## **Supporting Information**

# Piezoelectricity in Wide Bandgap Semiconductor 2D Crystal GaN Nanosheet

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#### Details of CVD growth of GaN

The urea (CO(NH<sub>2</sub>)<sub>2</sub>) powders (~ 5 mg) were placed in the position of zone 2 with a ceramic boat, where the temperature can reach to 150°. The Ga pallet supported by a  $1 \times 1$  cm<sup>2</sup> W foil was placed in the position of zone 1, which is the central temperature of the furnace. Firstly, the Ga pallet would get molten and spread over the surface of W foil to form the Ga-W solid solution when the temperature of zone 1 was up to 1110°. Secondly, the temperature of zone 2 would reach to 150° so that the urea precursors were moved into the zone 1 with the flow of H<sub>2</sub>/Ar. The nitridation reaction would occur at zone 1 with the urea as the N source precursor and the molten Ga as the Ga source precursor and the GaN nanosheets could be formed on the surface of Ga-W solid solution in the CVD system.

#### Mechanism of CVD growth of GaN

When the Ga-W substrates were heated to 1110 °C, the surface tension of molten W (2361.5 mN/m) is much larger than that of molten Ga (628.7 mN/m). Therefore, the outmost surface of the substrate has spread with molten Ga to form GaN thick layers. The sub-surface is Ga-W solid solution. During the process of CVD growth of GaN, the urea precursors were moved into the reaction zone with the flow of  $H_2/Ar$ . The nitridation reaction would occur with the urea as the N source precursor and the molten Ga as the Ga source precursor and the GaN layers could be formed on the outmost surface of the substrate. Because the Gibbs free energy of WN (-121 KJ/mol) is lower than that of GaN (-18 KJ/mol), W atoms are more easily combined with N atoms to form W-N bonds. Therefore, the exist of Ga-W solid solution can relatively hinder the thickening of GaN layers owing to the stronger nitridation ability of W atoms than that of Ga atoms. But the molten point of W is ultra-high (3410 °C). The tube furnace cannot reach to this value. The amount of molten W is too small. Therefore, the thickness of Ga-W solid solution is too thin. As a result, the Ga-W solid solution has limited nitridation ability to impede the thickening of GaN nanosheets. Therefore, we can obtain the relatively thick GaN crystals with CVD system. Then the obtained GaN can be thin down through the mechanical exfoliation to get the correspond 2D GaN nanosheets.

### Reproducibility and scalability for producing 2D GaN nanosheets

The process of growth of 2D GaN nanosheets can be divided into two steps. Firstly, the CVD system is used to grow GaN thick crystals on the Ga-W substrate, which can be easily achieved under  $H_2/Ar$  atmosphere because the growth conditions are not strict. Secondly, the mechanical stripping is used to thin down the thickness of GaN. Finally, we can obtain GaN nanosheets with the thickness ranging from tens to hundreds of nanometers, as shown in Figure S3 (a) and (b). Through our method, we can easily obtain the 2D GaN nanosheets with good reproducibility and scalability.



Figure S1. Sketch map of the CVD process for the growth of GaN nanosheets.



Figure S2 (a) HRTEM atomic image of GaN nanosheets, the insert is the IFFT of the marked area with white square lines, (b) FFT of GaN crystals.



Figure S3. (a) The morphology of GaN nanosheets on the Ga-W substrate, (b) The morphology of GaN nanosheets transferred to the SiO<sub>2</sub> substrate.



Figure S4. The AFM topography of GaN nanosheets with the thickness of (a) 245 nm and (b) 32 nm.



Figure S5. (a) Surface potential of sample without control. (b) Surface potential of sample with reverse voltage control.



Figure S6. (a) Resonant frequency tracking in DART-PFM mode. (b) Out-of-plane amplitude maps after fitting with the SHO model.



Figure S7. Histogram of the distribution of piezoelectric amplitude data.