Low temperature growth of clean single layer hexagonal boron nitride flakes and film for graphene-based field-effect transistors

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Two dimensional (2D) materials have attracted intense attention owing to their unique physical and chemical properties and the realization of artificial structure and functionality [1]. Insulating hexagonal boron nitride (h-BN), a one-atom thick crystal with a large bandgap (6 eV) [2], superior chemical and thermal stability, and a higher thermal conductivity, is known as “white graphene” due to their similar structure—a lattice mismatch of only 1.6%. The surface of h-BN is chemically inert and lacks charge traps and dangling bonds compared with conventional SiO₂ substrates [3–6]. h-BN films as dielectric layers with all of these properties enable integration with graphene for improving the performance of graphene devices [3–7]. In general, while the complete control over h-BN properties *via* single growth technique such as quality, crystallinity or layer number is difficult, controlling certain parameters of h-BN in growth process is allowed. In particular, the realization of low cost and high quality h-BN remains a great challenge at present.

Recently, several methods have been developed to grow monolayer or multi-layer h-BN film. In these methods, chemical vapor deposition (CVD) is very popular for obtaining high yield, better quality and controllable h-BN film on different metal substrates (Cu, Ni, Pt) at growth temperature as high as 1,000°C [8–18]. While exfoliated h-BN films are demonstrated to be a super dielectric layer for graphene devices, in practice, the performance of as-grown products as dielectric substrate of graphene field-effect transistor (FET) devices is dependent on the quality of h-BN and interface properties between graphene and h-BN [8]. For industrial-level synthesis, the growth at low

temperature is preferred for compatibility with the existing technology as energy consumption is reduced. Indeed, a method with low-energy consumption is highly desired for the synthesis of clean high-quality h-BN at low temperature. Plasma-enhanced CVD (PECVD) allows the low-temperature growth of h-BN on metals [19,20], or even on dielectric surface [21,22]. The PECVD process, usually at 400°C, is able to reduce energy cost and potentially useful for various applications in material fabrication industries. However, there are several challenges by this technique to grow h-BN film. On the one hand, amorphous state could be present in h-BN thin films produced by PECVD mainly due to the low preparation temperatures. On the other hand, ammonia borane molecules used as precursor are dissociated into three products: hydrogen, polyiminoborane (BH₂NH₂, solid), and borazine ((HBNH)₃, gas; active species for h-BN growth) [5,9,12–15], in which very active monomeric aminoborane forms polymeric aminoborane non-crystalline nanoparticles (50–100 nm) [12]. At present, there is no report overcoming these challenges.

Here we prepared high-quality triangle-shaped h-BN flakes and films on Cu surface *via* PECVD method at low temperature (500°C), which is the lowest growth temperature reported to date *via* solid precursor (ammonia borane complex). The synthesized h-BN flakes are regular triangular-shaped, monolayer and single crystalline by adjusting the growth parameters, such as the ratios of Ar to H₂, plasma power and the distance between Cu foil and the reaction precursor. The side effect of aminoborane nanoparticles is eliminated by wrapping the precursor up

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in tin foil. More importantly, graphene FET using the h-BN films as the dielectric layers shows the highest hole and electron mobility of 10,500 and 4,750 $\text{cm}^2 \text{V}^{-1} \text{s}^{-1}$, respectively, which are higher than those using high temperature CVD [4-7,10], demonstrating high quality of the as-grown h-BN.

We used solid ammonia borane as the source, which has equal number of boron and nitrogen, to explore h-BN growth on Cu surface by PECVD. A heating belt was used to heat the source, and Ar and H_2 gases were used as carrier gas for flowing evaporated source molecules into the reaction chamber. Borazine molecules are produced at temperatures ranging between 90 and 110°C, and they are active species for the h-BN growth on Cu (Fig. 1a). However, monomeric aminoborane is very active, and tends to aggregate to polymeric aminoborane particles that have poor crystallinity but stability at ambient conditions. Experimentally, we found that the triangular shaped h-BN flakes were generally covered with some white nanoparticles (Fig. S1), which were formed by the polymerization of active monomeric aminoborane carried to the h-BN surface by gas flow subjected to plasma bombardment. To reduce the amount of active monomeric aminoborane on Cu surface where h-BN flakes and films grow, we wrapped the precursor up in tin foil to

prevent the diffusion of active monomeric aminoborane to the reaction zone. Fig. S2a, S2b show optical images of the tin foil surface before and after growth, respectively, where many h-BN nanoparticles are formed on tin surface after growth.

The synthesis of h-BN flakes and films on Cu surface was carried out using Ar and H_2 as the carrier gas with 40 sccm and 20 sccm flow rates (standard cubic centimeters per minute) at different growth temperatures (500, 550, 650 and 750°C), a pressure of about 110–120 Pa, and PECVD power of 70 W. To tune the nucleation density of h-BN, Cu foils were annealed to reduce grain boundaries and roughness of Cu surface under Ar/ H_2 atmosphere at 1,060°C for 6 h. Fig. 1b–e show the scanning electron microscopy (SEM) images of h-BN triangular flakes grown on annealed Cu foils at various growth temperatures, where the size distribution of h-BN flakes changed in the range of 1–2, 2–3, 3–4, and 4–5 μm with increasing growth temperatures. Fig. 1f shows h-BN size statistics analyzed from the corresponding SEM images for four examples, illustrating that h-BN nucleation density is reduced at high temperature associated with lower supersaturation and high thermal motion of precursors [5,23]. In addition, the estimated growth rates for each case essentially remain unchanged (about 1–1.5

Figure 1 (a) The schematic showing remote radiofrequency (RF) PECVD setup. (b–e) SEM images of h-BN synthesized at different temperatures (500, 550, 650 and 750°C, respectively). The rest growth parameters are same: using Ar and H_2 as the carrier gas with a flow rate of about 40 and 20 sccm, a pressure of about 110–120 Pa, and PECVD power of 70 W. (f, g) The size distribution and nucleation number as a function of growth temperature. (h, i) SEM images showing that h-BN domains grow into near-fully covered and continuous film for longer growth time of 65 and 70 min, respectively. All scale bars are 10 μm .

$\mu\text{m min}^{-1}$), which is 1–1.5 times faster than those for CVD grown h-BN at high growth temperature (1,000°C) [5]. This result is reasonable as PECVD enables fast dissociation and dehydrogenation rates of precursors compared with conventional thermal CVD. The formation of continuous h-BN film is allowed in this technique, and the increase of growth duration yields gradual merging of as-synthesized h-BN (Fig. 1h), eventually leading to a continuous film without any gap (Fig. 1i).

Fig. 2a and b show the X-ray photoemission spectroscopy (XPS) spectra of the as-grown h-BN on Cu foil at 750°C. The observed B 1s and N 1s binding energies are peaked at 190.37 and 397.97 eV, respectively (Fig. 2a, b), corresponding to the previously-reported locations of h-BN products [5,8–18]. The atomic ratio of B/N analyzed

from the XPS data is about 1.10, approaching 1/1 stoichiometry in h-BN. As shown in Fig. S3a–c, the XPS spectra of h-BN films with different grown temperatures (500, 550 and 650°C) are similar to the spectrum shown in Fig. 2a, indicating that the configuration for B and N atoms of all grown samples adopts the hexagonal phase B–N bonds. To investigate the characteristics of monolayer h-BN films at different growth temperatures, transparent quartz substrates were used as target substrates for the h-BN transfer, and then the samples were characterized by optical absorption. Fig. S4a shows a typical UV-visible absorption spectrum of as-grown monolayer h-BN. h-BN film has approximately no absorbance within the visible-light range, but a large absorption within the UV range, indicating an optical band

Figure 2 (a, b) XPS spectra of B 1s and N 1s for the transferred h-BN samples, respectively. (c, d) Typical AFM images of h-BN flake and film transferred onto SiO₂/Si substrates by PMMA-assisted method, respectively. Insets in each image also show the height of h-BN grain along the labeled lines. All scale bars are 2 μm . (e, i, m and q) Typical TEM images of continuous h-BN films synthesized at 500, 550, 650 and 750°C, respectively. (f–h, j–l, n–p and r–t) SAED patterns recorded on different locations labeled in image of e, i, m and q, respectively.

gap (OBG). OBG is determined to be around 6.04 eV in Fig. S4b [14,24]. The slightly larger value of OBG compared with that of multi-layer or bulk h-BN resulted from the lacking of layer-layer interaction for single layer h-BN [25]. Indeed, our value is in good agreement with that of monolayer h-BN obtained by the theoretical calculation (6.0 eV) [26] and previous experiments [5,9,12,15].

The transferred h-BN flakes and films on SiO₂/Si substrates were characterized by atomic force microscopy (AFM) (Fig. 2c and d). Fig. S5a and S5b show the SEM images of triangular h-BN flakes and films transferred onto the SiO₂/Si substrate, respectively. According to the previous report [5], we annealed the transferred h-BN samples at 950°C for 25 min to get rid of PMMA residues on h-BN surface in air. The treated h-BN flakes and films are about 0.540 and 0.530 nm in height (inset), which are in good agreement with the values obtained for monolayer h-BN [5,12,15]. The Raman spectrum of the h-BN film on the SiO₂/Si substrate shows a peak at 1,369 cm⁻¹, originating from the B–N bond stretching of the BN films (Fig. S6).

Transmission electron microscopy (TEM) was further used to characterize the crystalline structure of the as-grown h-BN film at different growth temperatures (500, 550, 650 and 750°C). Fig. 2e–t show a series of TEM images of the h-BN films grown with different temperatures transferred on the TEM grid along with selected area electron diffraction (SAED) patterns obtained at various locations. As shown in Fig. 2e–h, the film has a uniform contrast in Fig. 2e, and the recorded SAED spots exhibit a 6-fold symmetry that is characteristic of h-BN, suggesting its good crystallinity. Meanwhile, SAED spots obtained at various positions in an specific region of film exhibit the similar orientation (Fig. 2f–h), and two sets of diffraction patterns were also observed at marked position as shown in Fig. 2h, indicating that the h-BN film consists of small domains with various orientations due to small size in the individual flake. However, for the growth temperatures of 650°C and 750°C, the synthesized h-BN film with large domains only shows one set of six-fold symmetric SAED pattern at different locations of the films with same orientation, demonstrating that it is single crystal. The result reflects that h-BN grains are continuously growing as a function of time.

To understand h-BN growth by PECVD, we investigated the effects of various critical parameters on the h-BN growth. Here we discuss the ratios of Ar/H₂ flow, plasma powers, growth temperature, growth duration, and the distance between growth zone and the plasma generator. Accordingly, carrier gas composition is sig-

nificant to h-BN morphology. When Ar/H₂ flows (40/0 and 40/10 sccm) were used in the growth stage, random shaped objects in Fig. 3a and 3b were obtained. Changing the Ar/H₂ flow to 40/20 sccm resulted in the formation of h-BN flakes with triangular shape (Fig. 3c).

Fig. 3d–f display typical SEM images of h-BN flakes grown at different powers of plasma of 50, 70 and 90 W in Ar/H₂ (40/20 sccm) gas flow, respectively. The average grain size is about 1 μm at the condition of 50 W plasma, and the size is about 4 μm using 70 W plasma. The size is about 0.8 μm using 90 W plasma. The plots of h-BN size and the density of nucleation vs. plasma power in Fig. S7a, b display that a maximum size is grown at 70 W and is reduced to 2–3 μm at a power higher than 90 W. This result probably originates from a high concentration of hydrogen species produced at a high plasma power. Here, the hydrogen plays two roles in h-BN growth: an activator and an etching reagent [27]. Furthermore, we found the distance between growth zone and plasma generator obviously influences the synthesized h-BN. When the distance is about 20 and 25 cm, we observed the h-BN flakes with flower-like morphology due to the effective etching (Fig. 3g, h), which is similar to the graphene etching [28]. To decrease the influence of etching, we moved the plasma generator away the growth zone to 35 cm, and then regular and no-etched h-BN flakes were synthesized as shown in Fig. 3i.

In addition, we fabricated FET devices of monolayer graphene domains grown on liquid Cu surface [29,30] using oxidation-cleaned h-BN monolayer films synthesized at different growth temperatures as back-gated and top-gated dielectrics in Fig. 4a, respectively. Briefly, single-layered h-BN film was transferred onto 300 nm SiO₂/Si substrate and oxidized in air, followed by transferring graphene flakes onto h-BN films. Device fabrication protocols were based on the previous reports [29,30], and electrical transport measurements were conducted in ambient condition. Fig. 4b and Fig. S8a–d show a series of optical images of graphene/h-BN films synthesized at different growth temperatures for FET devices. We tested more than 30 devices, and compared the difference of two type models in electrical properties. Compared with SiO₂ substrates, the clean h-BN films for dielectrics exhibit the improved carrier mobility of graphene and the shift of Dirac point towards lower voltage. Fig. 4c shows typical transfer curves for h-BN films used as back-gated dielectric layers. The devices show typical p-type characteristic in agreement with our previous results of graphene/h-BN devices [5]. The shifting of Dirac points for four cases using h-BN as back-gated dielectric layers

Figure 3 (a–c) SEM images of triangular-shaped h-BN domains grown on Cu foil surface with different ratio of gas flow ($\text{Ar}/\text{H}_2 = 40/0, 40/10$ and $40/20$ sccm), plasma power (70 W) and the distance between Cu foil and plasma generator (35 cm). (d–f) SEM images showing the as-grown triangular-shaped h-BN domains with different plasma power (50, 70 and 90 W, respectively), gas ratio (40/20 sccm) and the distance between Cu foil and plasma generator (35 cm). (g–i) SEM images of the as-grown triangular-shaped h-BN domains with different distances between Cu foil and the plasma generator (20, 25 and 35 cm, respectively), gas ratio (40/20 sccm) and plasma power (70 W). All scale bars are 5 μm .

can be clearly seen in Fig. 4c, with $V_g = 12, 5, 10$ and 12 V for h-BN films synthesized at different temperatures (500, 550, 650 and 750°C), respectively. This is tunable by including graphene on SiO_2/Si substrates as its Dirac point usually shifts to a much positive value with similar protocol for the device fabrication and device tests [28]. The shifting of Dirac points for graphene devices reflects graphene doping by substrates or environment. Indeed, the graphene devices on cleaned h-BN films grown at different temperatures show higher hole mobility than that on SiO_2/Si substrates. For instance, hole carrier mobility is about 1,515, 974, 2,356, 3,321 and $10,500 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ at $V_{ds} = 0.01$ V for SiO_2 substrate, h-BN films synthesized at 500, 550, 650 and 750°C , respectively (Fig. S9). The hole carrier mobility of the devices using the low temperature h-BN as dielectric layers is lower than that on the SiO_2 substrates due to the H_2O or other residue. The calculated electron carrier mobility for h-BN films synthesized at 500, 550, 650, 750°C , respectively, is about 302, 392, 1,162 and $4,750 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ at $V_{ds} = 0.01$ V as shown in Fig. S9. The mobility of different devices is improved by h-BN films as back-gated dielectrics grown at a higher temperature, due to the fact that the high temperature results in high crystallinity of h-BN. These results indicate that the quality of substrate is vital for

improving the carrier mobility of graphene devices.

To prevent the degradation of graphene devices from the influence of water or gases in the air, we transferred the h-BN synthesized at 750°C to the post-tested devices as the top dielectric layer and the transferred process were similar to the previous report [5]. According to the tested results (Fig. 4e and f), we found the Dirac point of graphene devices with h-BN as top-gated dielectric layers upshifted to $V_g = 40$ and 32 V (650 and 750°C), respectively, due to the trapped water in the transfer at the h-BN/graphene interface. Owing to the high temperature stability of h-BN in air, the h-BN/graphene/h-BN/ SiO_2/Si devices were annealed at 80°C in vacuum for 3 and 24 h to remove water residues due to the fabricated process. As shown in Fig. 4e and f, the Dirac point of the graphene devices with h-BN as top-gated dielectric downshifted to $V_g = 38$ and 20 V (650 and 750°C), respectively, but all these values are higher than those values in Fig. 4c due to less water residue. Meanwhile, the hole carrier mobility for the un-annealed devices in vacuum is 1,545, 3,321 and $4,000 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ at $V_{ds} = 0.01$ V (SiO_2/Si substrate, 650 and 750°C), respectively (Fig. 4g–i). While annealed for 3 and 12 h in vacuum, the hole carrier mobility is 2,863 and 2,829 (650°C), $5,500 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ (750°C) at $V_{ds} = 0.01$ V. The calculated electron carrier mobility is about 1,261

Figure 4 (a) The schematic diagram of graphene FET devices with h-BN as dielectrics: (1) h-BN as back gate dielectric; (2) h-BN as back gate and top gate dielectric. (b) The optical image of the device with h-BN dielectric grown at 750°C. (c) Transfer curves of graphene/SiO₂ and graphene/h-BN films synthesized at different temperatures. (d) Transfer curves of graphene/SiO₂ capped by h-BN films synthesized at 750°C as the top gate dielectric and annealed in vacuum at 80°C for 3 and 12 h. (e) Transfer curves of graphene/h-BN synthesized at 650°C capped by h-BN films synthesized at 750°C as the top gate dielectric and annealed in vacuum ambient at 80°C for 3 and 12 h. (f) Transfer curves of graphene/h-BN synthesized at 750°C capped by h-BN films synthesized at 750°C as the top gate dielectric and annealed in vacuum ambient at 80°C for 3 h. (g-i) Histograms of hole and electron mobility distribution for SiO₂ and h-BN synthesized at different temperatures (650 and 750°C) as back- and top-gated dielectrics.

and 1,066 (650°C), 3,064 $\text{cm}^2 \text{V}^{-1} \text{s}^{-1}$ (750°C) at $V_{ds} = 0.01$ V, respectively. This trend is very important for improving the performance of the devices through decreasing the doping water in the air.

In summary, the high quality h-BN grew on Cu surface *via* PECVD method with low growth temperature and neat surface. Moreover, we elucidate that the quality of as-synthesized h-BN is modulated by growth temperature, and as dielectrics of graphene FET, the quality of as-grown h-BN is vital for the device performance.

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低温制备高质量六方氮化硼晶畴、薄膜及其在石墨烯基场效应晶体管中的应用

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摘要 二维六方氮化硼是一种理想的石墨烯电学器件介电层材料, 然而, 制备低成本和高质量的氮化硼材料仍是一个挑战. 本文使用等离子体化学气相沉积法, 500°C下在铜箔衬底上制备了三角形的BN晶畴及其薄膜. 通过使用锡箔纸包裹原料的方法避免了残留原料在BN表面的沉积. 当BN作为石墨烯场效应晶体管的介电层时, 基于石墨烯的场效应器件空穴与电子的迁移率分别为10500和4750 cm² V⁻¹ s⁻¹, 明显优于在高温条件下制备的BN作为介电层的石墨烯器件, 间接表明了该方法可得到高质量的BN. 另外, 本工作也揭示了氮化硼的质量对石墨烯场效应器件的性能至关重要.