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Dry Release Transfer of Graphene and Few-layer *h*-BN by utilizing Thermoplasticity of Polypropylene Carbonate for Fabricating van der Waals Heterostructures

The dry release transfer of two-dimensional (2D) materials such as graphene, hexagonal boron nitride (*h*-BN), and transition metal dichalcogenides (TMDs) is a versatile method for fabricating high-quality van der Waals heterostructures. Until now, polydimethylpolysiloxane (PDMS) sheets have been widely used for the dry release transfer of TMD materials. However, this method has been known to have limitations that make it difficult to transfer few-layer-thick graphene and *h*-BN because of the difficulty to fabricate these materials on PDMS. As an alternative method, we demonstrate the dry release transfer of single- and bi-layer graphene and few-layer *h*-BN in this study by utilizing poly(propylene) carbonate (PPC) films [1]. Because of the strong adhesion between PPC and 2D materials around room temperature, we demonstrate that single- to few-layer graphene, as well as few-layer *h*-BN, can be fabricated on a spin-coated PPC film/290-nm-thick SiO₂/Si substrate via the mechanical exfoliation method. In addition, we show that these few-layer crystals are clearly distinguishable using an optical microscope with the help of optical interference [Fig. 1(a)]. Because of the thermoplastic properties of PPC film, the adhesion force between the 2D materials and PPC significantly decreases at about 70 °C. Therefore, we demonstrate that single- to few-layer graphene, as well as few-layer *h*-BN flakes, on PPC can be easily dry-transferred onto another *h*-BN substrate [Figs. 1(b) and 1(c)]. This method enables a multilayer van der Waals heterostructure to be constructed with a minimum amount of polymer contamination. We demonstrate the fabrication of encapsulated *h*-BN/graphene/*h*-BN devices and graphene/few-layer *h*-BN/graphene vertical-tunnel-junction devices using this method. Transport measurements revealed high quality of these devices [Fig. 1(d)]. Further, we also demonstrate a fabrication of Top-gate/*h*-BN/graphene/*h*-BN/bi-layer graphene/*h*-BN/graphene/*h*-BN/graphite gate heterostructure using this method; this requires 8 times of successive dry transfer of graphene and *h*-BN. Since devices fabricated by this method do not require an edge-contact scheme, our finding provides a simple method for constructing high-quality graphene and *h*-BN-based van der Waals heterostructures.

References

- [1] K. Kinoshita, R. Moriya, M. Onodera, Y. Wakafuji, S. Masubuchi, K. Watanabe, T. Taniguchi, and T. Machida, npj 2D Materials and Applications **3**, 22 (2019).

Figures

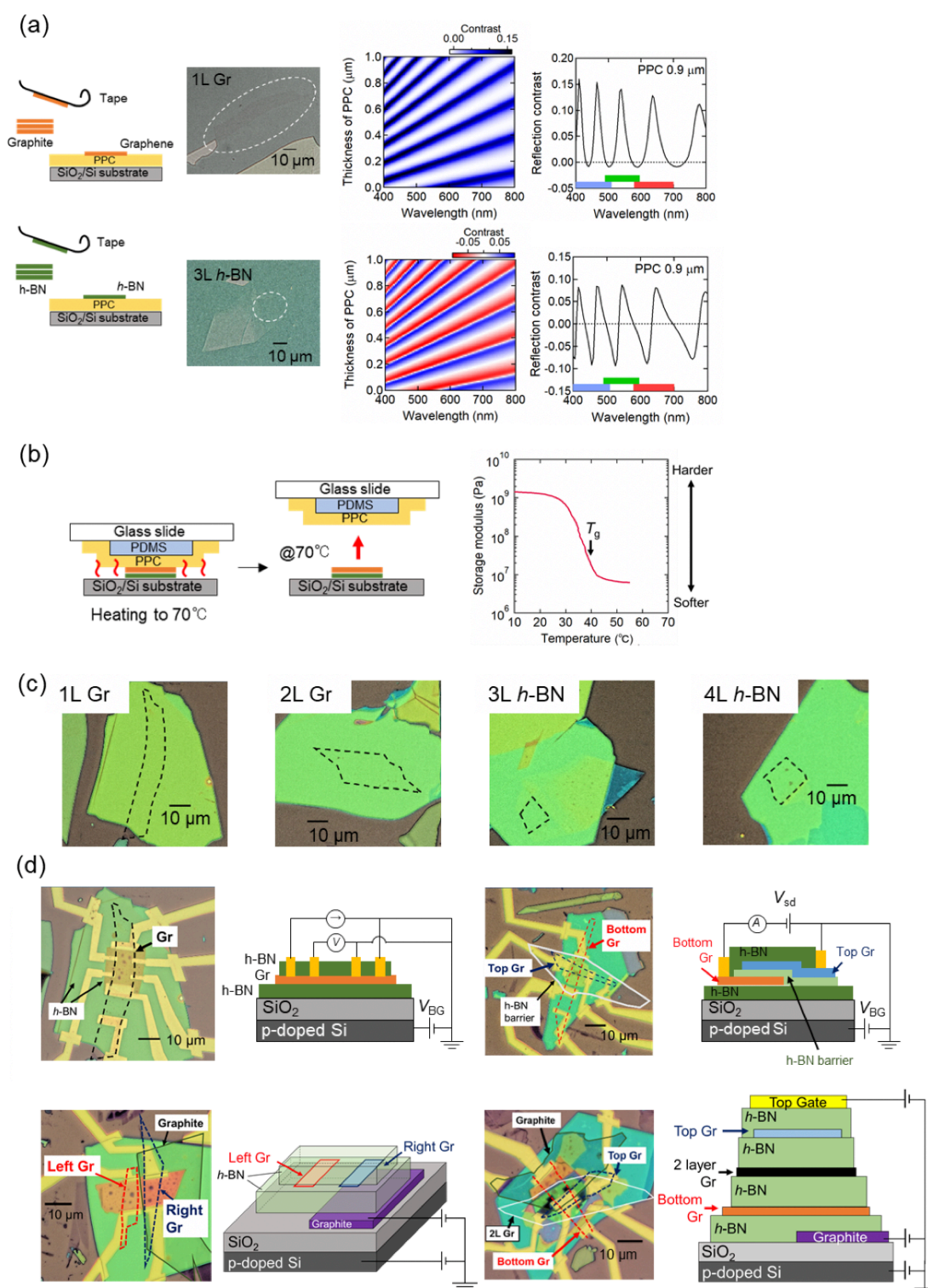


Figure 1: (a) Optical micrograph and calculated optical contrast for monolayer graphene or three-layer *h*-BN prepared on spin-coated PPC/SiO₂/Si substrate. (b) Illustration for dry release transfer method of 2D materials. The temperature dependence of storage modulus of PPC is also shown. (c) Optical micrograph of single-layer graphene, bi-layer graphene, three-layer *h*-BN, and four-layer *h*-BN on thick *h*-BN, respectively. (d) Devices prepared by this method. Left top: *h*-BN/graphene/*h*-BN Hall bar device. Right top: *h*-BN/graphene/few layer *h*-BN/graphene/*h*-BN tunneling transistor. Bottom: multi-stack vdW heterostructures including bottom graphite gate.