

Large-area synthesis and transfer of multilayer hexagonal boron nitride for enhanced graphene device arrays

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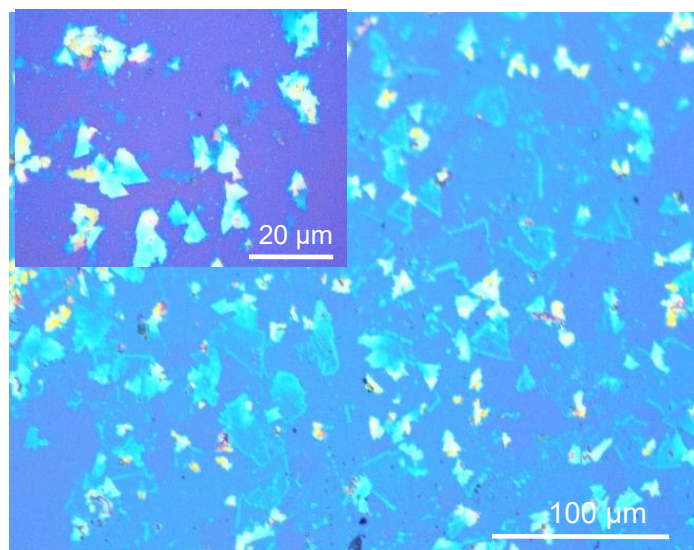
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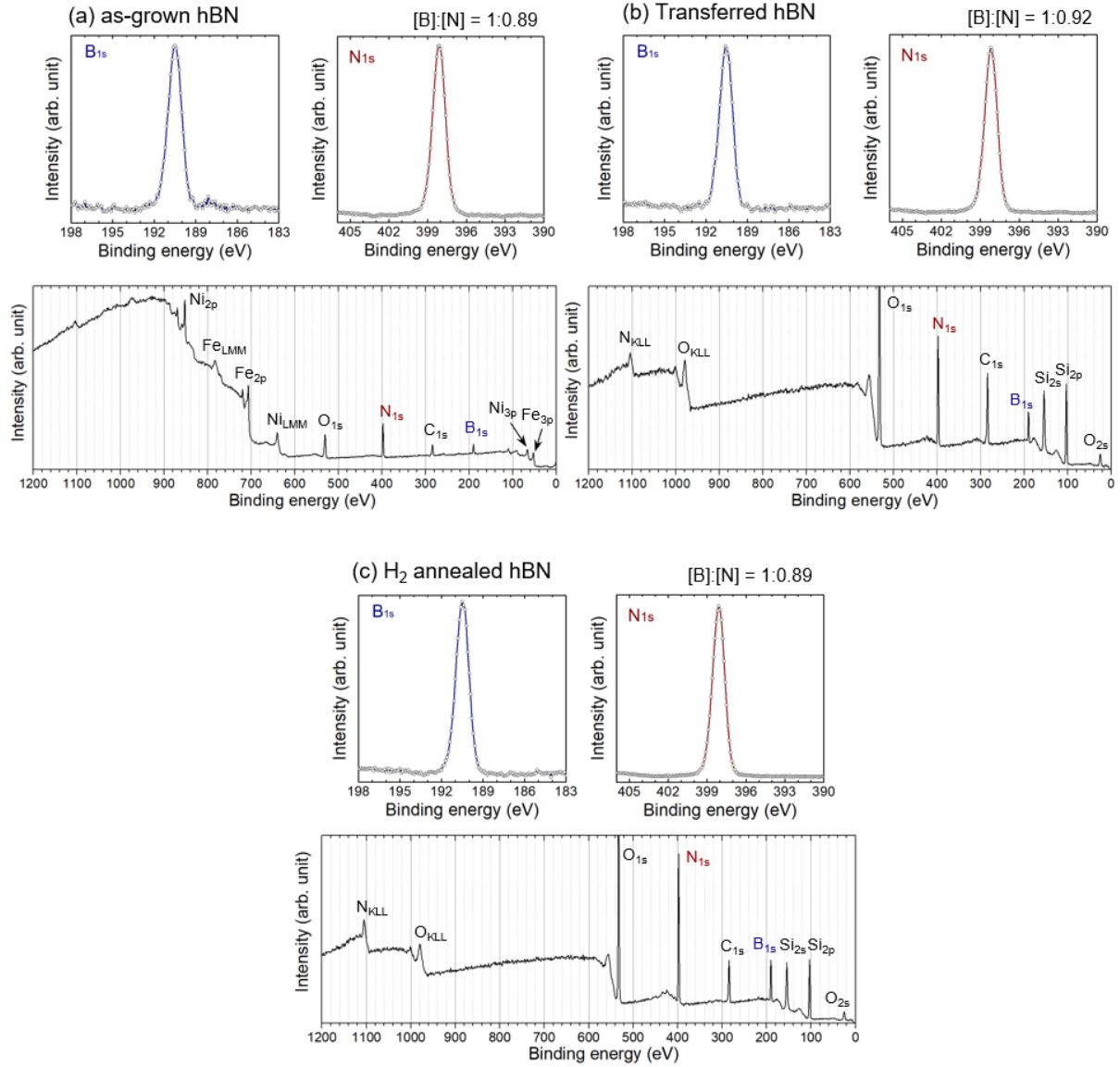
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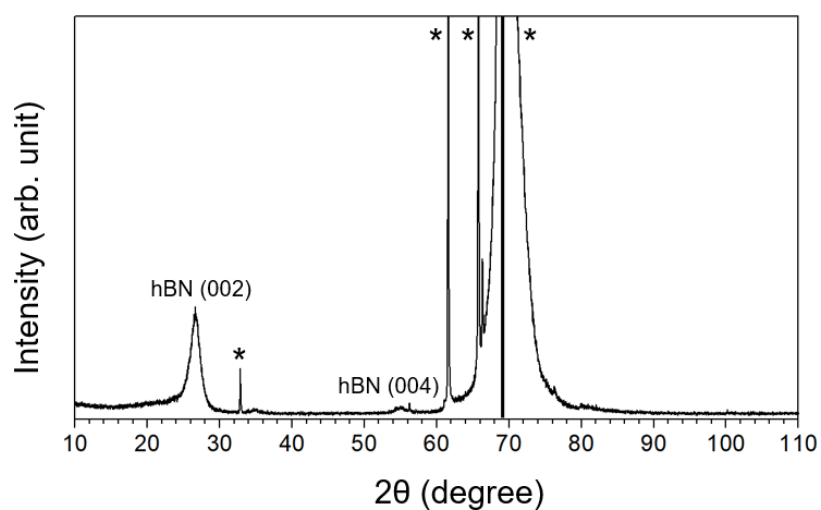
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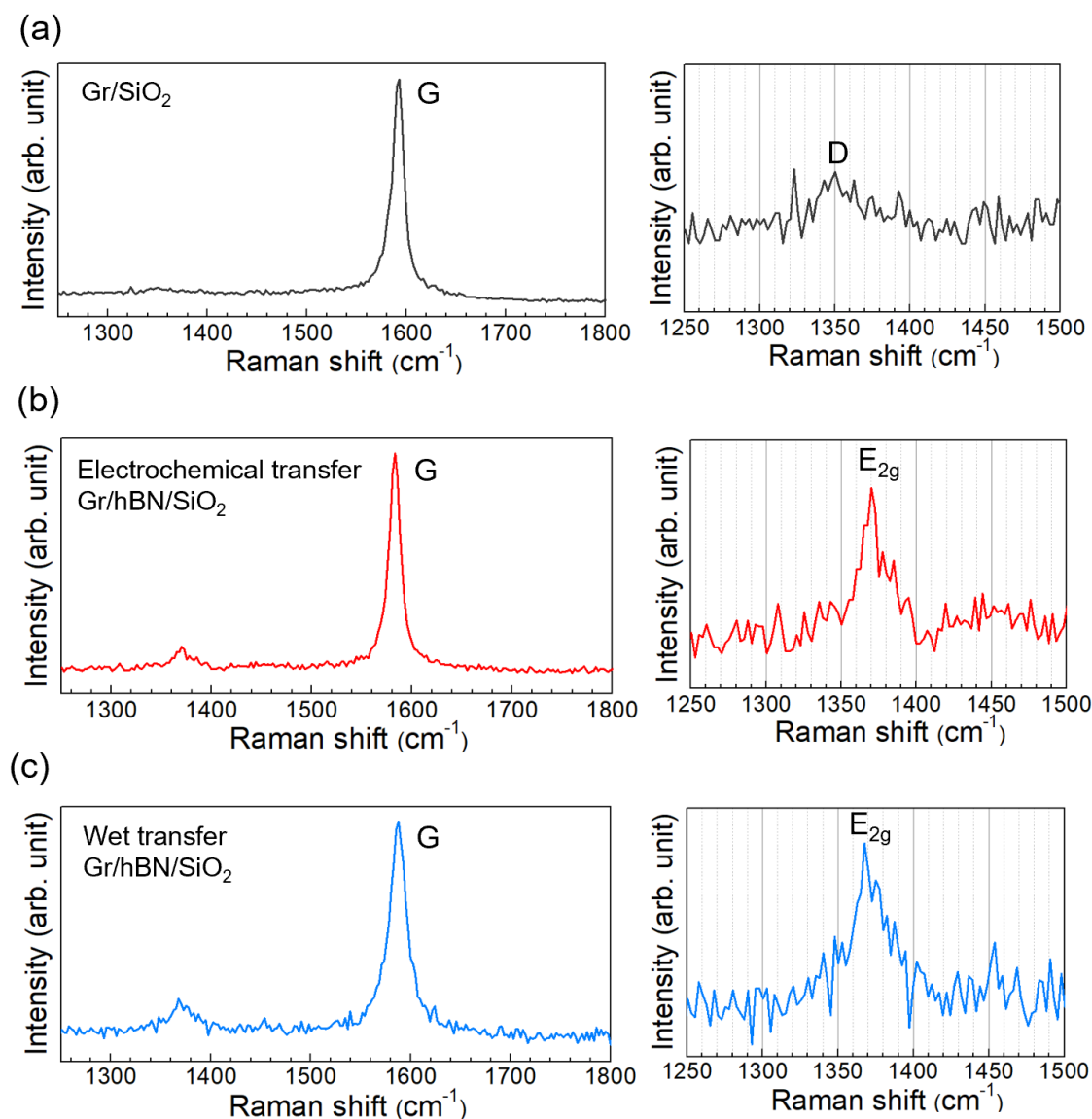
Supplementary Fig. 1. Optical images of multilayer hBN grown on Fe foil. The CVD-grown hBN on Fe foil was transferred on a SiO₂ substrate. These optical micrographs show inhomogeneous hBN thickness. The yellow grains have thickness over 30 nm, as determined by AFM. The Fe foil (purity > 99%, 20 μm thickness) purchased from Nilaco was used for the CVD growth.



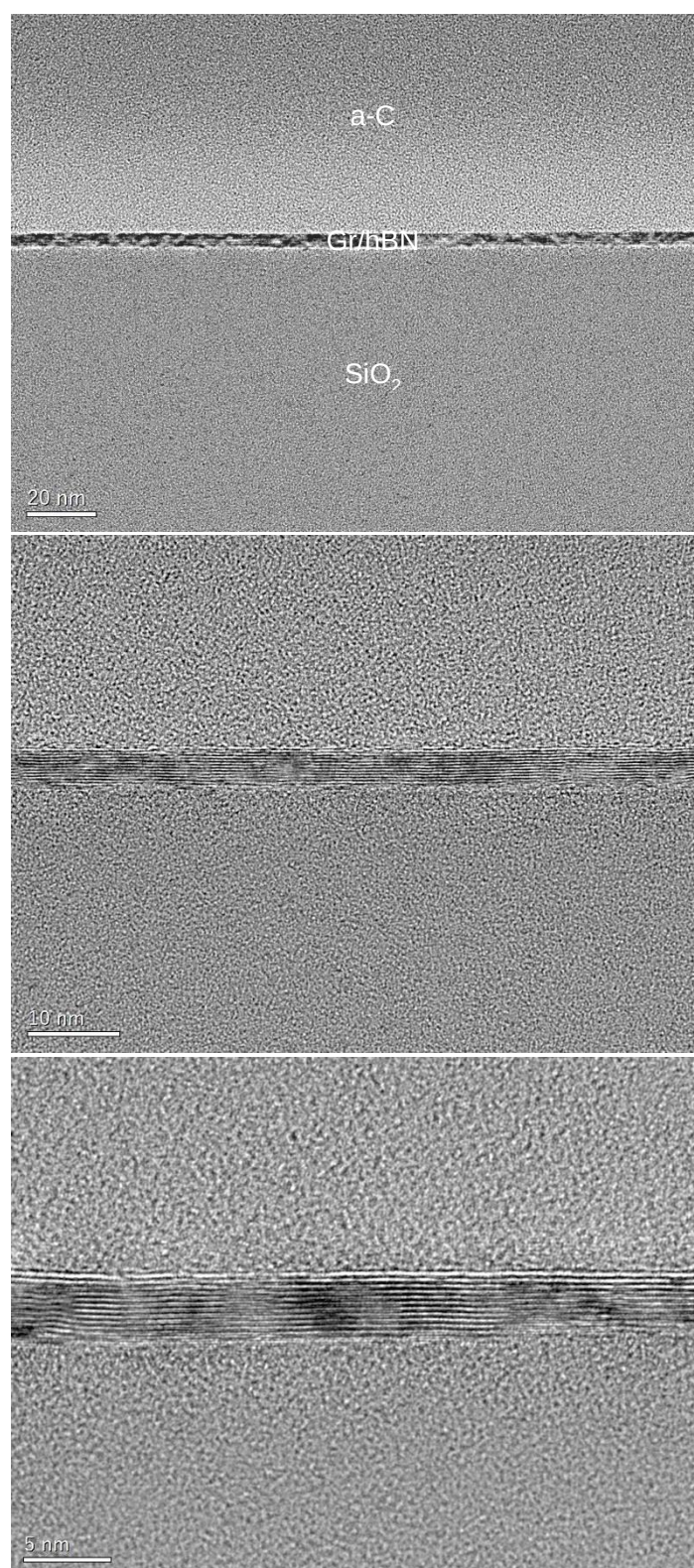
Supplementary Fig. 2. XPS spectra of CVD-grown hBN samples. **a**, As-grown hBN on Fe-Ni foil. **b**, Transferred hBN on SiO₂ substrate. **c**, H₂-annealed hBN on SiO₂ substrate. High resolution XPS spectra of B_{1s} and N_{1s} and wide scan spectra are shown for each sample. The C_{1s} peak became weaker after the H₂ annealing (see **b** and **c**), indicating the removal of surface contamination. The O_{1s} peaks in **b,c** are mainly originated in the SiO₂ substrate.



Supplementary Fig. 3. XRD profile of the electrochemically transferred hBN on a SiO₂/Si substrate. The interlayer distance calculated from the (002) peak is 3.33 Å ($c = 6.66$ Å), in agreement with the distance of hBN crystals. Because the present hBN is not very thick, the hBN(002) diffraction has a relatively broad peak. Asterisks (*) indicate the diffraction from the SiO₂/Si substrate.

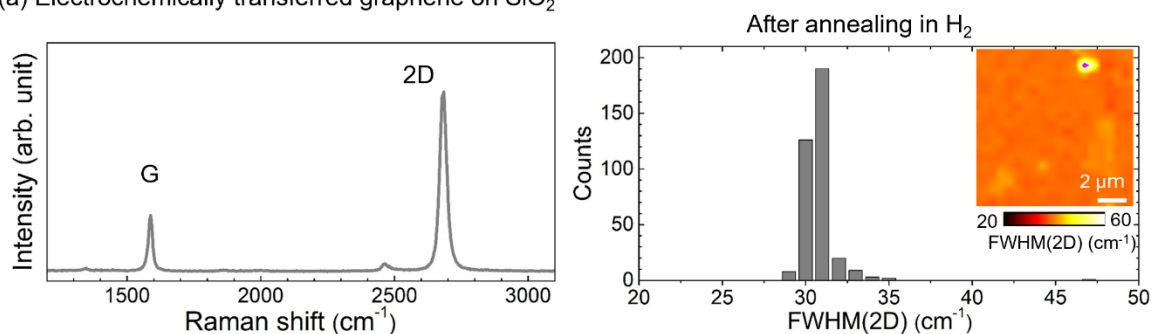


Supplementary Fig. 4. Magnified Raman spectra. **a**, Selected spectral regions of the Raman spectra shown in Fig. 3 of the main text. Right panels display the graphene D band and hBN E_{2g} band. Because D and E_{2g} bands appear at slightly different positions (D band: $\sim 1350 \text{ cm}^{-1}$, E_{2g} band: $1365\sim 1370 \text{ cm}^{-1}$), the peaks observed at $1360\sim 1400 \text{ cm}^{-1}$ in **b** and **c** can be assigned to multilayer hBN and not in graphene.

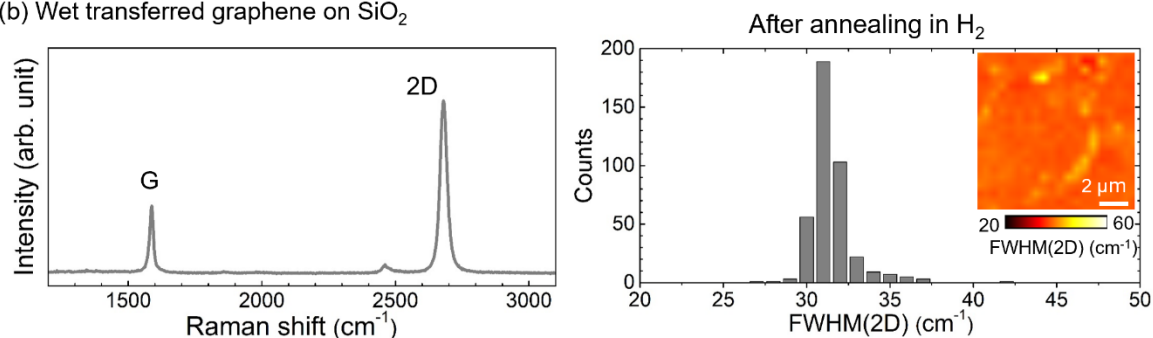


Supplementary Fig. 5. Cross-sectional TEM images of a Gr/hBN_{ele} stack. Top surface of graphene is covered with a-C layer to protect from any damages induced during the FIB process. The hBN is highly uniform with a clear layered structure.

(a) Electrochemically transferred graphene on SiO₂



(b) Wet transferred graphene on SiO₂



Supplementary Fig. 6. Raman analysis of monolayer graphene transferred by different methods. Typical Raman spectra and FWHM(2D) distributions of monolayer graphene transferred by electrochemical (a) and wet etching (b) methods. Insets show the Raman mappings of the FWHM(2D) spatial distribution. The negligibly weak D band indicates that the crystalline quality of monolayer graphene is high in both cases. Different from the case of hBN transfer, the quality of monolayer graphene did not strongly depend on the transfer method.

Supplementary Table 1. Comparison of average carrier mobilities ($\text{cm}^2 \text{V}^{-1}\text{s}^{-1}$) of heterostack devices consisting of monolayer graphene and multilayer hBN, measured at room temperature. The values in the parentheses are the maximum values.

	Carrier type	hBN/Gr/hBN	Gr/hBN	Gr/SiO ₂	Typical channel size ^{*1}
This work	μ_{hole}	5,477 (10,219)	4,196 (7,074)	3,106 (5,217)	W_{ch} 3 μm
	μ_{electron}	5,551 (9,571)	4,692 (7,284)	3,031 (5,384)	L_{ch} 9 μm
Ref. 1	μ_{hole}	-	7,490 (24,000)	6,091	W_{ch} 1.5 μm
	μ_{electron}	-	4,407	5,494	L_{ch} 2 μm
Ref. 2	μ	(5,000~10,000)	-	(3,200~4,100)	W_{ch} 2 μm L_{ch} 4 μm
Ref. 3	μ_{electron}	-	3,030	(1,750)	W_{ch} 11~18 μm L_{ch} 3~10 μm
Ref. 4	μ	(7,400)	(6,300)	(2,800)	NA
Ref. 5	μ	-	(1,200-750)	(260) ^{*2}	W_{ch} 27 μm L_{ch} 7.5 μm

^{*1} W_{ch} and L_{ch} are channel width and length, respectively.

^{*2} SiN was used as a substrate instead of SiO₂.

Kim *et al.* reported an average hole (electron) mobility of 7,490 (4,407) $\text{cm}^2 \text{V}^{-1}\text{s}^{-1}$ (highest value of 24,000 $\text{cm}^2 \text{V}^{-1}\text{s}^{-1}$) for graphene/hBN stacked devices, and only observed an increase with respect to SiO₂ for the hole mobility.¹ Shi *et al.* fabricated hBN/graphene/hBN sandwich structures and obtained carrier mobilities of 5,000-10,000 $\text{cm}^2 \text{V}^{-1}\text{s}^{-1}$ (3,200-4,100 $\text{cm}^2 \text{V}^{-1}\text{s}^{-1}$ for graphene/SiO₂), similar to those reported here.² Both of these works used very thick hBN as a substrate, as seen from the much more intense E_{2g} band than that of our graphene-hBN heterostructure. More importantly, these works used EB lithography to fabricate devices with small channel sizes (channel width and length are 1.5-2 μm and 2-4 μm , respectively).^{1,2} The EB lithography allows to fabricate devices at desired positions by avoiding wrinkles and bubbles in the channels or at least by selecting positions with low wrinkle and bubble densities.

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