

Supporting Information

Ultrasensitive Label-Free MiRNA Sensing Based on Flexible Graphene Field-Effect Transistor Without Functionalization

Jianwei Gao^a, Yakun Gao^a, Yingkuan Han^{a,b}, Jinbo Pang^c, Chao Wang^a, Yanhao Wang^a, Hong Liu^{c,d}, Yu Zhang^{a,*}, Lin Han^{a,*}

^a Institute of Marine Science and Technology, Shandong University, Qingdao 266237, China; gaoxiii@foxmail.com; 17854262215@163.com; 980606701@qq.com; wangyanhaooo@126.com

^b School of Microelectronics, Shandong University, Jinan 250010, China; yingkuan.han@foxmail.com

^c Institute for Advanced Interdisciplinary Research (iAIR), University of Jinan, Jinan 250022, China; jinbo.pang@hotmail.com

^d State Key Laboratory of Crystal Materials, Shandong University, Jinan, Shandong, 250100, China; hongliu@sdu.edu.cn

*Corresponding authors email: yuzhang@sdu.edu.cn; hanlin@sdu.edu.cn

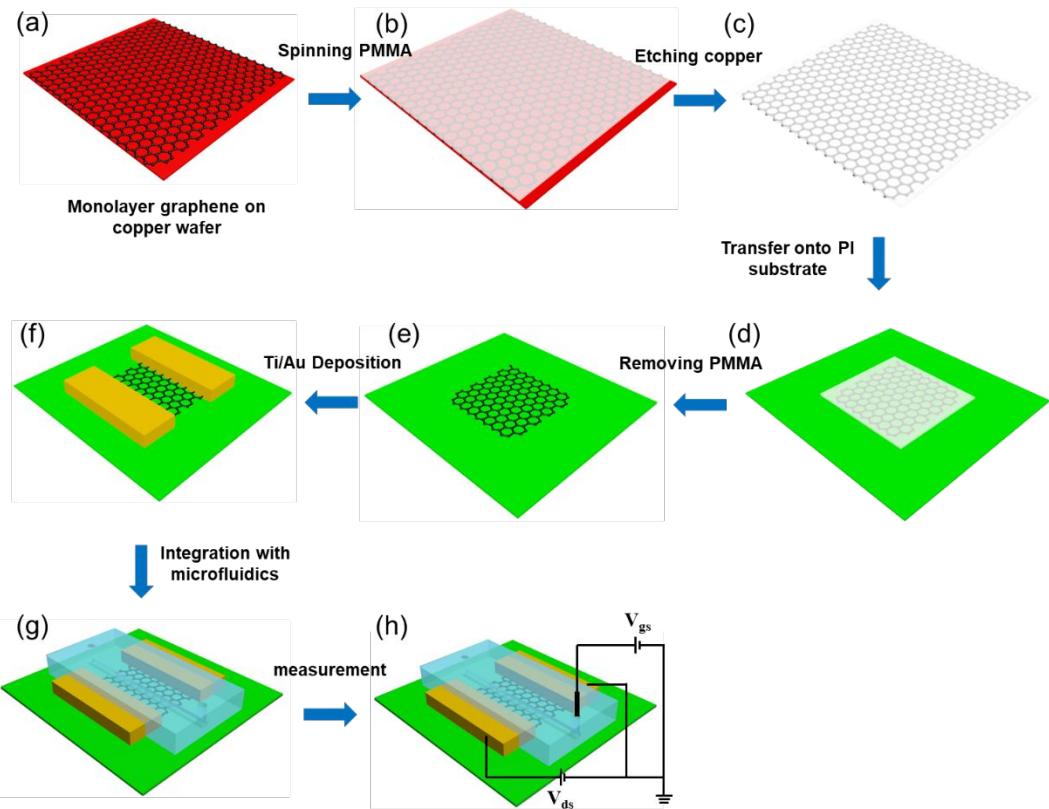


Figure S1. Fabrication process of graphene-field effect transistor biosensor.

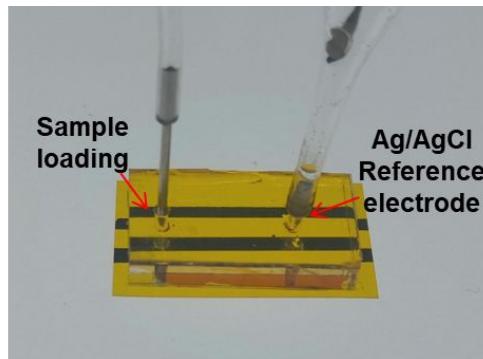


Figure S2. A photo of graphene transistor biosensor on PI substrate integrated with microfluidic.

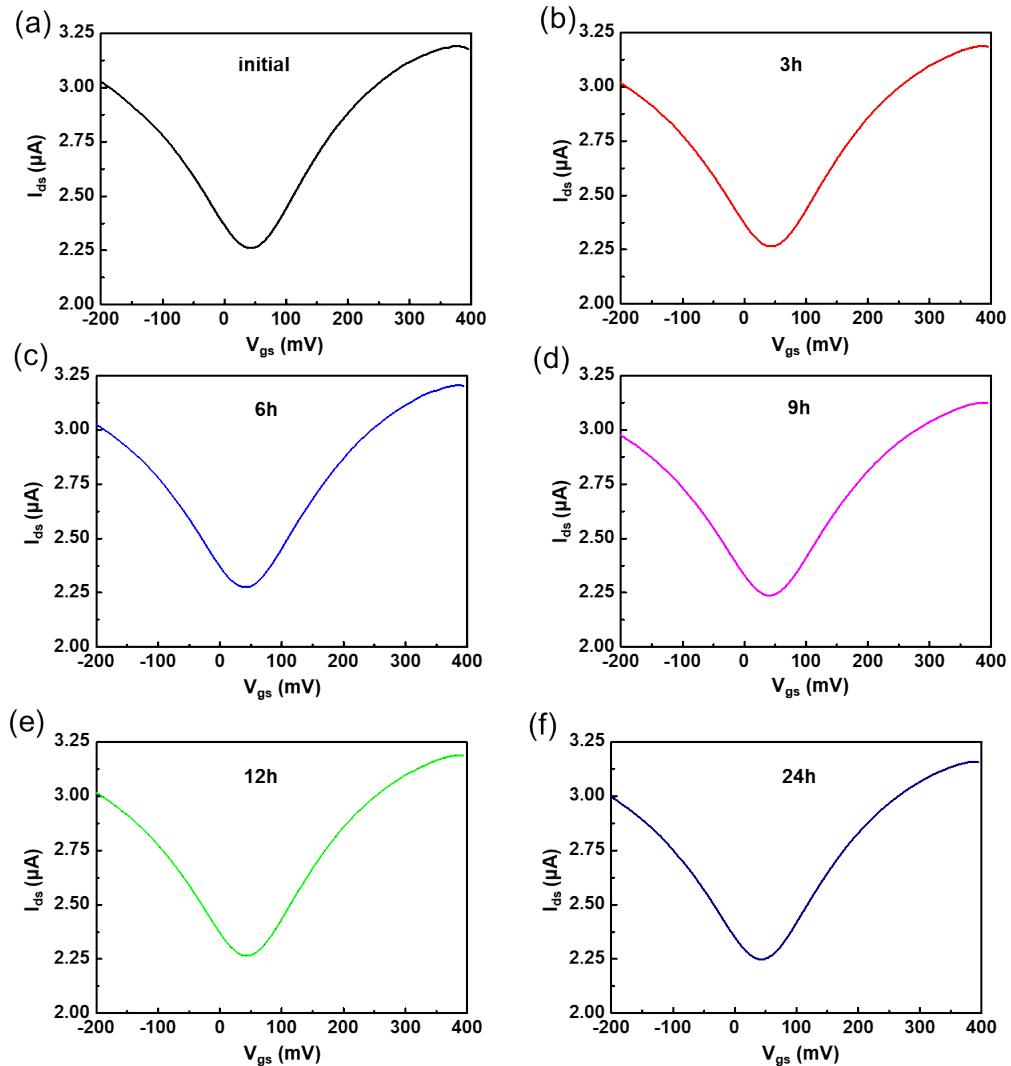


Figure S3. Transfer characteristics of a Gr-FET measured in PBS at (a) 0, (b) 3 hour, (c) 6 hour, (d) 9 hour, (e) 12 hour, (f) 24 hour, $V_{ds} = 50$ mV.

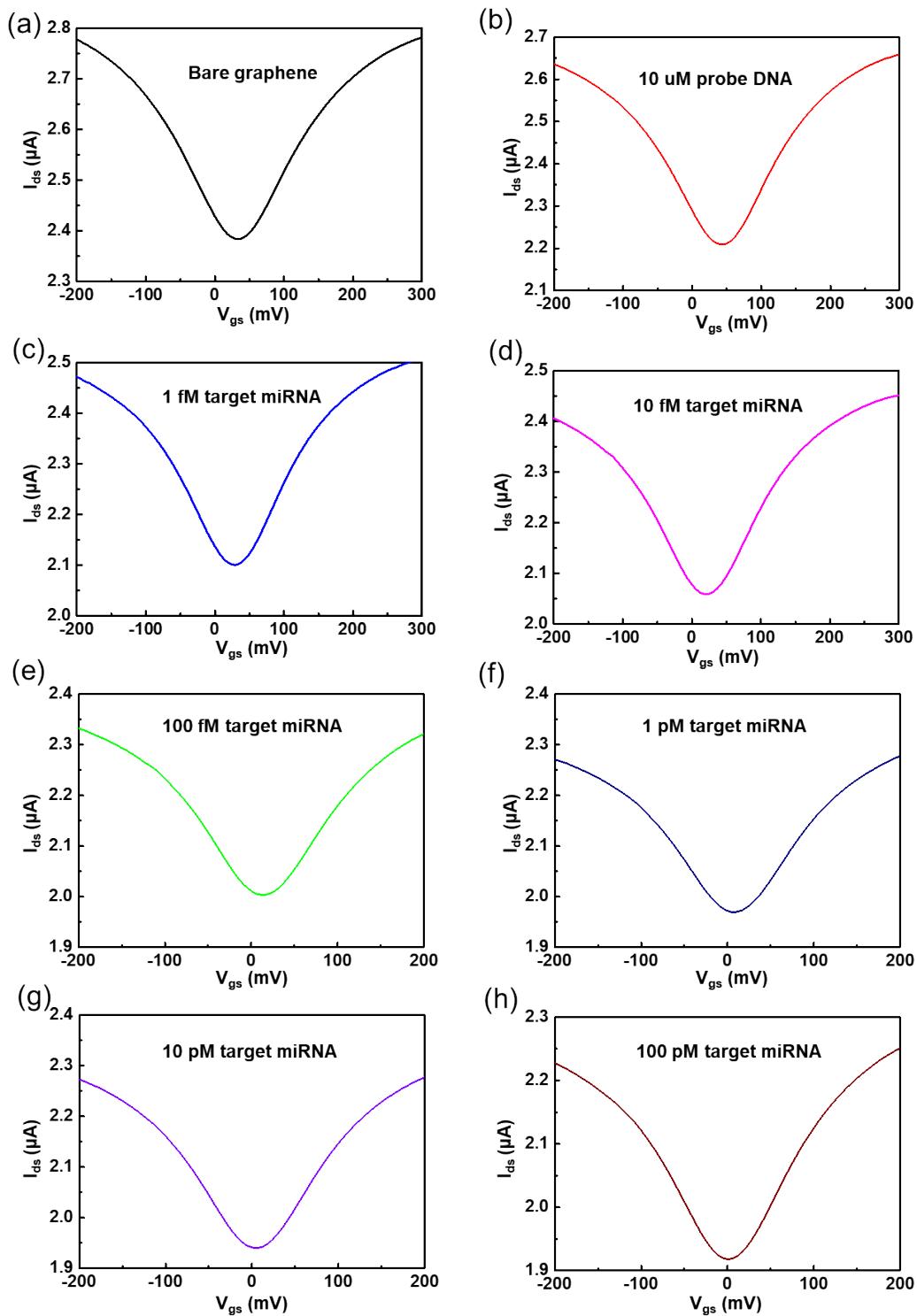


Figure S4. Transfer characteristics of as-fabricated Gr-FET biosensor responding to gate solutions with different target molecules (a) PBS, (b) 10 uM probe DNA, (c) 1 fM target miRNA, (d) 10 fM, (e) 100 fM (f) 1 pM (g) 10 pM (h) 100 pM before bending

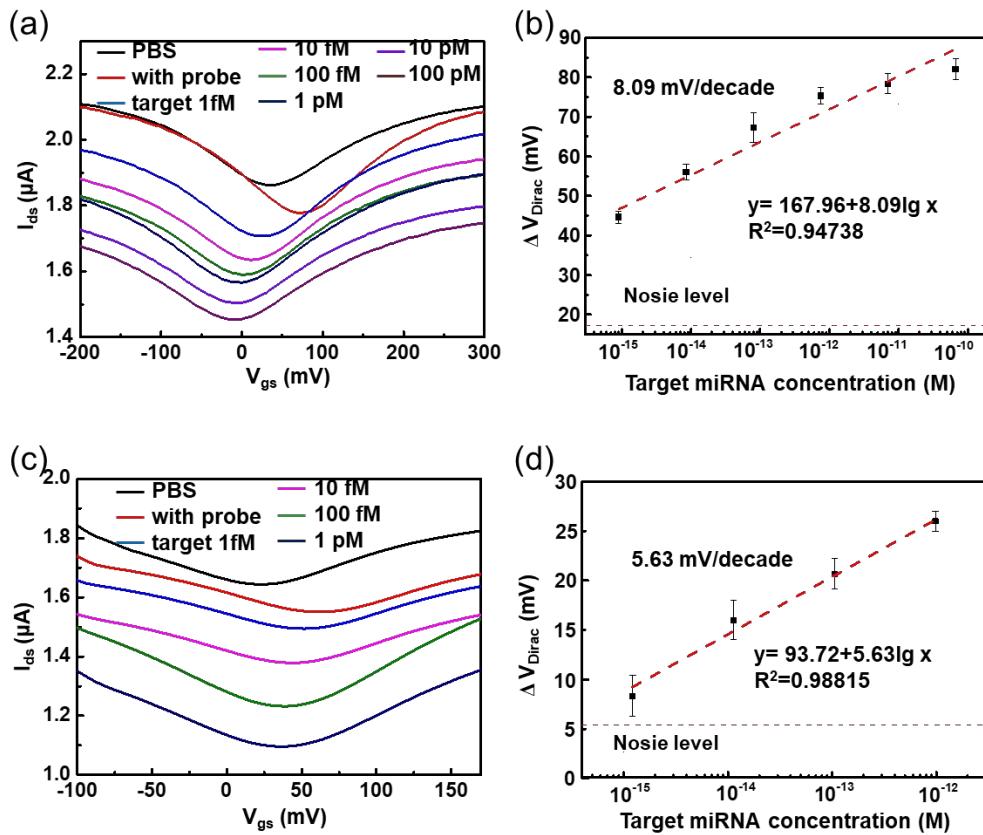


Figure S5. Properties of miRNA sensors in different fabrication batches. (a) Transfer characteristics of as-fabricated Gr-FET biosensor responding to gate solutions with different target molecules. (b) ΔV_{Dirac} dependence of the concentration of target miRNA corresponding to as-fabricated biosensor, and dashed line represents the noise level (~ 17 mV) from the blank control test; (c) Transfer characteristics of Gr-FET biosensor responding to gate solutions with different target molecules. (d) ΔV_{Dirac} dependence of the concentration of target miRNA corresponding to as-fabricated biosensor, and dashed line represents the noise level (~ 6 mV) from the blank control test

Table S1. Performance and Structure of Graphene-Based Solution-Gated FET DNA/miRNA Biosensor

FET channel		Linker	Target	Gate electrode	Reaction solution	DNA immobilization time	Detection limit (M)	Reference
Material	Substrate							
Graphene	Si/SiO ₂	None	DNA	Ag	1× PBS	4h	1 × 10 ⁻¹²	1
Graphene	Si/SiO ₂	None	DNA	Ag	PBS ^①	16h	1 × 10 ⁻¹¹	2
Graphene	Si/SiO ₂	PBASE	DNA	Pt	0.01× PBS	2h	1 × 10 ⁻¹¹	3
Graphene	sapphire	PBASE	DNA	Ag/AgCl	0.01× PBS	2h	1 × 10 ⁻¹³	4
Graphene	Si/SiO ₂	PBASE	DNA	Au-planar, integrated	PBS ^②	14h	1 × 10 ⁻¹⁷	5
rGO	Si/SiO ₂	Au-S	miRNA	Ag	1× PBS	3h	1 × 10 ⁻¹⁴	6
Graphene	Si/SiO ₂	PBASE	DNA	Ag	1× PBS	2h	1 × 10 ⁻¹⁴	7
Graphene	glass	Au-S	DNA	Au	1× PBS	12h	1 × 10 ⁻¹⁵	8
Graphene	PI	Poly A	miRNA	Ag/AgCl	1× PBS	30min	1 × 10 ⁻¹⁴	This work

Note: ① 0.25 M NaCl and 10 mM PBS; ② 10 mM PBS, 150 mM NaCl and 50 mM MgCl₂

REFERENCE

- (1) Chen, T. Y.; Loan, P. T. K.; Hsu, C. L.; Lee, Y. H.; Wang, J. T. W.; Wei, K. H.; Li, L. J. Label-free detection of DNA hybridization using transistors based on CVD grown graphene. *Biosensors and Bioelectronics*, 2013, 41, 103-109.
- (2) Dong, X.; Shi, Y.; Huang, W.; Chen, P.; Li, L. J. Electrical detection of DNA hybridization with single-base specificity using transistors based on CVD-grown graphene sheets. *Advanced Materials*, 2010, 22(14), 1649-1653.
- (3) Xu, S.; Zhan, J.; Man, B.; Jiang, S.; Yue, W.; Gao, S.. Zhou, Y. Real-time reliable determination of binding kinetics of DNA hybridization using a multi-channel graphene biosensor. *Nature communications*, 2017, 8, 14902.
- (4) Xu, S.; Jiang, S.; Zhang, C.; Yue, W.; Zou, Y.; Wang, G.; Wang, J. Ultrasensitive label-free detection of DNA hybridization by sapphire-based graphene field-effect transistor biosensor. *Applied Surface Science*, 2018, 427, 1114-1119.
- (5) Campos, R.; Borme, J.; Guerreiro, J. R.; Machado Jr, G.; Cerqueira, M. F.; Petrovykh, D. Y.; Alpuim, P. Attomolar label-free detection of DNA hybridization with electrolyte-gated graphene field-effect transistors. *ACS sensors*, 2019, 4(2), 286-293.
- (6) Cai, B.; Huang, L.; Zhang, H.; Sun, Z.; Zhang, Z.; Zhang, G. J. Gold nanoparticles-decorated graphene field-effect transistor biosensor for femtomolar MicroRNA detection. *Biosensors and Bioelectronics*, 2015, 74, 329-334.
- (7) Zheng, C.; Huang, L.; Zhang, H.; Sun, Z.; Zhang, Z.; Zhang, G. J. Fabrication of ultrasensitive field-effect transistor DNA biosensors by a directional transfer technique based on CVD-grown graphene. *ACS applied materials interfaces*, 2015, 7(31), 16953-16959.
- (8) Li, S.; Huang, K.; Fan, Q.; Yang, S.; Shen, T.; Mei, T. Li, J. Highly sensitive solution-gated graphene transistors for label-free DNA detection. *Biosensors and Bioelectronics*, 2019, 136, 91-96..