

Supporting Information

Self-assembly graphene nanomask for epitaxial growth of nonplanar and planar GaN

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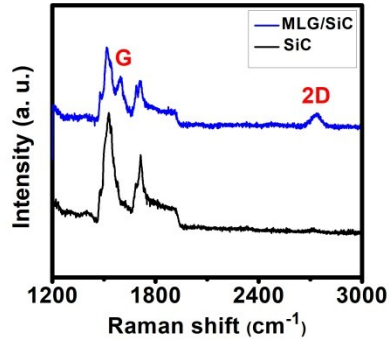


Figure S1. Raman spectrum of the MLG/SiC and pure SiC substrate at the wave number of 1200 to 3000 cm^{-1} . Clear G and 2D bands of MLG and secondary bands of SiC can be seen.

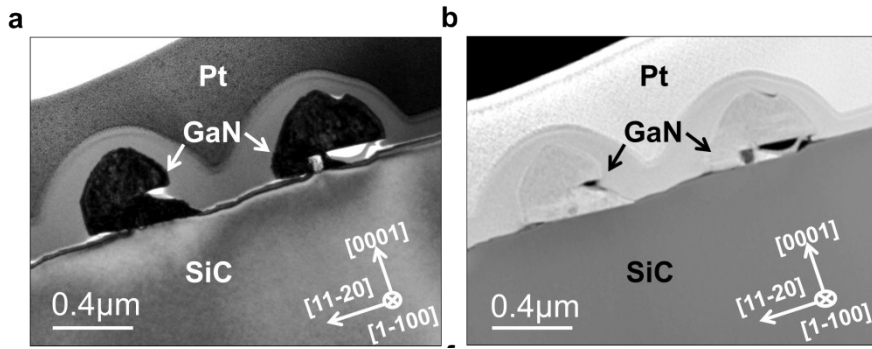


Figure S2. (a) TEM and (b) HAADF images of GaN nanowalls SNSAG on MLG/SiC.

Table S1. The difference of diffraction spots distance of GaN and SiC. (a and c: the lattice parameters; hkl: crystal indices; d: interplanar crystal spacing). The small $\Delta 1/d$ results the SAED patterns of GaN and SiC are overlap.

	a [nm]	c [nm]	(hkl)	d [nm]	1/d [1/nm]	$\Delta 1/d$ [1/nm]
GaN	0.3189	0.5185	(002)	0.259	3.857	0.122
			(-210)	0.159	6.272	0.237
SiC	0.3073	1.0053	(004)	0.251	3.979	
			(-210)	0.154	6.508	

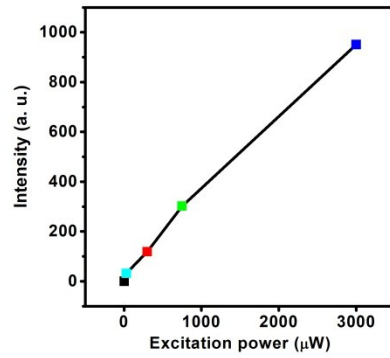


Figure S3. Plot of integrated intensity of the emission as a function of the excitation power.

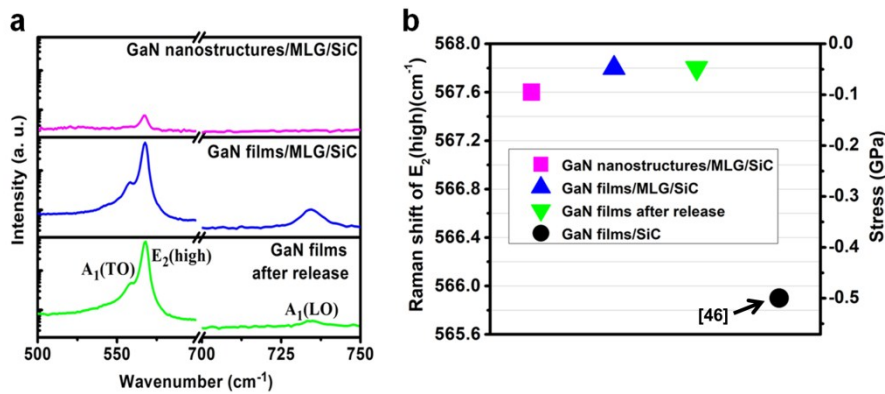


Figure S4. (a) Raman spectra of the GaN nanostructures/MLG/SiC, GaN films/MLG/SiC, and GaN films after release at the wavenumber of 500 to 750 nm. (b) Raman shift of E_2 (high) and stress of GaN. Black circle correspond to 1.65 μm -thick GaN films grown directly on 4H-SiC with similar miscut angle of 3.4° ,¹ the stress was -0.5 GPa (10 times than that of GaN/MLG/SiC), and there were a lot of cracks formed in GaN/SiC.

Table S2. Stress of GaN nanostructures and films grown on MLG/SiC and after release.

Sample	E_2 (high) [nm]	Stress ^a [GPa]
GaN nanostructures/MLG/SiC	567.6	-0.10
GaN films/MLG/SiC	567.8	-0.05
GaN films after release	567.8	-0.05

^{a)} The stress is calculated by the formula as follow:

$$\sigma = \frac{\Delta\omega}{K} (\text{cm}^{-1} \text{GPa}^{-1})$$

where σ is the biaxial stress, $\Delta\omega$ is the E_2 frequency shift and K is the Raman stress factor. The unstressed GaN E_2 (high) at 568.0 cm^{-1} , $K=4.2 \text{ cm}^{-1} \text{GPa}^{-1}$.²

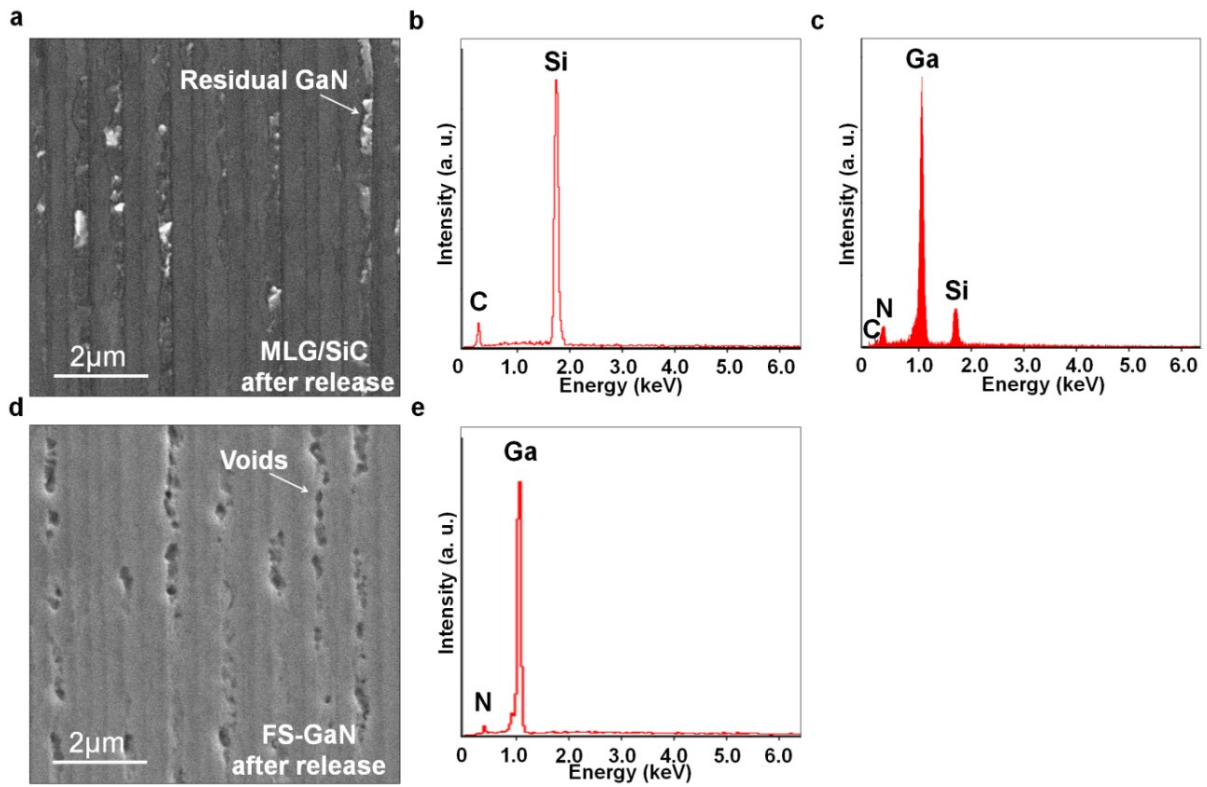


Figure S5. (a) Surface morphology of MLG/SiC after GaN films release. (b) and (c) EDX spectra of MLG/SiC and residual GaN on MLG/SiC after release. (d) Surface morphology of the back surface of FS-GaN after release. (e) EDX spectra of the voids on the surface of GaN microrod. It indicates that the GaN films are SNSAG on MLG/SiC.

Table S3. The crystalline quality of GaN films vdWE/RE/SAG/SNSAG on graphene.

Structure	Growth type	Thickness (μm)	Crystalline	FWHM(0002) (arcsec)	TDDs ($\times 10^8 \text{cm}^{-2}$)	Method	Ref.
GaN/ZnO/graphene/SiO ₂ /Si	vdWE	~5	Grain boundary	-	12~24	TEM	14
GaN/ZnO/graphene/SiO ₂ /Si	vdWE	~5	Grain boundary	-	13	TEM	15
GaN/graphene/SiO ₂ /Si	vdWE	10	Grain boundary	-	8-36	TEM	16
GaN/graphene/SiC	RE	2.5	Single crystalline	222	10	TEM	27
GaN/graphene/SiC	SAG	1.5	Inversion domain	-	30	TEM	28
GaN/graphene/GaN/sapphire	SAG	40	Single-crystalline	154	0.18	EPD	29
GaN/graphene/GaN/sapphire	SAG	20	Single-crystalline	276	1.7~1.8	CL	30
GaN/graphene/SiC	SNSAG	150	Single-crystalline	43	0.059	CL	This work

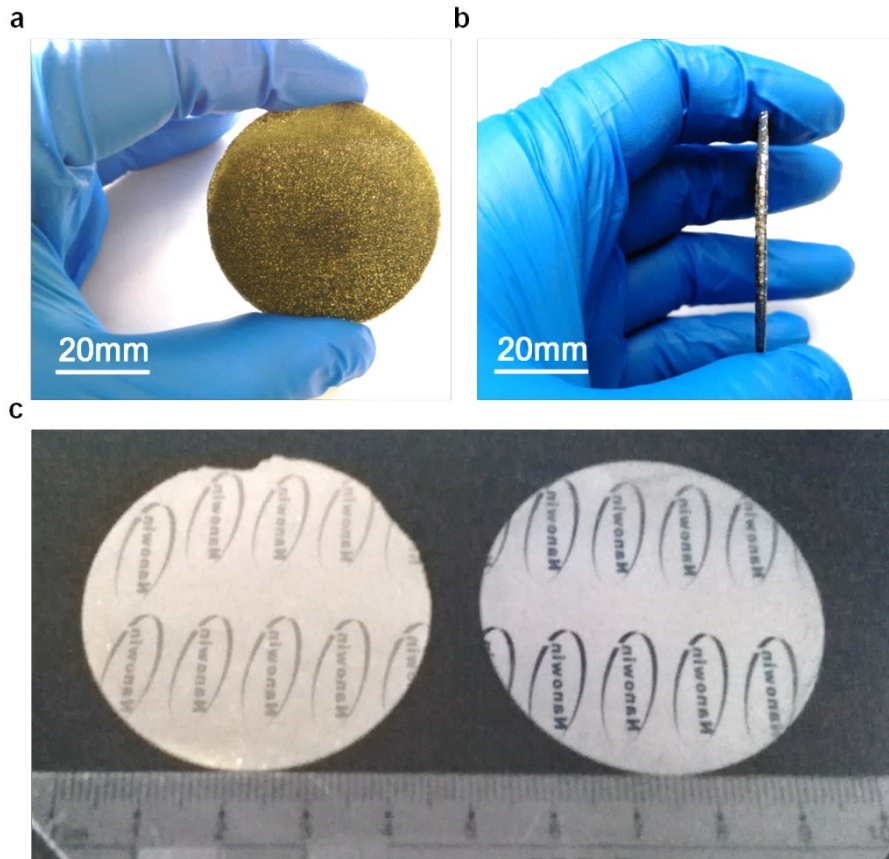


Figure S6. (a) Photograph of crack-free, 1.2 mm-thick GaN films SNSAG on MLG/SiC. (b) Cross-section photograph of the GaN films SNSAG on MLG/SiC. (c) Photograph of crack-free, mechanical exfoliated FS-GaN (left) after single side chemical mechanical polishing and the corresponding MLG/SiC substrate (right). Mirror reflection of Nanowin logo which was set above the samples when taking photo. The bow of the substrate was about $19\ \mu\text{m}$ (radius of bowing is $17\ \text{m}$), which is lower than the one fabricated by laser lift-off (radius of bowing was $4.6\ \text{m}$ and bow was about $70\ \mu\text{m}$).³ Such low bow value may be caused by nearly-free stress of GaN films after growth.

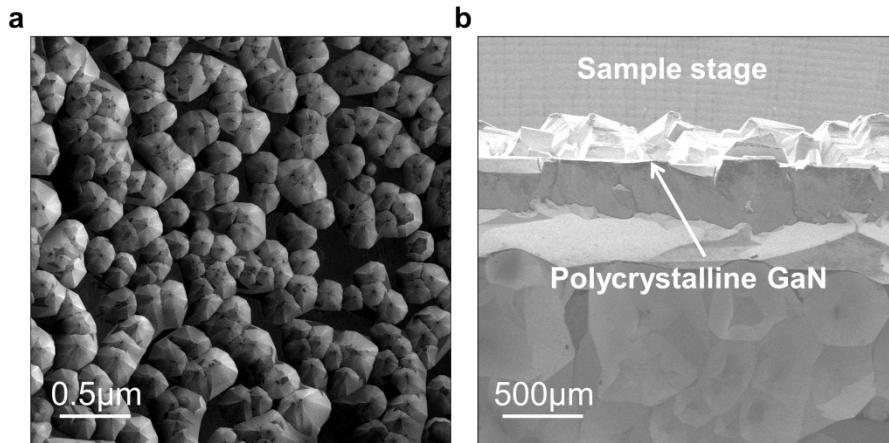


Figure S7. (a) SEM image of surface morphology of the GaN films SNSAG on MLG/SiC. (b) SEM image of the edge morphology of GaN films SNSAG on MLG/SiC. The surface of GaN films is not smooth as the surface deterioration of GaN films when thickness is more than 1 mm. In addition, because of polycrystalline GaN exists at the edges of substrate, directly mechanical exfoliation of GaN can not obtain FS-GaN without cracks. Therefore, polycrystalline GaN should be eliminated by cylindrical grinding machine, and then the GaN films can be mechanically exfoliated without any stressor layer.

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