Supporting Information for:

Wafer-size and single-crystal $MoSe_2$ atomically thin films grown on GaN substrate for light emission and harvesting

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1. SEM characterization of the $MoSe_2$/GaN samples from Phase 1-3.

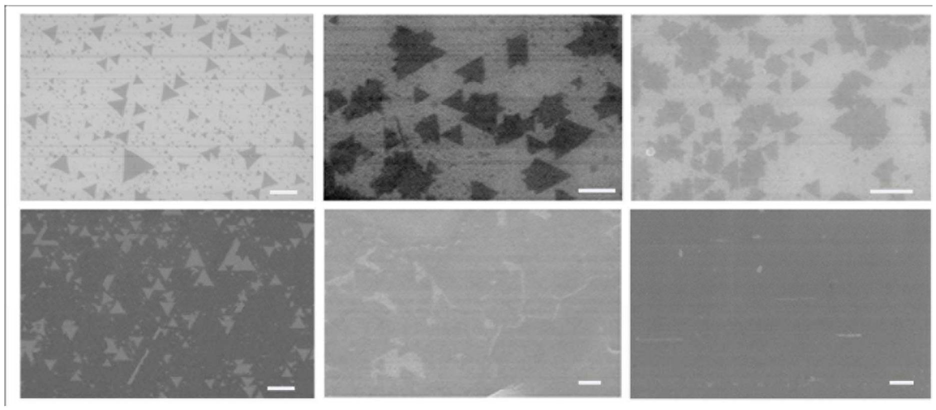

Figure S1: SEM images at different distances from the source, showing the increasing coverage of GaN surface. Scale bar: 2 μm

2. XPS characterization of the $MoSe_2$/GaN sample.

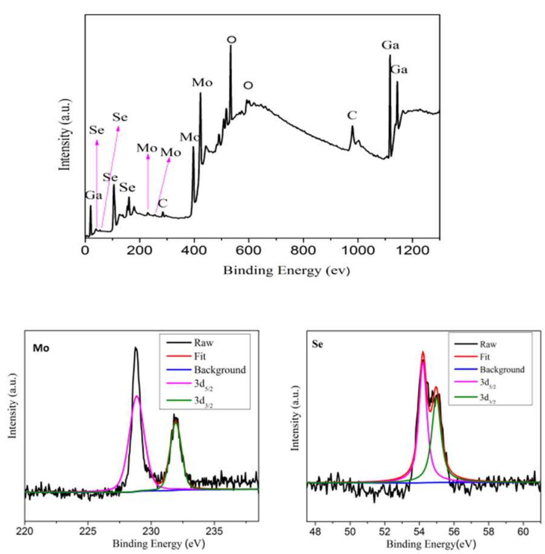

Figure S2: XPS survey and Mo- and Se-related peaks.

Four elements are present in the spectra acquired: Mo and Se from the CVD products, as well as Ga and N from the GaN substrate. Mo $3d_{3/2}$ and $3d_{5/2}$ core levels peaks are located at ~232 and 228.8 eV, respectively, while the peak of Se 3d is located around 54.0 eV, in agreement with the values obtained in other $MoSe_2$ systems.[1,2] The peaks at 228.8 and 232 eV are attributed to the doublet Mo $3d_{5/2}$ and Mo $3d_{3/2}$ binding energies, respectively, for Mo^{4+}. The peaks at 54.3 and 55.1 eV, corresponding to the Se $3d_{5/2}$ and Se $3d_{3/2}$ orbitals of divalent selenide ions (Se^{2-}). The XPS analysis demonstrates that the obtained material is $MoSe_2$ without other impurities.

3. Raman spectrum of the Phase 1 $MoSe_2$/GaN sample.

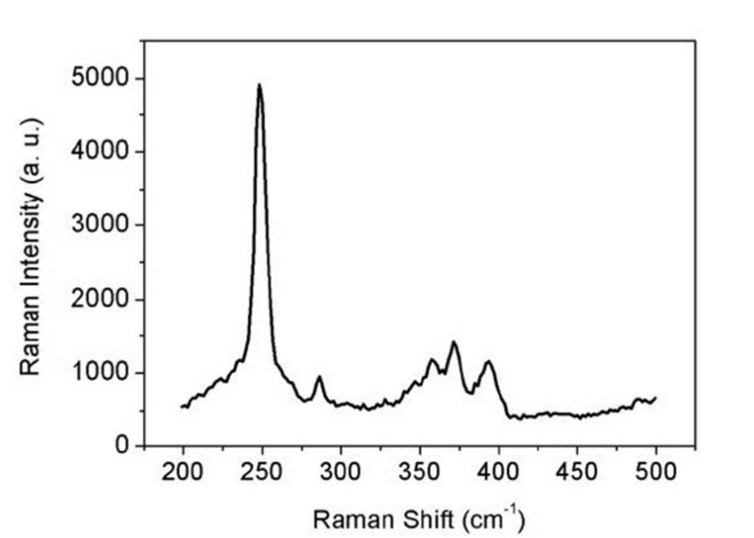

Figure S3: Raman spectrum of the Phase 1 sample.

4. Raman spectrum of the GaN surface area in the Phase 2 sample.

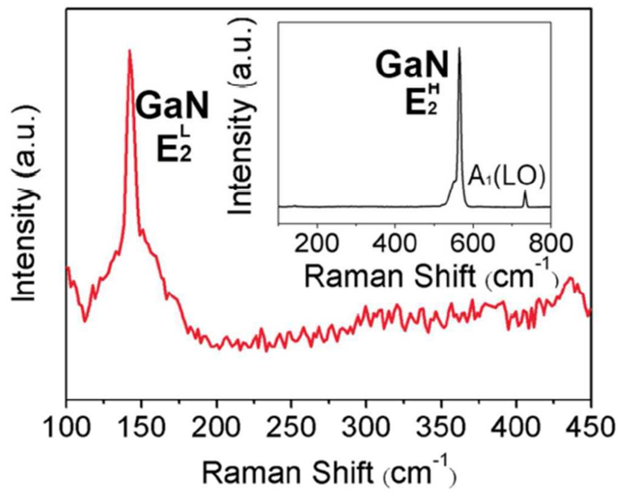

Figure S4: Raman spectrum of point A in Figure. 3 a.

5. PL characterizations of $MoSe_2$ films with different phase.

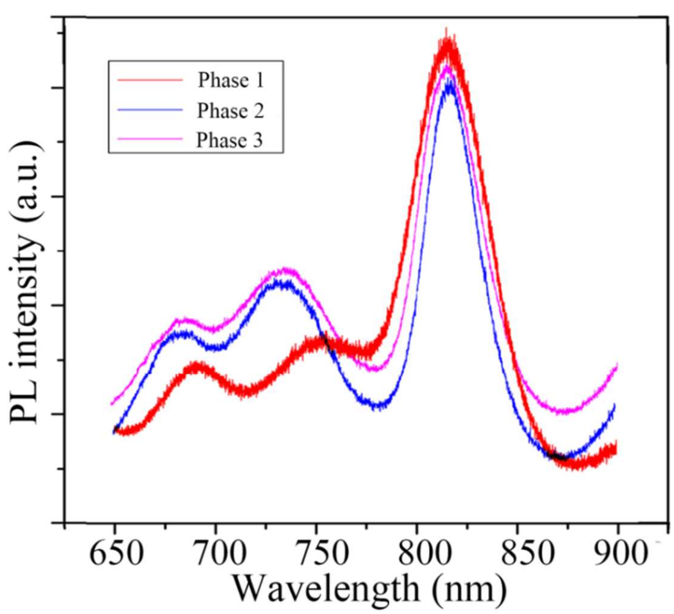

Figure S5: PL spectra of the as-grown samples with different growth modes. We can obviously observed that the PL peaks of Phase 1-3 are similar.

6. Raman spectrum of the GaN surface area in the Phase 2 sample.

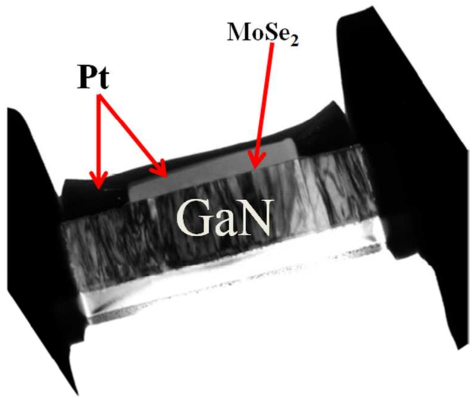

Figure S6: Low-magnification TEM image of the FIB-processed sample slice. $MoSe_2$ film exists between GaN substrate and the covered Pt.

7. $MoSe_2$ growth on p-type GaN on a sapphire substrate.

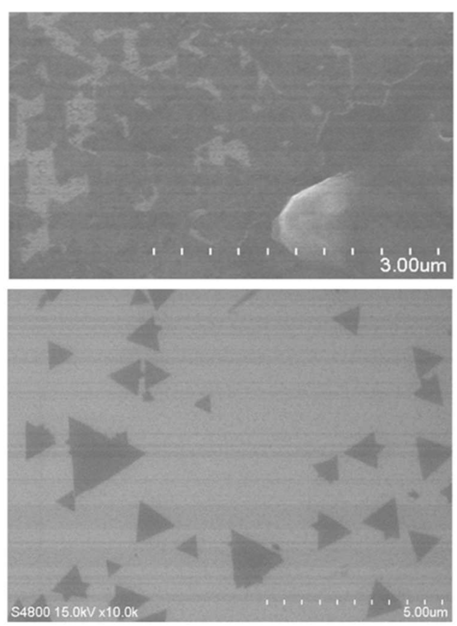

Figure S7: SEM images of the $MoSe_2$ grown on a p-type GaN surface.

8. UPS characterizations of $MoSe_2$ on a GaN substrate and on a p-type GaN film.

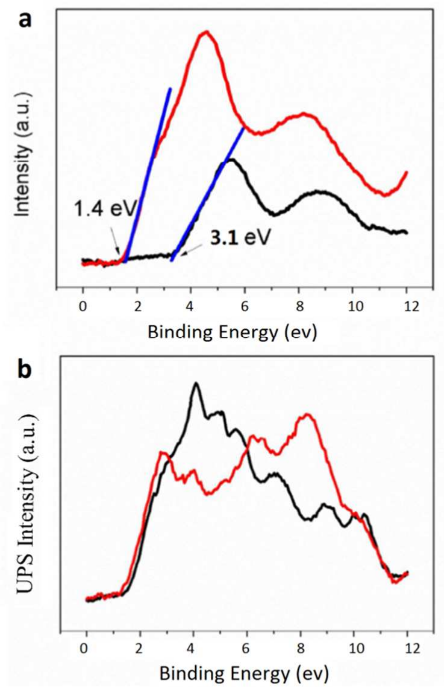

Figure S8: (a) UPS spectra of $MoSe_2$ (red) and the GaN substrate (black). (b) UPS spectra of $MoSe_2$ (black) and the p-type GaN substrate (red).

Referances

（1） Gong, Y. J.; Liu, Z.; Lupini, A. R.; Shi, G.; Lin J. H.; Najmaei, S.; Lin, Z.; Elias, L.; Berkdemir, A.; You, G.; Terrones, H.; Terrones, M.; Vajtai, R.; Pantelides, S. T.; Pennycook, S. J.; Lou, J.; Zhou, W.; Ajayan, P. M. Band Gap Engineering and Layer-by-Layer Mapping of Selenium-Doped Molybdenum Disulfide. *Nano Lett.* **2013,** 14, 442-449.

（2） Wang, X.; Gong, Y.; Shi, G.; Chow, W. L.; Keyshar, K.; Ye, G.; Liu, Z.; Ringe, E.; Tay, B. K.; Ajayan, P. M. Chemical Vapor Deposition Growth of Crystalline Monolayer $MoSe_2$. *ACS Nano* **2014**, 8, 5125-5131.