Supporting Information

Toward van der Waals Epitaxy of Transferable Ferroelectric Barium Titanate Films via A Graphene Monolayer

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Figure S1. Pretreatment of graphene. (a) SEM image of the as-transferred graphene. (b) SEM image of the graphene after pretreatment. (c) A map of the graphene 2D peak intensity from the Raman spectrum taken on the as-transferred graphene. (d) A map of the graphene 2D peak intensity from the Raman spectrum taken on the graphene after pretreatment.



Figure S2 XPS O1s peak and fitting of the graphene before (lower) and after (upper) pretreatment. The black dots are experimental data and the green lines are fitted profile.



Figure S3 A cross-sectional SEM image of the film shown in Figure 2d.



Figure S4. (a) XRD pattern of a 60 nm-thick BTO film grown on graphene/SiO₂/Si at 700°C. (b) XRD pattern of a 60 nm-thick BTO film grown on SiO₂/Si without graphene. (c) X-ray reflectivity (XRR) pattern of the 60 nm BTO film grown on graphene/SiO₂/Si. (d) Raman spectrum of graphene after the BTO film growth process.



Figure S5. A photo of the exfoliated BTO film on a tape.



Figure S6. (a, c) A SEM image and the EDX spectrum of the exfoliated BTO film. (b, d) A SEM image and the EDX spectrum of the remaining SiO_2/Si substrate.



Figure S7. (a, b) Piezoresponse (phase and amplitude) of the exfoliated BTO film, with a 10 μ m × 10 μ m square written by +5V, a 6 μ m × 6 μ m concentric square written by -5V and a 2 μ m × 2 μ m square written by +5V; the pattern was read immediately after being written.



Figure S8. (a) The phase (upper) and amplitude (lower) responses measured using a piezoresponse force microscope on a single location of the BTO film on Nb doped STO substrate as a function of the sweeping tips bias, which show a hysteresis loop with 180° phase change and a butterfly-like curve, respectively. (b, c) Piezoresponse (phase and amplitude) of the exfoliated BTO film, with a 6 μ m × 6 μ m square written by +6V, a 2 μ m × 2 μ m concentric square written by -6V; the pattern was read out 1 hour after being written.



Figure S9 (a) XPS Ba 3d peak and fitting (b) XPS Ti 2p peak and fitting for the BTO film grown on graphene/SiO₂/Si. The black dots are experimental data and the green lines are fitted profile while black peaks are the deconvoluted peaks.