Supporting Information

Low-temperature growth of two-dimensional layered chalcogenide crystals on liquid

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Characterization of 2D GaSe crystals

2D GaSe crystals were characterized by optical microscopy (OM, Olympus DX51 microscope), scanning electron microscopy (SEM, Hitachi S-4800, acceleration voltage 1-30 kV), atomic force microscopy (AFM, Vecco Nanoscope IIIa, tapping mode), and transmission electron microscopy (TEM, FEI Tecnai F20, acceleration voltage 200 kV) equipped with an energy dispersive X-ray spectrometer (EDX). X-ray diffraction spectroscopy (XRD) was performed by the XPert Pro (Cu Kα1 λ = 1.540598 Å, Kα2 λ = 1.544426 Å, 80 kV, 100mA). X-ray photoelectron spectroscopy (XPS) analysis was conducted on a Kratos Axis Ultra spectrophotometer with the monochromatic Al X-ray at low pressures of 5×10^{-9} to 1×10^{-8} Torr. Electrical measurements were performed in a Micromanipulator 6200 probe station with Keithley 4200 semiconductor analyzer.

The photodetector devices were in series with fixed resistor (R) for the a.c. photovoltage measurement. Photocurrent (V_{AC}/R) maps were achieved by a lock-in amplifier with a mechanical chopper modulated laser and the Keithley 2400 providing the bias voltages. A 532 nm Griot He–Ne laser was focused on samples through a Ni-U Nikon optical microscope with a microstage with alignment accuracy of better than 0.1 μm. The laser power was calibrated by an optical power meter (Newport, model 840-C).

EDX results of 2D GaSe and $Ga_xIn_{1-x}Se$ crystals

The 2D GaSe crystals were transferred from liquid metal surface to Cu grid according to the PMMA-mediated transfer technique, and then were characterized by a transmission electron microscope (TEM) equipped with energy dispersive X-ray spectrometer (EDX). The elemental analyses indicate that Ga and Se elements are with an atomic ratio of 1:1 (Figure S1a).

The EDX spectrum of 2D $Ga_xIn_{1-x}Se$ crystals shows obvious In peaks (Figure S1b).

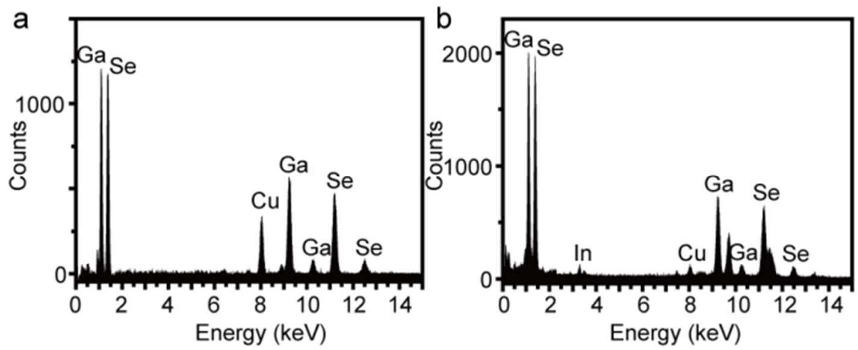

Figure S1. (a,b) Typical of 2D GaSe and $Ga_xIn_{1-x}Se$ crystals, respectively. The Cu signals come from the TEM grid.

AFM characterization of 2D GaSe crystals

We used AFM to determine the thickness of synthesized 2D GaSe crystals after transferred onto 300 nm SiO_2/Si substrate. The thickness of 2D GaSe crystals can be tuned by adjusting the growth time. For short growth time (1 min), we can obtain samples with a uniform height of about 4.1 nm (Figure 2d). Furthermore, we prepared thicker GaSe films using longer growth time (3 min and 10 min), as shown in Figure S2a and b.

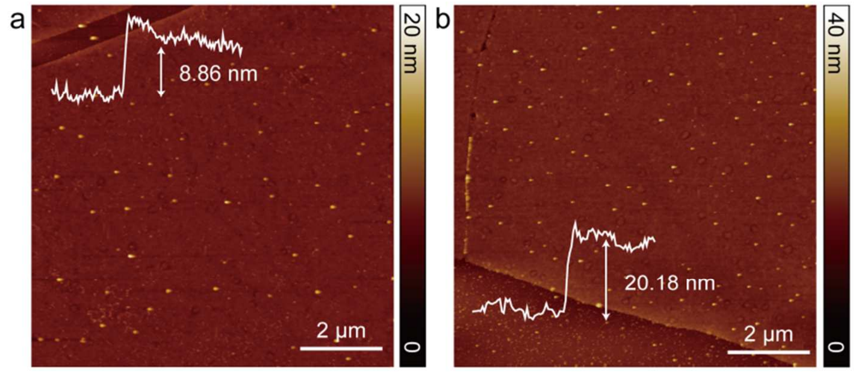

Figure S2. AFM images of 2D GaSe crystals with the thicknesses of ~8.86 nm and ~20.18 nm prepared with growth time of 3 min (a) and 10 min (b), respectively. The samples were transferred onto 300-nm-thick SiO_2 of Si substrates.

The transfer of 2D GaSe crystals onto EVA/PET

The 2D GaSe crystals grown on liquid substrates can be transferred onto EVA/PET using a hot press lamination method. Lamination process was realized using an SG 330-SCL lamination machine (Shanghai Shenguang Office Equipment Co. Ltd). The EVA layer melts at the specific temperature to adhere to the GaSe crystals. The temperature was adjusted by a knob according to the thickness of EVA/PET film. The temperature of ~100 ^{o}C was suitable for the EVA/PET film with the thickness of 125 μm in this work. The lamination machine has a set lamination speed of ~1 cm/s.

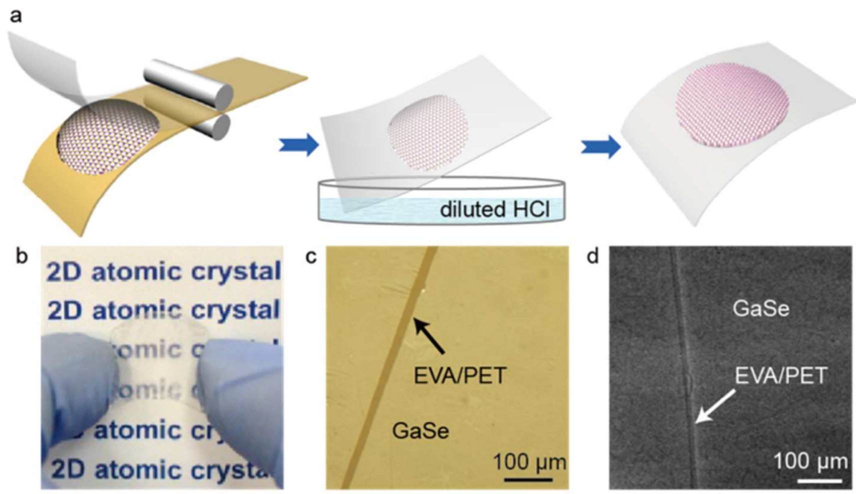

Figure S3. (a) Schematic of the transfer process from PI to EVA/PET. (b) Photograph image of the 2D GaSe crystals on the transparent and flexible PET substrate. (c) Typical OM image of the GaSe film on EVA/PET after hot-lamination transfer. (d) The corresponding SEM image.