Spatially Composition-graded Monolayer $WSe_{2x}Te_{2-2x}$ Nanosheets

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Alloying in two-dimensional (2D) transition metal dichalcogenides (TMD) has allowed bandgap engineering and phase transformation, which provide more flexibility and functionality for electronic and photonic devices [1]. To date, many ternary TMD alloys with homogenous compositions have been synthesized. However, realization of bandgap modulation spatially within a single TMD nanosheet remains largely unexplored. In this work, we demonstrate the synthesis of spatially composition-graded $WSe_{2x}Te_{2-2x}$ flakes using an in situ chemical vapor deposition (CVD) method. The photoluminescence (PL) and Raman spectra line-scanning characterization indicate a spatially graded bandgap, which increases from 1.46 eV (center) to 1.61 eV (edge) within one monolayer flake. Furthermore, the electronic devices based on this spatially graded material exhibit tunable transfer characteristics.

The schematic and the temperature profile of the one-step CVD growth of spatially composition-graded $WSe_{2x}Te_{2-2x}$ monolayer flakes are shown in Fig. 1a and 1b respectively. The growth includes two high-temperature processes. During the first stage of the growth (step i), the temperature was held at 800 ℃ for 10 minutes. In the second stage of the growth (step ii), the temperature was decreased to 695 ℃, while the movable boat carrying additional tellurium (Te) powder was pushed into the heating zone via magnet. The temperature was kept at 695 ℃ for 10 minutes. This process enables the growth of spatially graded $WSe_{2x}Te_{2-2x}$ monolayer flakes. The optical image of a synthesized $WSe_{2x}Te_{2-2x}$ flake is shown in Fig. 1c. The PL line scan is taken from the center to edge of the flake (marked in Fig. 1c) and the PL spectra are shown in Fig. 1d. The PL peak energy ranges from 1.46 eV (center) to 1.61 eV (edge). The corresponding PL contour plot (Fig. 1e) clearly shows a continuous blue shift of the PL peak as the laser spot moves from center to edge. Furthermore, the Raman line-scanning spectra and the contour plot of the flake also confirmed a graded-composition in the flake, as shown in Fig. 1f and 1g. As the laser spot moves towards the edge, both the E_{2g}^1 and A_{1g} peaks blueshift, while the intensity ratio between A_{1g} and E_{2g}^1 peak gradually increases, which is consistent with the Raman spectrum taken in the exfoliated $WSe_{2x}Te_{2-2x}$ with various Te concentrations [3]. These results indicate that this CVD growth method realizes continuous tuning of bandgap in lateral direction within a single monolayer $WSe_{2x}Te_{2-2x}$ flake. Moreover, the AFM image and extracted height profile manifest monolayer thickness and a uniform surface, as shown in Fig. 1h and 1i. The bandgap engineering in spatially composition-graded $WSe_{2x}Te_{2-2x}$ enables tunable multi-channel electronic devices. Fig. 1j and 1k show the optical image and transfer curves of the multi-channel transistors formed in various regions respectively. As the channel location moves from the center to the edge regions, the on-state drain current first increases, reaches to maximum value in middle region, and then decreases again. This phenomenon can be explained by the two competing sources of resistance: the Schottky contact resistance and the channel resistance. As the tellurium concentration increases, the bandgap of the $WSe_{2x}Te_{2-2x}$ alloy decreases, which leads to higher carrier concentration and lower channel resistance [3]. On the other hand, increasing tellurium concentration will result in significantly increase of electron affinity, which will increase the hole barrier height at the Pd/$WSe_{2x}Te_{2-2x}$ contacts and increase the Schottky contact resistance for hole transport [2]. Thus, the total resistance reaches minimum, when the tellurium concentration is moderate, which is consistent with the experimental observation. This work provides the groundwork for the synthesis of composition-graded TMD materials and opens a new route toward tailoring the electrical properties of TMDs by bandgap engineering in the future.

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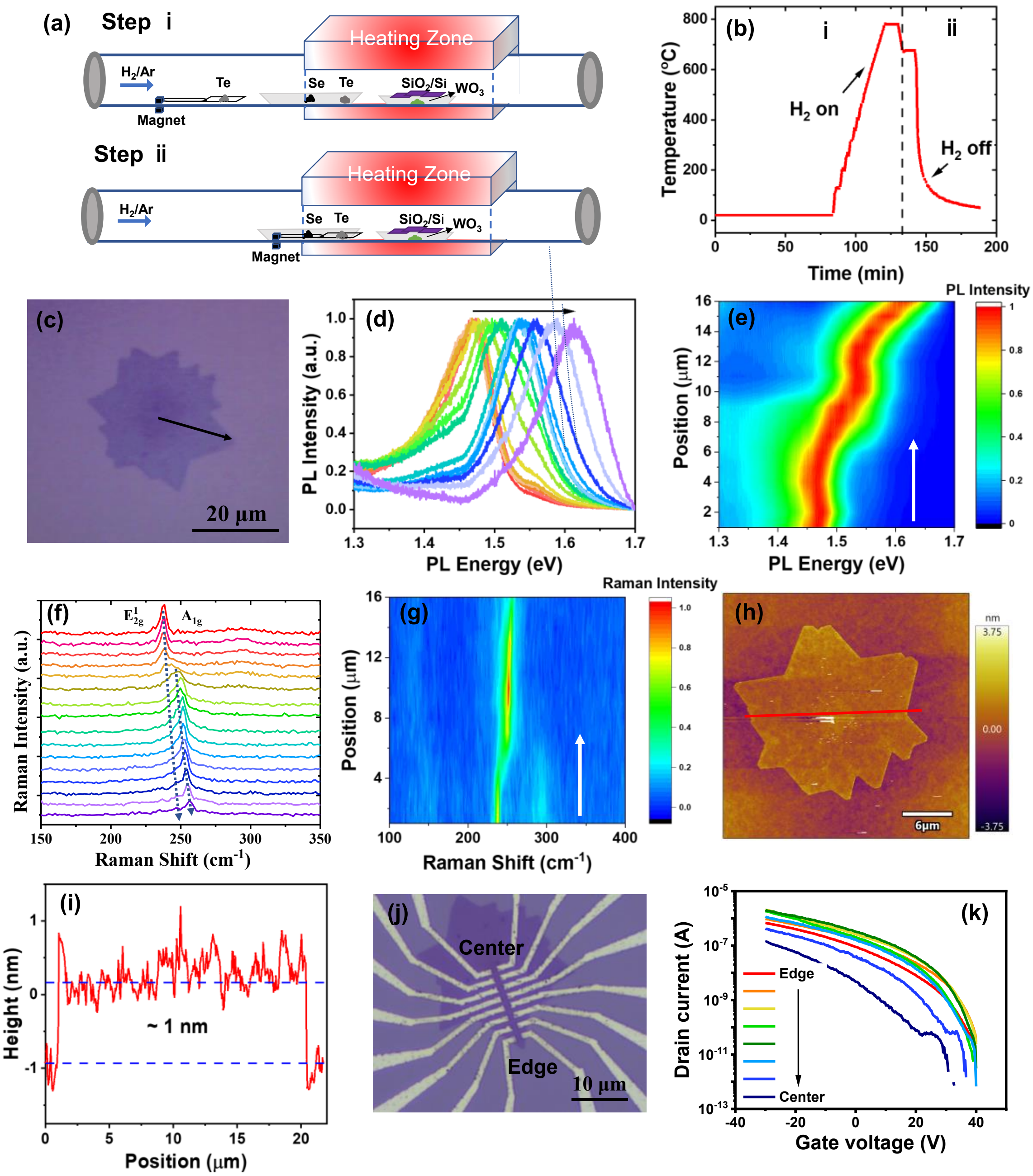

Figure 1. (a) Schematic and (b) corresponding temperature profile of in situ CVD growth of spatially gradient $WSe_{2x}Te_{2-2x}$ monolayer flakes. (c) Optical image of one monolayer flake. (d) PL line-scan spectra and (e) corresponding contour plot indicate continuous bandgap modulating from the center to the edge within one flake. (f) Raman line-scan spectra and (g) contour plot show a gradual blue-shift of E_{2g}^{1} and A_{1g} peaks from center to edge. (h) AFM image and (i) extracted height profile manifest monolayer thickness and a uniform surface. (j) Optical image and (k) transfer curves of the multichannel transistors located in various regions.

References:

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