conditions are chosen from all experimental conditions of 12,000, and 100 patterns are prepared to compare how quickly the maximum score of LEAP is reached to the score derived in BO. The predicted score in the random simulations is calculated based on a model function constructed in BO. Moreover, the maximum LEAP score in each simulation pattern is normalized to compare the number of trials required to reach the maximum score. The LEAP in BO reaches the maximum score after only 23rd trials, as shown in Figure 2a, while the LEAP in random simulation reaches the maximum score after an average of 485 trials. Figure 2b shows the evolution of predicted LEAP in BO and random simulation for the selected fastest five patterns, as a function of trials. These results clearly show that the Bayesian algorithm is much more effective and a superior optimization than the random algorithm for the optimization of mechanical exfoliation process. These results suggest that a robotic system using the BO algorithm for mechanical exfoliation would be useful for efficient fabrication of many other monolayer 2D materials.

Collaboration Works

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松田一成，学術変革領域研究(A)，２.５次元構造の分析技術開発（計画研究）

松田一成，学術変革領域研究(A)，２.５次元物質科学の総括（分担金）

松田一成，基盤研究(S)，原子層人工ヘテロ構造におけるバレースピン量子光学の開拓と応用

松田一成，基盤研究(S)，低次元状態の埋め込みに基づく新物質科学（分担金）

松田一成，挑戦的研究（萌芽），バレースピン制御とデバイス応用：バレートロニクスに向けた課題と挑戦

朝田秀一，特別研究員奨励費，革新的エネルギー変換に向けた原子層人工ヘテロ構造の自発的光起電力に関する研究

2. Others

松田一成，科学技術振興機構 戦略的創造研究推進事業（CREST），二次元半導体・ヘテロ構造の量子光プラットフォームの構築と応用

松田一成，宮内雄平，東京都公立大学法人，配向制御ナノチューブを基盤にした日米共同クリーンエネルギー技術開発

篠北啓介，科学技術振興機構 創発的研究支援事業（FOREST），半導体モアレ超構造を用いた量子電磁力学の創生

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