Dry transfer and optical properties of CVD-grown transition metal dichalcogenides

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Improving the transfer technique of 2D materials is an important issue for their fundamental study and future applications. Even though much progress has been made, the search for exfoliated thin and sufficiently large flakes of 2D materials is still very time-consuming. In contrast, the use of triangular single crystals of transition metal dichalcogenides (TMDCs) allows for easy determination of crystal orientation and thus easy control of twist angles. However, there have been only a few reports on dry transfer and its effect on the optical properties of CVD-grown TMDCs. One of the major technical difficulties is the efficient peeling of CVD-grown monolayers from growth substrates. In general, CVD-grown monolayers are known to adhere strongly to growth substrates compared to exfoliated flakes. Compared with the solution or chemical assisted transfer of CVD grown TMDCs, the dry transfer should keep the intrinsic properties of as grown TMDCs because the solution processes induce physical or chemical property modulations of TMDCs. To expand the versatility of this growth-assisted approach, it is highly desirable to demonstrate the dry transfer and investigate its effects on physical properties.

Here, we report the dry transfer and optical properties of CVD-grown TMDCs using a simple acrylic resin stamp [1]. We introduced the melting and solidification process of an acrylic resin stamp in contact with the sample to improve the adhesion of the stamp. This improvement allows us to efficiently pick up the single crystals of various TMDC monolayers with desired grain size and density from the SiO₂ surface, and to perform high-throughput and continuous dry transfer. Furthermore, we also fabricated the hBN-encapsulated TMDC monolayers and various twisted bilayers including MoSe₂/MoS₂, MoSe₂/WSe₂, MoSe₂/WS₂. The interlayer interaction and quality of dry-transferred, CVD-grown TMDCs were characterized by using photoluminescence (PL), cathodoluminescence (CL) spectroscopy, and cross-sectional electron microscopy. The prominent PL peaks of interlayer excitons can be observed for the MoSe₂/MoS₂ with small twist angles at room temperature (Fig.1). We also found that the optical spectra were locally modualted due to the nanosized bubbles, which are formed by the presence of interface carbon impurities (Fig.2). The present findings provide a widely applicable potential of the present method and enable an efficient search of the emergent optical and electrical properties of TMDC-based vdW heterostructures.

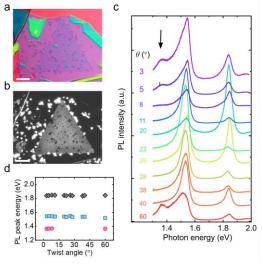
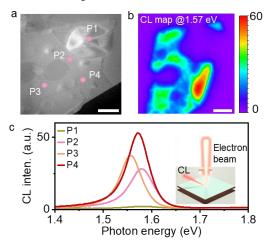


Figure 1. (a) Optical and (b) PL images of hBN encapsulated MoS₂/MoSe₂ heterobilayers. Scale bars are 10 μm. (c) Room-temperature PL spectra of twisted area with various twist angle. (d) PL peak positions of intralayer exciton from MoS₂ and MoSe₂, and interlayer exciton from MoSe₂/MoS₂ heterobilayer with different twist angles.



Figre 2. (a) CL analysis of hBN-encapsulated monolayer MoSe₂. (a) STEM image of hBN-encapsulated monolayer MoSe₂. (b) CL map at 1.57 eV of the same area as (a). Scale bars in (a) and (b) are 1 μm. (c) CL spectra recorded at the positions in (a).

References

[1] H. Naito, W. Zhang, Y. Miyata et al., *Nanoscale Adv.* **2023**, *5*, 5115.