

## Supporting Information

### Epitaxial Growth of Structure-Tunable ZnO/ZnS Core/Shell Nanowire Arrays using HfO<sub>2</sub> as Buffer Layer

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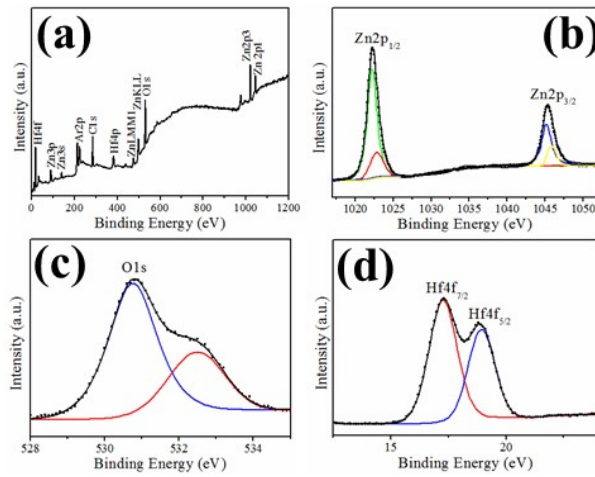
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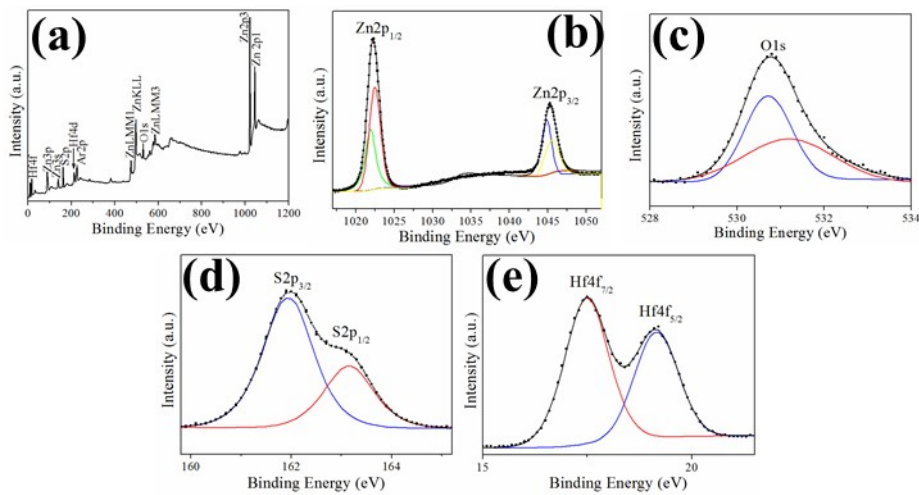
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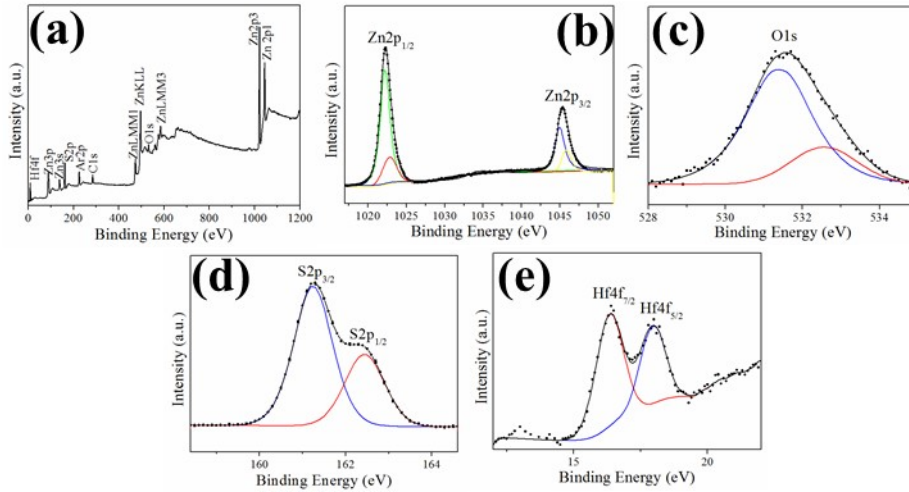
<sup>c</sup> Center of Super-Diamond and Advanced Films (COSADF) and Department of Materials Science and Engineering, City University of Hong Kong SAR, P.R.China.



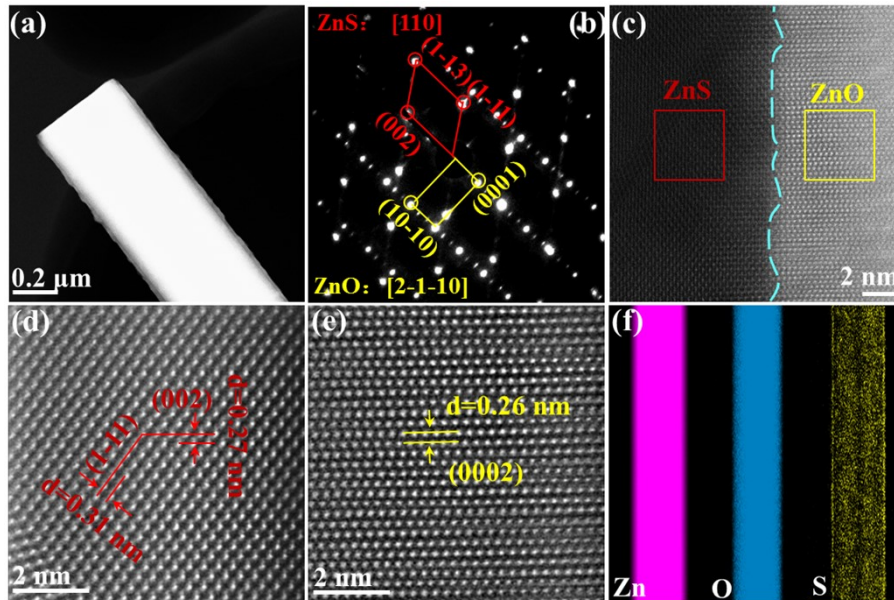
**Figure S1.** XPS spectra and fitting results of ZnO/HfO<sub>2</sub> nanowire arrays (a) XPS survey spectrum of the ZnO/HfO<sub>2</sub> nanowire arrays. (b) XPS spectra of Zn2p peaks, (c) O1s peaks and (d) S2p peaks. The thickness of HfO<sub>2</sub> is 2 nm.



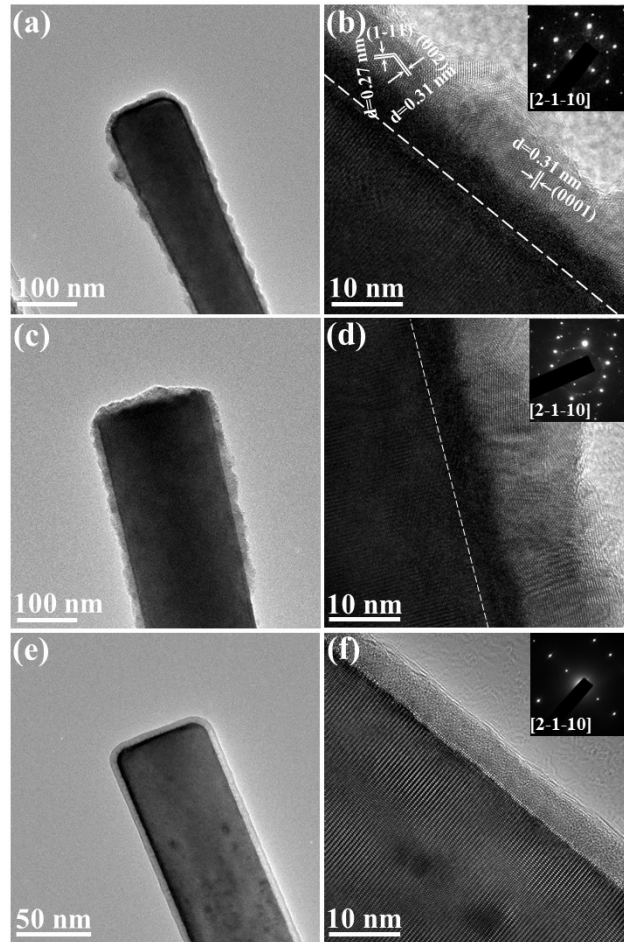
**Figure S2.** XPS spectra and fitting results of ZnO/ZnS core/shell nanowire arrays with 1 nm HfO<sub>2</sub> as buffer layer. (a) XPS survey spectrum of the sample. (b) XPS spectra of Zn2p peaks, (c) O1s peaks, (d) S2p peaks and (e) Hf4f peaks.



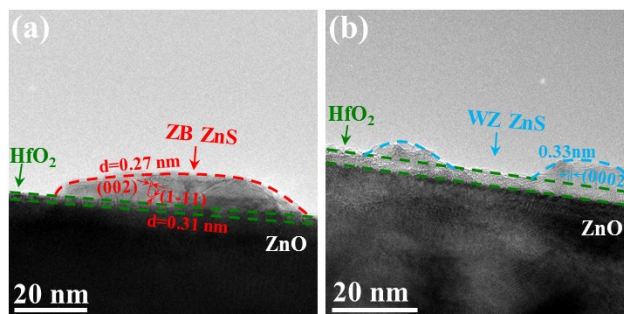
**Figure S3.** XPS spectra and fitting results of ZnO/ZnS core/shell nanowire arrays with 2 nm HfO<sub>2</sub> as buffer layer. (a) XPS survey spectrum of the sample. (b) XPS spectra of Zn2p peaks, (c) O1s peaks, (d) S2p peaks and (e) Hf4f peaks.



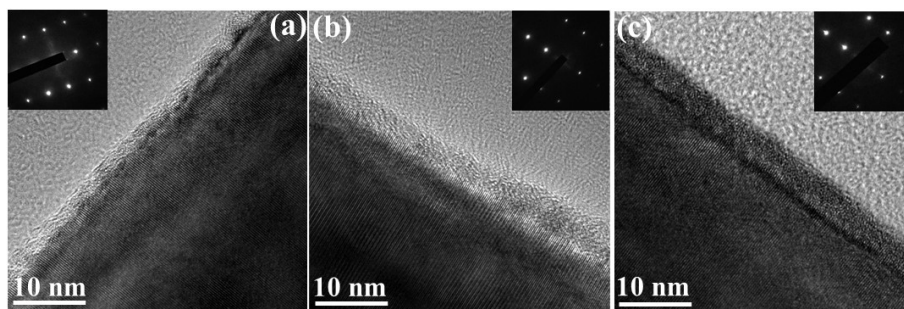
**Figure S4** Nanostructure and composition characterization of ZnO/ZnS core/shell nanowire without HfO<sub>2</sub> buffer layer. (a) Low-magnification ADF-STEM image. (b) Corresponding SAED pattern. (c) ADF-STEM image. (d-e) Enlarged ADF images of ZB ZnS and WZ ZnO, respectively. (f) Elemental maps of the core/shell heterostructure.



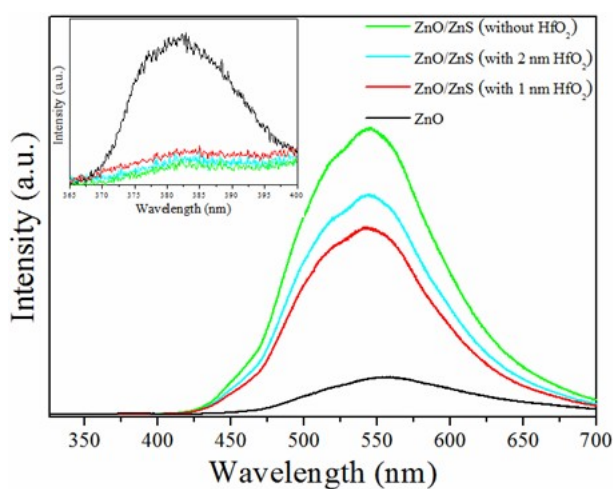
**Figure S5. Nanostructure and morphology characterization of ZnO/ZnS core/shell nanowire with HfO<sub>2</sub> buffer layer of various thicknesses.** (a,c and e) TEM image of the obtained ZnO/ZnS nanowire with an HfO<sub>2</sub> buffer layer, the thicknesses are 3 nm, 4 nm and 5 nm, respectively. (b, d and f) The corresponding HRTEM images, and the insets show the SAED patterns.



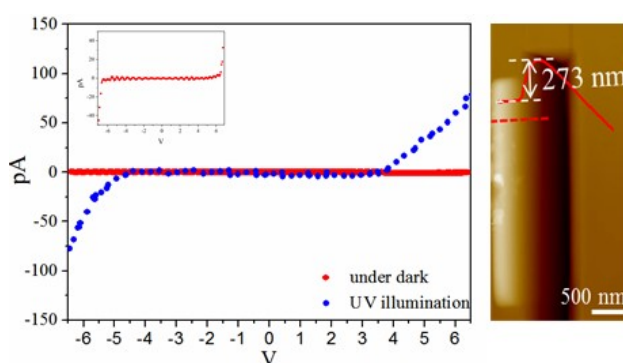
**Figure S6. HRTEM images of the obtained core/shell heterostructures using (a) 1 nm and (b) 2 nm HfO<sub>2</sub> buffer layer at the growth stage of 10 min.**



**Figure S7.** HRTEM images of the obtained (a) 1 nm, (b) 2 nm and (c) 4 nm HfO<sub>2</sub>-coated ZnO after heating while other thermal conditions obtained unchanged, insets are corresponding SAED patterns.



**Figure S8** PL spectra obtained at room temperature (RT) for ZnO/ZnS core/shell nanowire arrays with different thicknesses of and without HfO<sub>2</sub>, respectively.



**Figure S9. Photoelectric performance of the nanodevice based on pure ZnO nanowire arrays.** I-V curve of the ZnO nanowire arrays in the dark (red) and in the presence of 325 nm illumination (blue), respectively. The right inset is the corresponding AFM image and height information of the longitudinal section sample.