

Table S1. XRD-derived phase contents, lattice parameters, and average crystallite sizes. MG – manual grinding, SBM – short ball milling, LBM – long ball milling. *n/d* – not determined.

Powder (temperature, time/grinding)	Phase, content [%]	Lattice parameters <i>a</i> [Å], <i>c</i> [Å]	Av. crystallite size [nm]
650 °C, 36 h/LBM	h-GaN, 100	<i>a</i> =3.18, <i>c</i> =5.19	12
700 °C, 90 h/MG	h-GaN (small), 50	<i>a</i> =3.17, <i>c</i> =5.19	4
	h-GaN (large), 33	<i>a</i> =3.20, <i>c</i> =5.23	21
	c-GaN, 14	<i>a</i> =4.52	57
	<i>c-GaAs</i> , 3	<i>n/d</i>	<i>n/d</i>
700 °C, 90 h/SBM	h-GaN, 99.8	<i>a</i> =3.19, <i>c</i> =5.21	16
	<i>c-GaAs</i> , 0.2	<i>n/d</i>	<i>n/d</i>
700 °C, 90 h/LBM	h-GaN, 100	<i>a</i> =3.19, <i>c</i> =5.20	15
800 °C, 90 h/MG	h-GaN (small), 32	<i>a</i> =3.16, <i>c</i> =5.22	3
	h-GaN (large), 25	<i>a</i> =3.19, <i>c</i> =5.19	25
	c-GaN, 42.5	<i>a</i> =4.51	40
	<i>c-GaAs</i> , 0.5	<i>n/d</i>	<i>n/d</i>
800 °C, 90 h/SBM	h-GaN, 71	<i>a</i> =3.19, <i>c</i> =5.22	13
	c-GaN, 29	<i>a</i> =4.51	15
800 °C, 90 h/LBM	h-GaN, 62	<i>a</i> =3.20, <i>c</i> =5.20	14
	c-GaN, 38	<i>a</i> =4.51	16
900 °C, 6 h/MG	h-GaN, 33.5	<i>a</i> =3.18, <i>c</i> =5.29	26
	c-GaN, 65	<i>a</i> =4.51	55
	<i>c-GaAs</i> , 1.5	<i>n/d</i>	<i>n/d</i>
900 °C, 6 h/SBM	h-GaN, 68	<i>a</i> =3.19, <i>c</i> =5.21	16
	c-GaN, 32	<i>a</i> =4.51	23
900 °C, 6 h/LBM	h-GaN, 72	<i>a</i> =3.19, <i>c</i> =5.21	17
	c-GaN, 28	<i>a</i> =4.51	16
900 °C, 6 h/bulk wafer	h-GaN, 76	<i>a</i> =3.19, <i>c</i> =5.20	30
	c-GaN, 24	<i>a</i> =4.51	33
950 °C, 6 h/bulk wafer	h-GaN, 100	<i>a</i> =3.18, <i>c</i> =5.18	22

Table S2. Positions of deconvoluted peaks in micro-Raman determinations for GaAs substrates and nitrided products. MG – manual grinding, LBM – long ball milling, am – amorphous, cryst – crystalline, br – broad. References in square brackets are listed as such in the text.

shift [cm ⁻¹]/mode (assignment) [Ref.]	Raman shift [cm ⁻¹]							
	GaAs_MG	GaAs_LBM	700_MG	700_LBM	800_MG	800_LBM	900_MG	900_LBM
150/LA (am GaAs) [20a]; 144/E ₂ (low) (h-GaN) [20g]	156	ca.160	151			161		
210-240, br (am As); 198 (cryst As) [20b]; 205, br (am As) [20c]; <i>ca.</i> 200/DALA in GaAsN [20d]	197	195, 222	204, 213	197, 221	202, 221	196, 216	199, 226	200, 224
231-245/ TO, br (am GaAs) [20a,20c]; 255 (cryst As) [20b]; 246 (Ga metal) [20l]	239	243	238					
264-265/TO (cryst GaAs) [20c]	264	261	255	249		248	249	250
285-292/LO (cryst GaAs) [20a,20c]; red shift in GaAsN [20e]	285	283	279					
2 nd order GaAsN [20e]; B ₁ (low)silent (h- GaN) [20i]; nitrogen vacancies (h-GaN) [20j]			299, 315	297	304	296	292	299
nitrogen vacancies (h- GaN) [20j], octahedrally bonded Ga [20k]			411	411	417	410	412	415
480/2 nd order in GaAs[20a] or 470 in GaAsN [20e]; 467- 475/N-related LVM [20f]			480			460		513
554-558/E ₁ (TO) (h- GaN); 552/TO (c- GaN) [20h]; 532/A ₁ (TO) (h-GaN); 559/E ₁ (TO) (h-GaN); 568/E ₂ (high) (h-GaN); 555/TO (c-GaN) [20g]			550	558	555, 561	544	541	560
B ₁ (low)silent (h-GaN) [20i]; nitrogen vacancies (h-GaN) [20j]			601, 663	689	612, 684	602, 652	599, 668	607, 645
735/A ₁ (LO) (h-GaN), 739/LO (c-GaN) [20h]; 734/A ₁ (LO) (h- GaN); 741/E ₁ (LO) (h- GaN) [20g]			700, 756	713	718, 756	708, 754	704, 755	714, 757