SUPPLEMENTARY INFORMATION FOR

Interface energies of Ga_2O_3 phases with the sapphire substrate and the phase-locked epitaxy of metastable structures explained

Ilaria Bertoni*[a], Aldo Ugolotti[a], Emilio Scalise[a], Roberto Bergamaschini[a], and Leo Miglio[a]

[a] *Department of Materials Science, University of Milano-Bicocca, via Cozzi 55, 20125 Milan (Italy)*

phase	a[Å]	b[Å]	c[Å]
$\kappa - Ga_2O_3$	5.061	8.686	9.292
$\beta - Ga_2O_3$	14.763	3.048	5.809
$\alpha - Ga_2O_3$	5.001	5.001	13.448
$\alpha - Al_2O_3$	4.775	4.775	13.015

Table S1: Lattice parameters of optimized bulk cells, oriented according to the substrate. The slabs of the film were built in order to align the β [102] along the α [100], the β [010] along the α [120], the κ [100] along the α [100] and the κ [010] along the α [120].

		m_x / m_y misfit strain		
phase	x/y axis	vs $\alpha - Al_2O_3$	vs $\alpha - Ga_2O_3$	vs $\beta - Ga_2O_3$
$\kappa - Ga_2O_3$	[100]/[010]	4.8 % / 5.7 %	0.3 % / -1.2 %	2.8 % / -5.3 %
$\beta - Ga_2O_3$	[102]/[010]	9.5 % / 3.0 %	5.3 % / -1.6 %	-
$\alpha - Ga_2O_3$	[100]/[120]	4.5 % / 4.5 %	-	-
$\alpha - Al_2O_3$	[100]/[120]	-	-	-
		epitaxial relationship: $n_x \times n_y$ (film): $n_x \times n_y$ (substrate)		
phase	in-plane cell	vs $\alpha - Al_2O_3$	vs $\alpha - Ga_2O_3$	vs $\beta - Ga_2O_3$
$\kappa - Ga_2O_3$	conventional-rectangular	1×1:1×1	1×1:1×1	1×1:1×1
$\beta - Ga_2O_3$	conventional-rectangular	1×3:3×1	1×3:3×1	1×3:3×1
$\alpha - Ga_2O_3$	primitive-hexagonal	1×1:1×1	1×1:1×1	1×1:1×1

Table S2: Misfit strain with the substrate, calculated as: $m_i=(a_i^{film} - a_i^{substr})/a_i^{film}$. a_i is the lattice parameter of the film/substrate along the given direction.

phase	$\Delta\mu_\varepsilon$ $[meV/f.u.]$	γ_{epi} [meV/f.u.]	v [Å^3/$f.u.$]	h [Å]
on $\alpha - Al_2O_3$ substrate				
α	309	88	45.8	2.32
β	540	57	48.6	4.93
κ	314	86	48.1	4.88
on $\alpha - Ga_2O_3^{\varepsilon}$ substrate				
α	309	88	45.8	2.32
β	540	57	48.6	4.93
κ	314	86	48.1	4.88
on $\alpha - Ga_2O_3$ substrate				
α	0	70	48.5	2.24
β	150	53	51.3	4.74
κ	10	53	50.8	4.69
$\alpha - Al_2O_3$: $\gamma_{epi} = 113 meV/Å^2$				

Table S3: Elastic energy, surface energy of the epilayer, atomic volume of the cell and thickness of the layer for the different phases, given $\alpha - Al_2O_3$ substrate, $\alpha - Ga_2O_3$ strained interlayer and Ga_2O_3 fully relaxed interlayer.

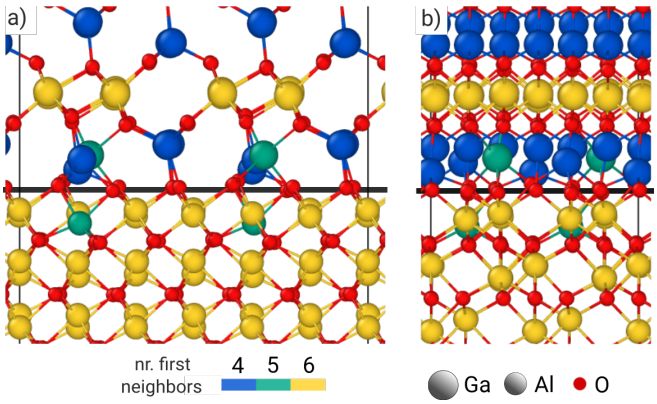

Figure S1: Front (a) and side (b) view of the $\beta - Ga_2O_3/\alpha - Al_2O_3$ interface nr. 2. The black line marks the plane of O atoms shared by both film and substrate.

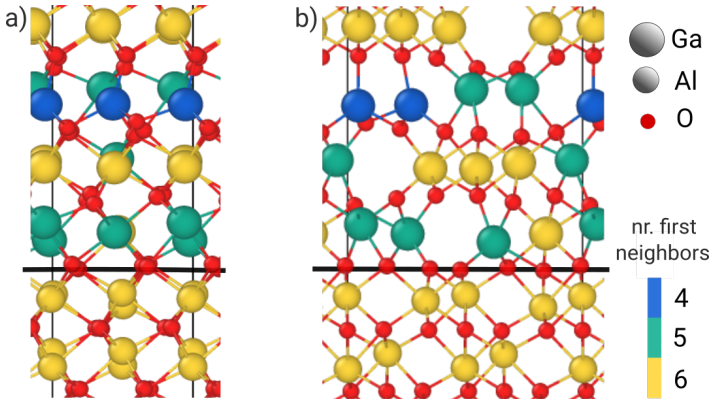

Figure S2: Front (a) and side (b) view of the $\kappa - Ga_2O_3/\alpha - Al_2O_3$ interface nr. 2. The black line marks the plane of O atoms shared by both film and substrate.

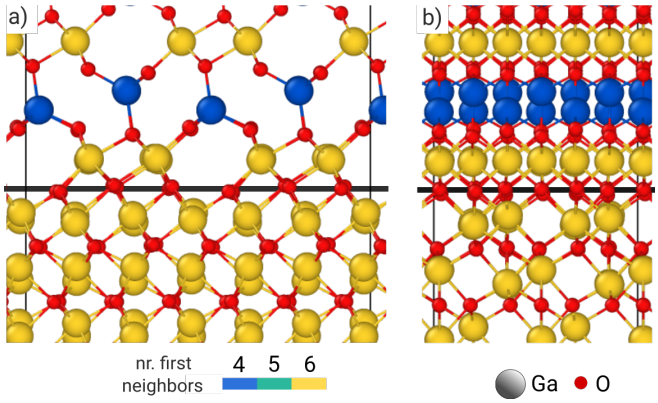

Figure S3: Front (a) and side (b) view of the $\beta - Ga_2O_3/\alpha - Ga_2O_3$ interface. The $\beta - Ga_2O_3$ interlayer is fully strained on $\alpha - Al_2O_3$ substrate. The black line marks the plane of O atoms shared by both film and substrate.

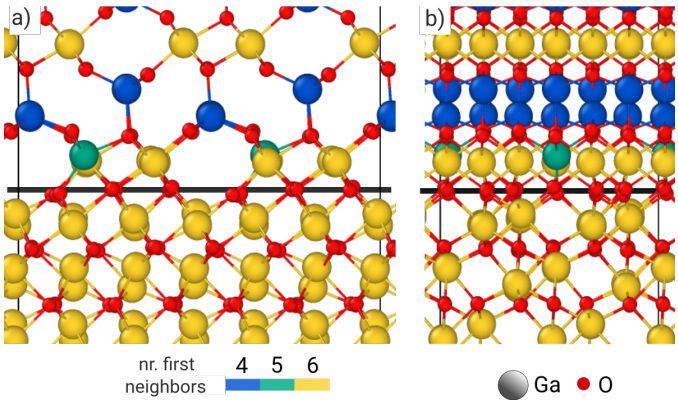

Figure S4: Front (a) and side (b) view of the $\beta - Ga_2O_3/\alpha - Ga_2O_3$ interface. The $\beta - Ga_2O_3$ interlayer is fully relaxed. The black line marks the plane of O atoms shared by both film and substrate.

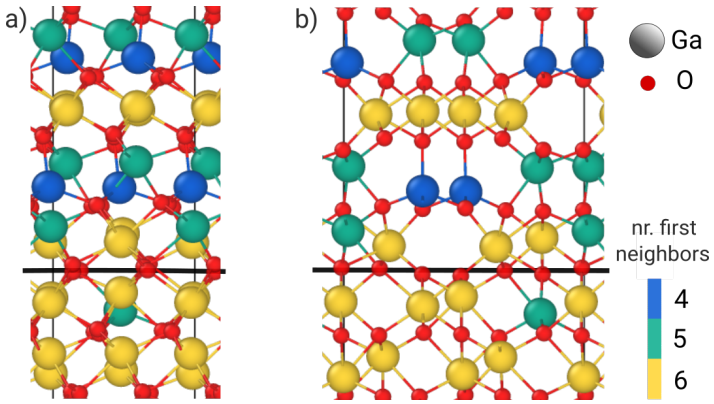

Figure S5: Front (a) and side (b) view of the $\kappa - Ga_2O_3/\alpha - Al_2O_3$ interface. The $\alpha - Ga_2O_3$ interlayer is fully strained on $\alpha - Al_2O_3$ substrate. The black line marks the plane of O atoms shared by both film and substrate.

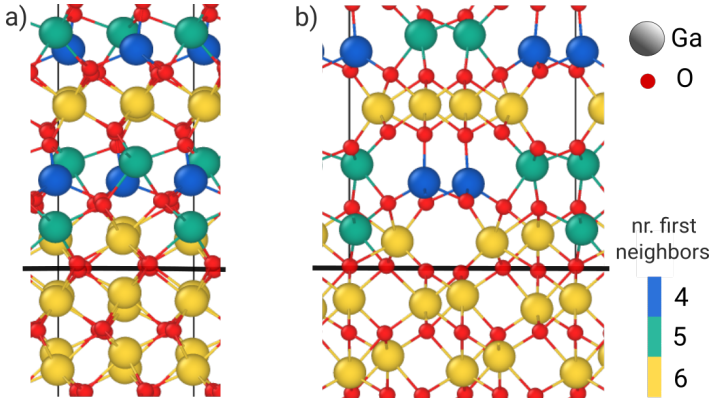

Figure S6: Front (a) and side (b) view of the $\kappa - Ga_2O_3/\alpha - Al_2O_3$ interface. The $\alpha - Ga_2O_3$ interlayer is fully relaxed. The black line marks the plane of O atoms shared by both film and substrate.

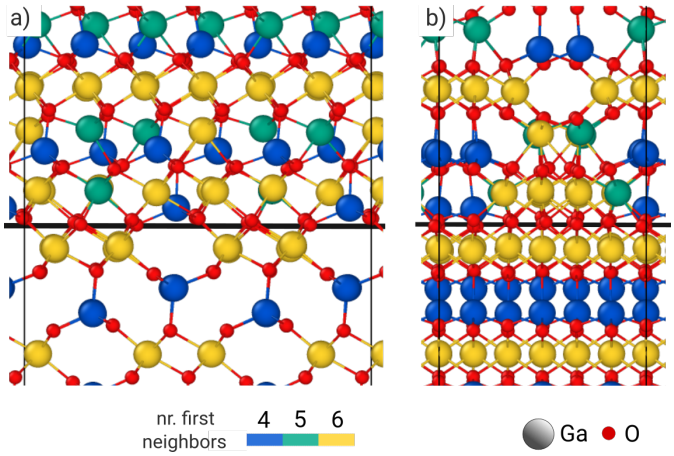

Figure S7: Front (a) and side (b) view of the $\kappa - Ga_2O_3/\beta - Ga_2O_3$ interface. The $\beta - Ga_2O_3$ interlayer is fully relaxed. The black line marks the plane of O atoms shared by both film and substrate.

The files of the optimized structures reported in the main manuscript and in the Supplementary Information are collected in the archive supplementary_information_geometries.zip