



Supporting Information

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Electronic Transport in Heterostructures of Chemical Vapor Deposited Graphene and Hexagonal Boron Nitride

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Figure S1. Graphene-hBN heterostructures. (a) Optical micrograph of electrodes in the TLM configuration contacting a graphene ribbon supported on five-layer CVD hBN. (b) Higher magnification optical micrograph of leads on five-layer hBN. (c) Optical image of PMMA etch mask (3 μm wide) spanning TLM design on a graphene-hBN stack.

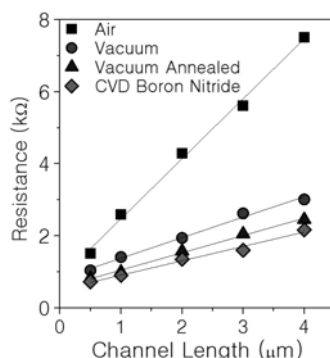


Figure S2. TLM measurements in four different systems (corresponding to Figure 3 in the main text). Resistance is measured for varying channel lengths on the same ribbon. Contact resistance is measured from the linear fit by extrapolating to a channel length of 0 μm . The extracted contact resistivity is reported in Figure 3c.

The field-effect mobility is derived using Equation 4 in Ref. 52:

$$R_T = R_C + (L/W)/(ne\mu)$$

R_T is the total resistance, R_C represents the contact resistance extracted from TLM, L and W represent the length and width of the graphene channel, respectively. We use the relation that $n = (n_0^2 + n_G^2)^{1/2}$ where n_0 is the carrier concentration at the Dirac point and n_G is the carrier concentration induced by the gate voltage. Experimentally, n_G is calculated using the relation $n_G = C_G(V_G - V_0)/e$ where C_G is the oxide capacitance per unit area, V_G is the gate voltage, V_0 is the gate voltage at the Dirac point, and e is the elementary charge. Using the parallel plate capacitor model, the calculated value of the areal gate capacitance C_G is 11.5 nF-cm⁻².

REFERENCES

1. Bolotin, K. I.; Sikes, K. J.; Jiang, Z.; Klima, M.; Fudenberg, G.; Hone, J.; Kim, P.; Stormer, H. L., Ultrahigh Electron Mobility in Suspended Graphene. *Solid State Commun.* **2008**, *146*, 351-355.
2. Bolotin, K. I.; Sikes, K. J.; Hone, J.; Stormer, H. L.; Kim, P., Temperature-Dependent Transport in Suspended Graphene. *Phys Rev Lett* **2008**, *101*.
3. Balandin, A. A.; Ghosh, S.; Bao, W. Z.; Calizo, I.; Teweldebrhan, D.; Miao, F.; Lau, C. N., Superior Thermal Conductivity of Single-Layer Graphene. *Nano Lett.* **2008**, *8*, 902-907.
4. Balandin, A. A., Thermal Properties of Graphene and Nanostructured Carbon Materials. *Nat. Mater.* **2011**, *10*, 569-581.
5. Behnam, A.; Lyons, A. S.; Bae, M. H.; Chow, E. K.; Islam, S.; Neumann, C. M.; Pop, E., Transport in Nanoribbon Interconnects Obtained from Graphene Grown by Chemical Vapor Deposition. *Nano Lett.* **2012**, *12*, 4424-4430.
6. Qi, Z. J.; Rodríguez-Manzo, J. A.; Botello-Méndez, A. R.; Hong, S. J.; Stach, E. A.; Park, Y. W.; Charlier, J.-C.; Drndić, M.; Johnson, A. T. C., Correlating Atomic Structure and Transport in Suspended Graphene Nanoribbons. *Nano Lett* **2014**.
7. Liu, G. X.; Wu, Y. Q.; Lin, Y. M.; Farmer, D. B.; Ott, J. A.; Bruley, J.; Grill, A.; Avouris, P.; Pfeiffer, D.; Balandin, A. A., *et al.*, Epitaxial Graphene Nanoribbon Array Fabrication Using Bcp-Assisted Nanolithography. *ACS Nano* **2012**, *6*, 6786-6792.
8. Lin, Y. M.; Valdes-Garcia, A.; Han, S. J.; Farmer, D. B.; Meric, I.; Sun, Y. N.; Wu, Y. Q.; Dimitrakopoulos, C.; Grill, A.; Avouris, P., *et al.*, Wafer-Scale Graphene Integrated Circuit. *Science* **2011**, *332*, 1294-1297.
9. Hao, Y. F.; Bharathi, M. S.; Wang, L.; Liu, Y. Y.; Chen, H.; Nie, S.; Wang, X. H.; Chou, H.; Tan, C.; Fallahzad, B., *et al.*, The Role of Surface Oxygen in the Growth of Large Single-Crystal Graphene on Copper. *Science* **2013**, *342*, 720-723.
10. Lee, J.-H.; Lee, E. K.; Joo, W.-J.; Jang, Y.; Kim, B.-S.; Lim, J. Y.; Choi, S.-H.; Ahn, S. J.; Ahn, J. R.; Park, M.-H., *et al.*, Wafer-Scale Growth of Single-Crystal Monolayer Graphene on Reusable Hydrogen-Terminated Germanium. *Science* **2014**.
11. Hwang, E. H.; Adam, S.; Das Sarma, S., Carrier Transport in Two-Dimensional Graphene Layers. *Phys. Rev. Lett.* **2007**, *98*.
12. Ando, T., Screening Effect and Impurity Scattering in Monolayer Graphene. *J Phys Soc Jpn* **2006**, *75*.
13. Nomura, K.; MacDonald, A. H., Quantum Transport of Massless Dirac Fermions. *Phys Rev Lett* **2007**, *98*.
14. Ishigami, M.; Chen, J. H.; Cullen, W. G.; Fuhrer, M. S.; Williams, E. D., Atomic Structure of Graphene on SiO₂. *Nano Lett.* **2007**, *7*, 1643-1648.
15. Katsnelson, M. I.; Geim, A. K., Electron Scattering on Microscopic Corrugations in Graphene. *Philosophical Transactions of the Royal Society of London, Series A: Mathematical, Physical Sciences and Engineering* **2008**, *366*.
16. Fratini, S.; Guinea, F., Substrate-Limited Electron Dynamics in Graphene. *Phys Rev B* **2008**, *77*.
17. Chen, J. H.; Jang, C.; Xiao, S. D.; Ishigami, M.; Fuhrer, M. S., Intrinsic and Extrinsic Performance Limits of Graphene Devices on SiO₂. *Nat. Nanotechnol.* **2008**, *3*, 206-209.
18. Bresnehan, M. S.; Hollander, M. J.; Wetherington, M.; LaBella, M.; Trumbull, K. A.; Cavalero, R.; Snyder, D. W.; Robinson, J. A., Integration of Hexagonal Boron Nitride with Quasi-Freestanding Epitaxial Graphene: Toward Wafer-Scale, High-Performance Devices. *Acs Nano* **2012**, *6*, 5234-5241.
19. Dean, C. R.; Young, A. F.; Meric, I.; Lee, C.; Wang, L.; Sorgenfrei, S.; Watanabe, K.; Taniguchi, T.; Kim, P.; Shepard, K. L., *et al.*, Boron Nitride Substrates for High-Quality Graphene Electronics. *Nat Nanotechnol* **2010**, *5*, 722-726.

20. Gannett, W.; Regan, W.; Watanabe, K.; Taniguchi, T.; Crommie, M. F.; Zettl, A., Boron Nitride Substrates for High Mobility Chemical Vapor Deposited Graphene. *Appl Phys Lett* **2011**, *98*.
21. Jain, N.; Durcan, C. A.; Jacobs-Gedrim, R.; Xu, Y.; Yu, B., Graphene Interconnects Fully Encapsulated in Layered Insulator Hexagonal Boron Nitride. *Nanotechnology* **2013**, *24*, 355202.
22. Han, G. H.; Rodriguez-Manzo, J. A.; Lee, C. W.; Kybert, N. J.; Lerner, M. B.; Qi, Z. J.; Dattoli, E. N.; Rappe, A. M.; Drndic, M.; Johnson, A. T. C., Continuous Growth of Hexagonal Graphene and Boron Nitride in-Plane Heterostructures by Atmospheric Pressure Chemical Vapor Deposition. *Acs Nano* **2013**, *7*, 10129-10138.
23. Wang, M.; Jang, S. K.; Jang, W. J.; Kim, M.; Park, S. Y.; Kim, S. W.; Kahng, S. J.; Choi, J. Y.; Ruoff, R. S.; Song, Y. J., *et al.*, A Platform for Large-Scale Graphene Electronics - Cvd Growth of Single-Layer Graphene on Cvd-Grown Hexagonal Boron Nitride. *Adv Mater* **2013**, *25*, 2746-2752.
24. Lee, K. H.; Shin, H. J.; Lee, J.; Lee, I. Y.; Kim, G. H.; Choi, J. Y.; Kim, S. W., Large-Scale Synthesis of High-Quality Hexagonal Boron Nitride Nanosheets for Large-Area Graphene Electronics. *Nano Lett* **2012**, *12*, 714-718.
25. Kim, K. K.; Hsu, A.; Jia, X. T.; Kim, S. M.; Shi, Y. M.; Dresselhaus, M.; Palacios, T.; Kong, J., Synthesis and Characterization of Hexagonal Boron Nitride Film as a Dielectric Layer for Graphene Devices. *Acs Nano* **2012**, *6*, 8583-8590.
26. Jo, I.; Pettes, M. T.; Kim, J.; Watanabe, K.; Taniguchi, T.; Yao, Z.; Shi, L., Thermal Conductivity and Phonon Transport in Suspended Few-Layer Hexagonal Boron Nitride. *Nano Lett* **2013**, *13*, 550-554.
27. Iqbal, M. W.; Iqbal, M. Z.; Jin, X.; Eom, J.; Hwang, C., Superior Characteristics of Graphene Field Effect Transistor Enclosed by Chemical-Vapor-Deposition-Grown Hexagonal Boron Nitride. *Journal of Materials Chemistry C* **2014**.
28. Gao, L. B.; Ren, W. C.; Xu, H. L.; Jin, L.; Wang, Z. X.; Ma, T.; Ma, L. P.; Zhang, Z. Y.; Fu, Q.; Peng, L. M., *et al.*, Repeated Growth and Bubbling Transfer of Graphene with Millimetre-Size Single-Crystal Grains Using Platinum. *Nat Commun* **2012**, *3*.
29. Kawamoto, N. N.; Luo, Z. T.; Kaplan, M.; Johnson, C., Synthesis of Large Area Graphene Films by Chemical Vapor Deposition. *Abstr Pap Am Chem S* **2011**, *241*.
30. Luo, Z. T.; Kim, S.; Kawamoto, N.; Rappe, A. M.; Johnson, A. T. C., Growth Mechanism of Hexagonal-Shape Graphene Flakes with Zigzag Edges. *Acs Nano* **2011**, *5*, 9154-9160.
31. Bhaviripudi, S.; Jia, X. T.; Dresselhaus, M. S.; Kong, J., Role of Kinetic Factors in Chemical Vapor Deposition Synthesis of Uniform Large Area Graphene Using Copper Catalyst. *Nano Lett* **2010**, *10*, 4128-4133.
32. Lee, G. H.; Yu, Y. J.; Cui, X.; Petrone, N.; Lee, C. H.; Choi, M. S.; Lee, D. Y.; Lee, C.; Yoo, W. J.; Watanabe, K., *et al.*, Flexible and Transparent Mos2 Field-Effect Transistors on Hexagonal Boron Nitride-Graphene Heterostructures. *Acs Nano* **2013**, *7*, 7931-7936.
33. Lauritsen, J. V.; Kibsgaard, J.; Helveg, S.; Topsoe, H.; Clausen, B. S.; Laegsgaard, E.; Besenbacher, F., Size-Dependent Structure of Mos2 Nanocrystals. *Nat Nanotechnol* **2007**, *2*, 53-58.
34. Fuchtbauer, H. G.; Tuxen, A. K.; Moses, P. G.; Topsoe, H.; Besenbacher, F.; Lauritsen, J. V., Morphology and Atomic-Scale Structure of Single-Layer Ws2 Nanoclusters. *Phys Chem Chem Phys* **2013**, *15*, 15971-15980.
35. Xia, F. N.; Perebeinos, V.; Lin, Y. M.; Wu, Y. Q.; Avouris, P., The Origins and Limits of Metal-Graphene Junction Resistance. *Nat Nanotechnol* **2011**, *6*, 179-184.
36. Sun, Z. Z.; Yan, Z.; Yao, J.; Beitler, E.; Zhu, Y.; Tour, J. M., Growth of Graphene from Solid Carbon Sources. *Nature* **2010**, *468*, 549-552.

37. Hao, Y. F.; Wang, Y. Y.; Wang, L.; Ni, Z. H.; Wang, Z. Q.; Wang, R.; Koo, C. K.; Shen, Z. X.; Thong, J. T. L., Probing Layer Number and Stacking Order of Few-Layer Graphene by Raman Spectroscopy. *Small* **2010**, *6*, 195-200.
38. Luo, Z. T.; Lu, Y.; Singer, D. W.; Berck, M. E.; Somers, L. A.; Goldsmith, B. R.; Johnson, A. T. C., Effect of Substrate Roughness and Feedstock Concentration on Growth of Wafer-Scale Graphene at Atmospheric Pressure. *Chem Mater* **2011**, *23*, 1441-1447.
39. Britnell, L.; Gorbachev, R. V.; Jalil, R.; Belle, B. D.; Schedin, F.; Katsnelson, M. I.; Eaves, L.; Morozov, S. V.; Mayorov, A. S.; Peres, N. M. R., *et al.*, Electron Tunneling through Ultrathin Boron Nitride Crystalline Barriers. *Nano Lett* **2012**, *12*, 1707-1710.
40. Wang, L.; Meric, I.; Huang, P. Y.; Gao, Q.; Gao, Y.; Tran, H.; Taniguchi, T.; Watanabe, K.; Campos, L. M.; Muller, D. A., *et al.*, One-Dimensional Electrical Contact to a Two-Dimensional Material. *Science* **2013**, *342*, 614-617.
41. Lu, Y.; Merchant, C. A.; Drndic, M.; Johnson, A. T. C., In Situ Electronic Characterization of Graphene Nanoconstrictions Fabricated in a Transmission Electron Microscope. *Nano Lett.* **2011**, *11*, 5184-5188.
42. Chaudhry, A., Interconnects for Nanoscale Mosfet Technology: A Review. *Journal of Semiconductors* **2013**, *34*, 066001.
43. Liao, A. D.; Wu, J. Z.; Wang, X. R.; Tahy, K.; Jena, D.; Dai, H. J.; Pop, E., Thermally Limited Current Carrying Ability of Graphene Nanoribbons. *Phys Rev Lett* **2011**, *106*.
44. Moser, J.; Barreiro, A.; Bachtold, A., Current-Induced Cleaning of Graphene. *Appl Phys Lett* **2007**, *91*.
45. Novoselov, K. S.; Geim, A. K.; Morozov, S. V.; Jiang, D.; Zhang, Y.; Dubonos, S. V.; Grigorieva, I. V.; Firsov, A. A., Electric Field Effect in Atomically Thin Carbon Films. *Science* **2004**, *306*, 666-669.
46. Liu, L.; Ryu, S. M.; Tomasik, M. R.; Stolyarova, E.; Jung, N.; Hybertsen, M. S.; Steigerwald, M. L.; Brus, L. E.; Flynn, G. W., Graphene Oxidation: Thickness-Dependent Etching and Strong Chemical Doping. *Nano Lett.* **2008**, *8*, 1965-1970.
47. Dan, Y. P.; Lu, Y.; Kybert, N. J.; Luo, Z. T.; Johnson, A. T. C., Intrinsic Response of Graphene Vapor Sensors. *Nano Lett* **2009**, *9*, 1472-1475.
48. Westenfelder, B.; Meyer, J. C.; Biskupek, J.; Kurasch, S.; Scholz, F.; Krill, C. E.; Kaiser, U., Transformations of Carbon Adsorbates on Graphene Substrates under Extreme Heat. *Nano Lett.* **2011**, *11*, 5123-5127.
49. Barreiro, A.; Bornert, F.; Rummeli, M. H.; Buchner, B.; Vandersypen, L. M. K., Graphene at High Bias: Cracking, Layer by Layer Sublimation, and Fusing. *Nano Lett.* **2012**, *12*, 1873-1878.
50. Liao, A.; Alizadegan, R.; Ong, Z. Y.; Dutta, S.; Xiong, F.; Hsia, K. J.; Pop, E., Thermal Dissipation and Variability in Electrical Breakdown of Carbon Nanotube Devices. *Phys Rev B* **2010**, *82*.
51. Murali, R.; Yang, Y. X.; Brenner, K.; Beck, T.; Meindl, J. D., Breakdown Current Density of Graphene Nanoribbons. *Appl Phys Lett* **2009**, *94*.
52. Liao, L.; Bai, J. W.; Qu, Y. Q.; Lin, Y. C.; Li, Y. J.; Huang, Y.; Duan, X. F., High-Kappa Oxide Nanoribbons as Gate Dielectrics for High Mobility Top-Gated Graphene Transistors. *Proc. Natl. Acad. Sci. U. S. A.* **2010**, *107*, 6711-6715.