Electronic Supplementary Material (ESI) for Nanoscale Advances.

Controlled growth of 2D ultrathin Ga_2O_3 crystals on liquid metal

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Methods

Preparation. The commercial Ga pellet was divided into small droplets in hot ethanol. A droplet of Ga (10 mg) was then placed on a W foil (50 mm, cut into 100 cm squares). Before loading gallium, the W foils were ultrasonicated and rinsed with acetone, ethanol and deionized water prior to being dried under a nitrogen stream. The Ga pellets had a purity of 99.9999 wt.% and the W foils with a purity of 99.95 wt.% were purchased from Alfa Aesar China (Tianjin) Co. Ltd. And Shanghai Minor Metals Co. Ltd.

CVD growth of Ga_2O_3. The CVD growth of Ga_2O_3 was carried out in a quartz tube furnace under ambient pressure. The growth protocol consisted of three steps: (1) heating the Ga-W substrates to 1020°C at a rate of 30–40°C/min under the flow of Ar; (2) exposure of the substrates to oxygen source at 1020°C for 30 min under the flow of Ar; and (3) cooling the substrates to room temperature at a rate of 20°C/min under Ar.

Characterization. Optical images were taken with an optical microscope (Olympus, Olympus DX51), and the SEM images were taken with a high-resolution SEM (Hitachi, SU8010) operating at 10 kV. Raman spectroscopy was performed with a laser micro-Raman spectrometer (Renishaw, Renishaw inVia, 532 nm excitation wavelength). The XRD spectra was performed on Rigaku SmartLab 9kw. XPS was performed on Kratos Analytical, the binding energies were calibrated by referencing the C 1s peak (284.8 eV).

Transferring the two dimensional (2D) ultrathin Ga_2O_3 crystals to the target substrates: Apply the tape to the substrate on which the ultrathin crystals grow, and then peel it off completely. Paste to Si/SiO_2, then heat to 80 ℃ for 10 minutes, so the tape becomes less sticky, release the crystals, then remove the tape, ultra-thin crystals are left on Si/SiO_2substrate.

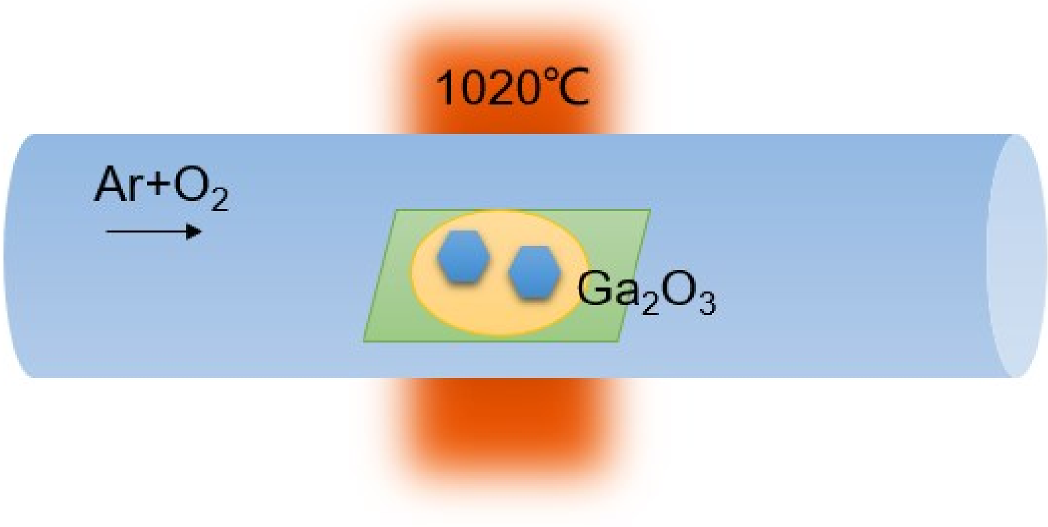

Figure S1. Sketch map of the CVD procedure for 2D ultrathin Ga_2O_3 crystals at the ambient pressure.

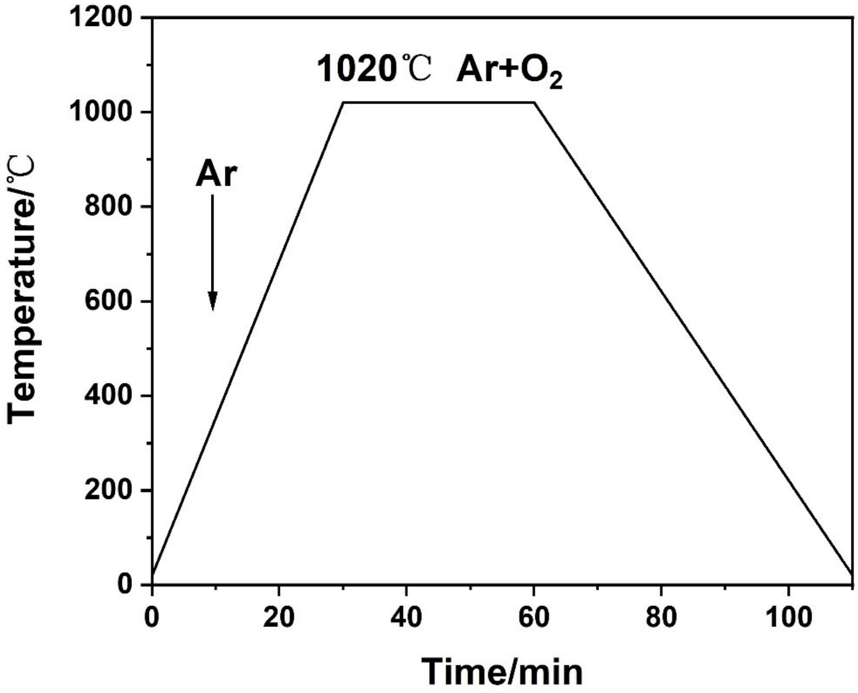

Figure S2. Schematic diagram of heating process of CVD system

Firstly, the Ga–W substrates (the W foil with a Ga pellet of 10 mg on its surface) were heated to 1020 ºC in the Ar atmosphere. Then, the Ga spreads over the entire substrate by itself into uniform and ultra-flat surface at elevated temperature due to the good wettability onto the W. The liquid gallium was then held at 1020 °C for 30 minutes, oxygen (10 sccm) was then pumped in to initiate the growth. Thereinto, with the surficial atomic Ga layer serving as templates and reacts with oxygen, high-quality 2D ultrathin Ga_2O_3 crystals can be obtained. Finally, the sample is quickly moved out of the high temperature zone and rapidly cooled in the environment to retain the crystals.

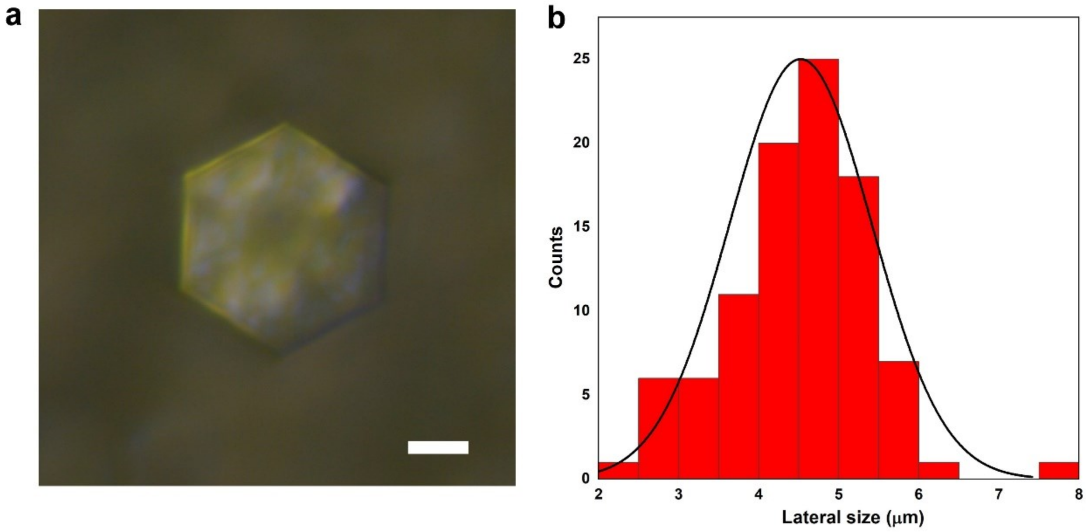

Figure S3. (a) Optical images of α-Ga_2O_3 crystals. (b)Statistical distributions of the lateral size 2D ultrathin Ga_2O_3 crystals. The scale bar in (a) is 2 μm.

The Figure S3a shows an optical image of a single hexagonal ultrathin Ga_2O_3 sheet. The morphology and regularity of the crystal can be seen. The lateral size distributions of 2D ultrathin Ga_2O_3 crystals on liquid Ga are analyzed. The statistical results shown in Figure S3b indicated the distribution of the grain sizes, where the average value of them was calculated to be 4.5 μm.

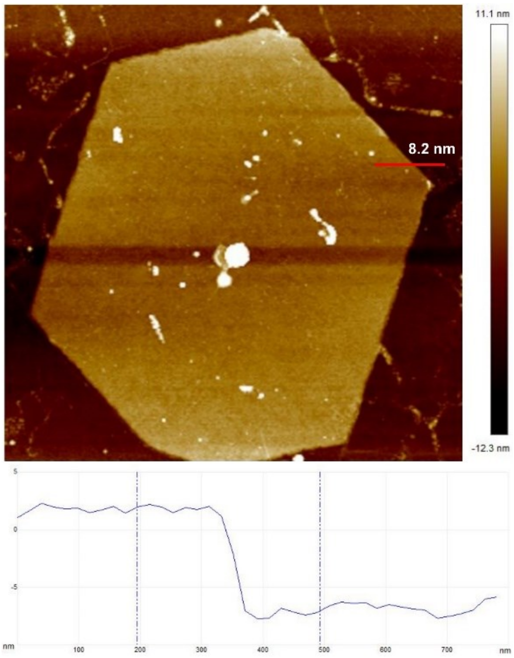

Figure S4. Thickness measurement of Ga_2O_3 sheet. It is noted that the thickness is around 8 nm.

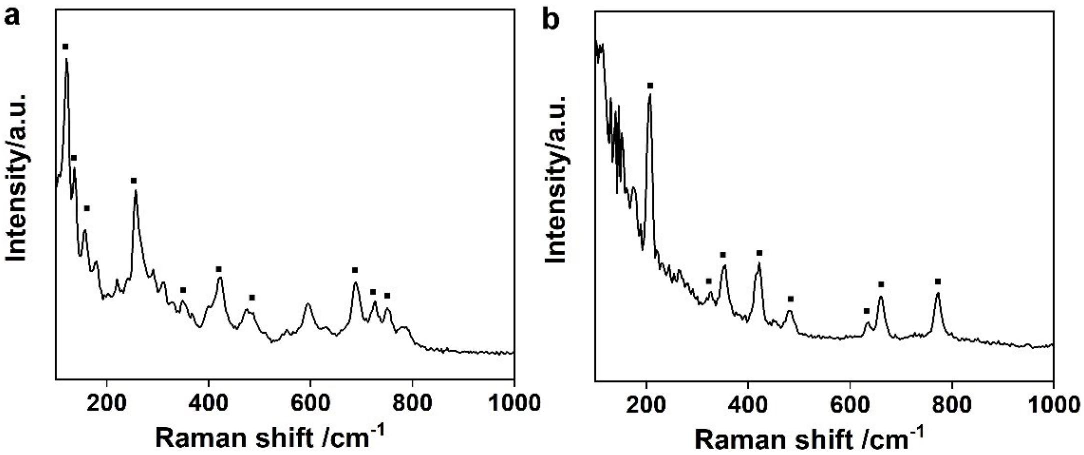

Figure S5. Raman spectrum of α-Ga_2O_3 and β-Ga_2O_3, respectively.

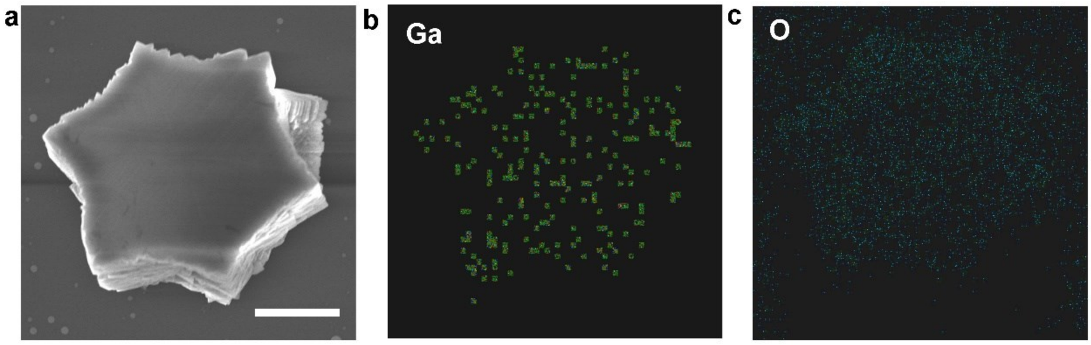

Figure S6. EDS mapping of the as grown Ga_2O_3 crystal. (a) SEM images of the hexagonal α-Ga_2O_3 crystal.(b), (c) EDS elemental mapping of Ga and O. The scale bar is 2 μm.

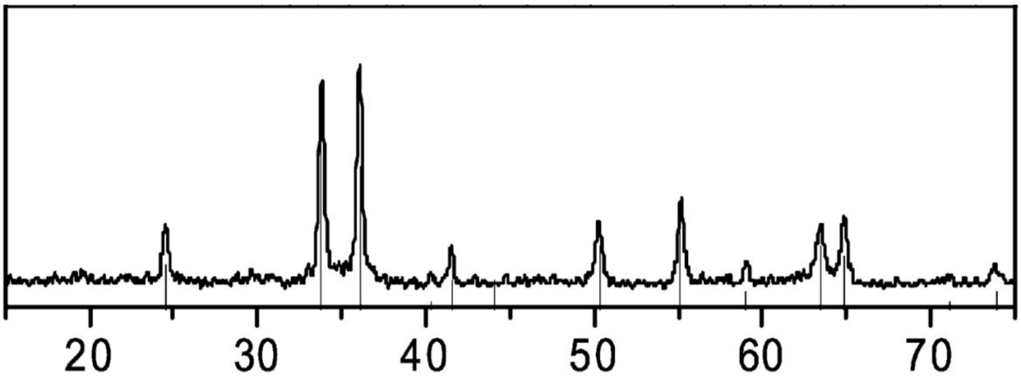

Figure S7. XRD spectrum of α-Ga_2O_3 crystals.

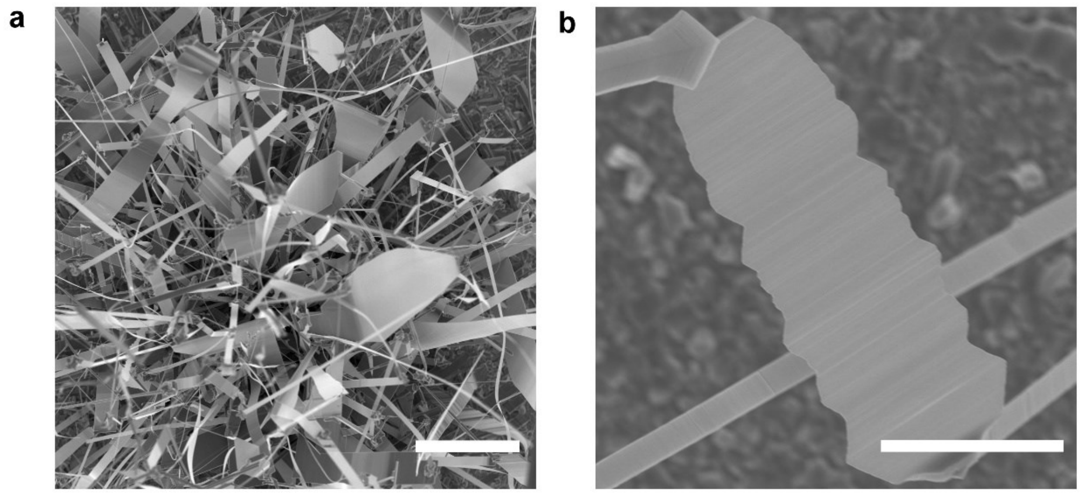

Figure S8. (a), (b) SEM images of β-Ga_2O_3 nanosheets. The scale bars in (a) is 20 μm and in (b) is 5 μm.

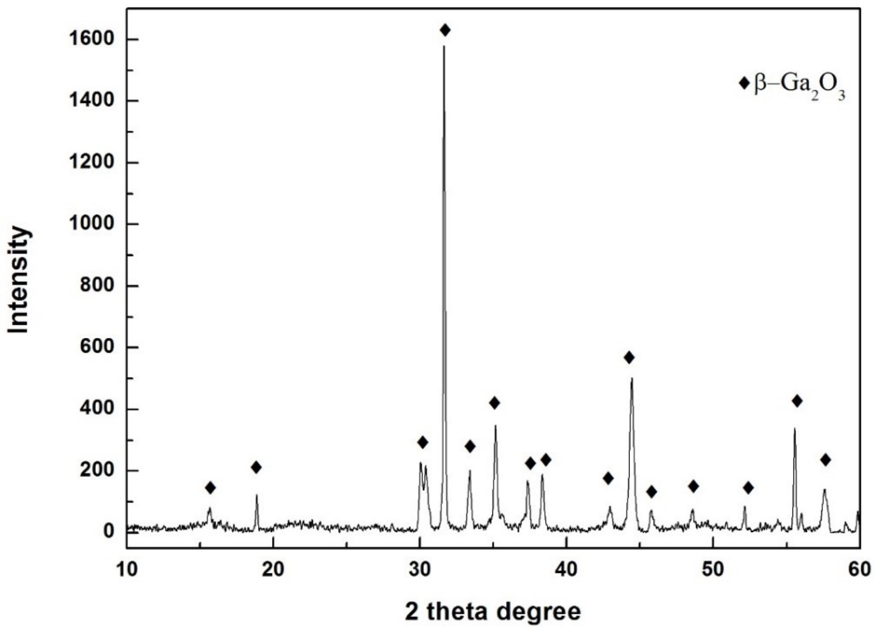

Figure S9. XRD spectrum of β-Ga_2O_3 crystals.

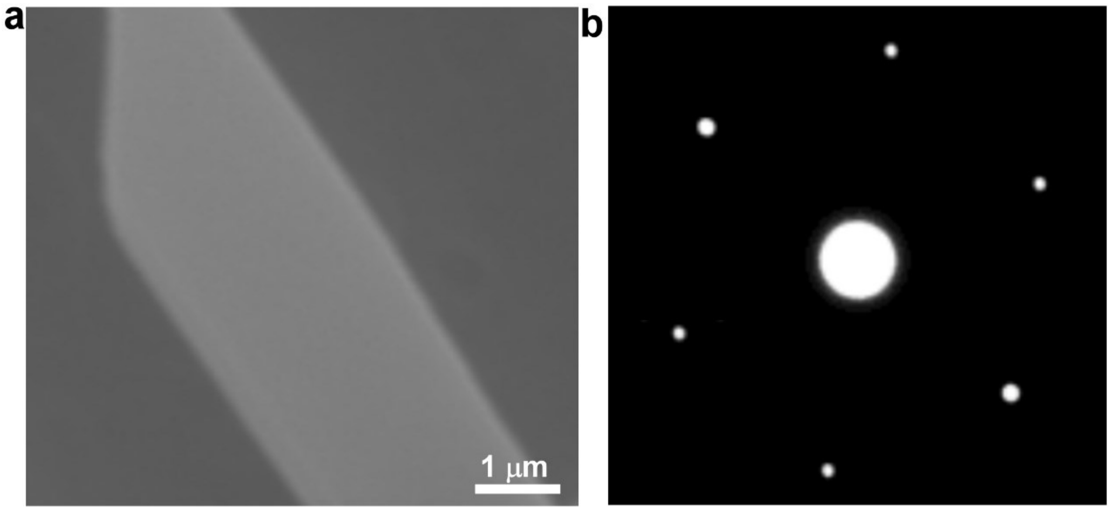

Figure S10. TEM measurement of the as-grown β-Ga_2O_3 samples.

Table S1 Effect of gas velocity on crystal thickness

Ga/mg	Ar/sccm	O_2/sccm	Temperature/℃	Growing time /min	crystal thickness /nm
10	100	5	1020	30	20
10	150	8	1020	30	15
10	200	12	1020	30	8

Table S2 Effect of oxygen content on crystal morphology

Ga/mg	Ar/sccm	O_2/sccm	Temperature/℃	Growing time/min	Crystal morphology
10	200	5	1020	30	small, irregularly shaped
10	200	8	102	30	suborbicular
10	200	13	1020	30	hexagonal