# Optical anisotropy induced by ultra-strong interfacial coupling in CVD-grown $WSe_2/ReSe_2$ vertical heterostructures†

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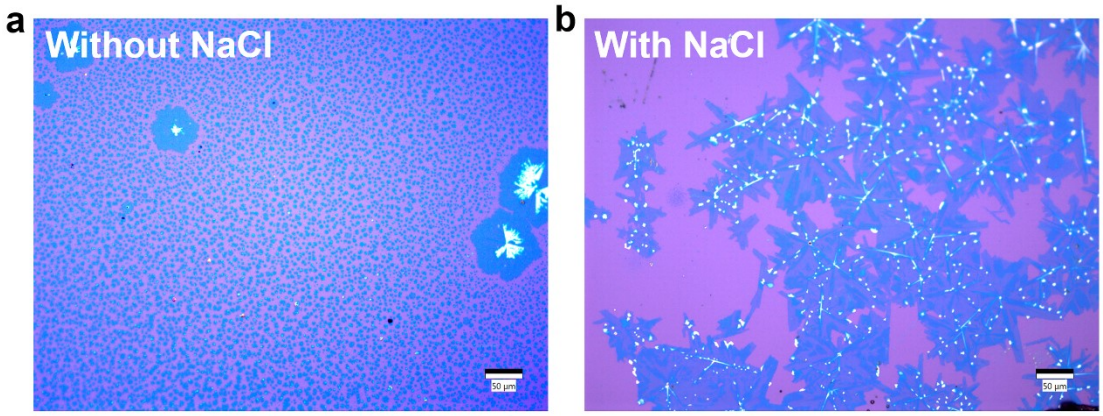


**Fig. S1** (a) Optical image of the CVD-grown $WSe_2/ReSe_2$ vertical heterostructures without NaCl assistance. (b) Optical image of the CVD-grown $WSe_2/ReSe_2$ vertical heterostructures with NaCl assistance.

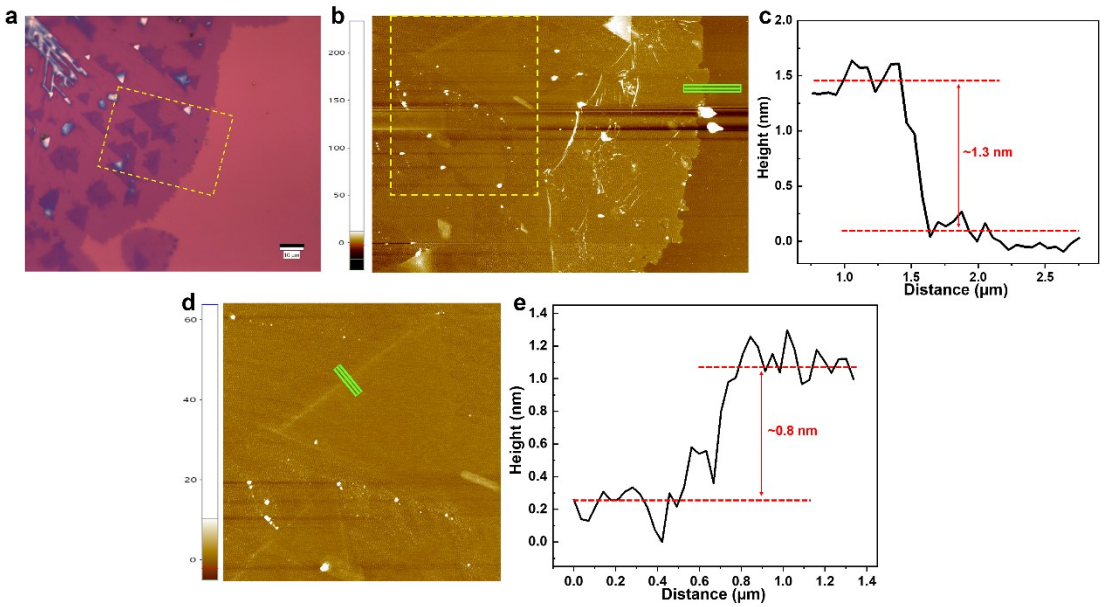


**Fig. S2** (a) Optical image of the CVD-grown $WSe_2/ReSe_2$ vertical heterostructures. (b) AFM scanning image of the yellow rectangular frame region marked in (a). (c) The height profile corresponding to the green line scanning marked in (b). (d) High-resolution AFM scanning image of the yellow rectangular frame region marked in (b). (e) The height profile corresponding to the green line scanning marked in (d).

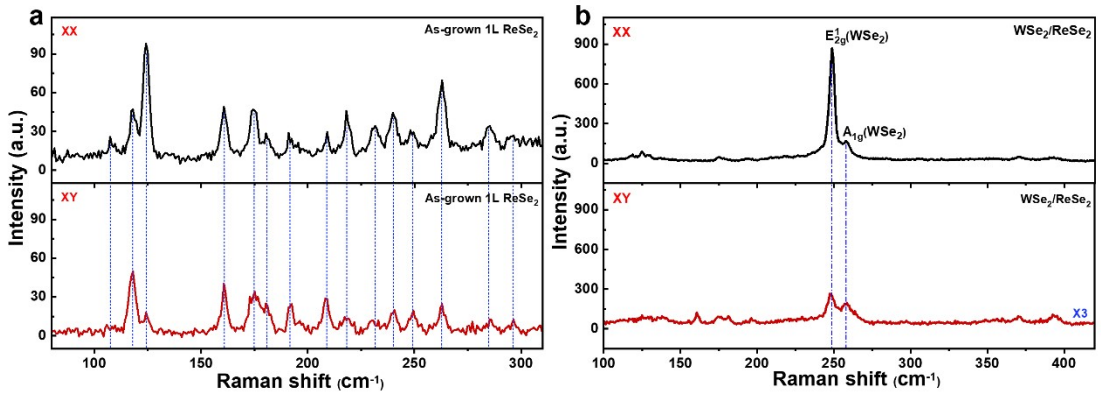


**Fig. S3** (a) Polarized Raman spectra of the as-grown monolayer $ReSe_2$ under parallel and crossed configurations. (b) Polarized Raman spectra of the as-grown $WSe_2/ReSe_2$ vertical heterostructures under parallel and crossed configurations.

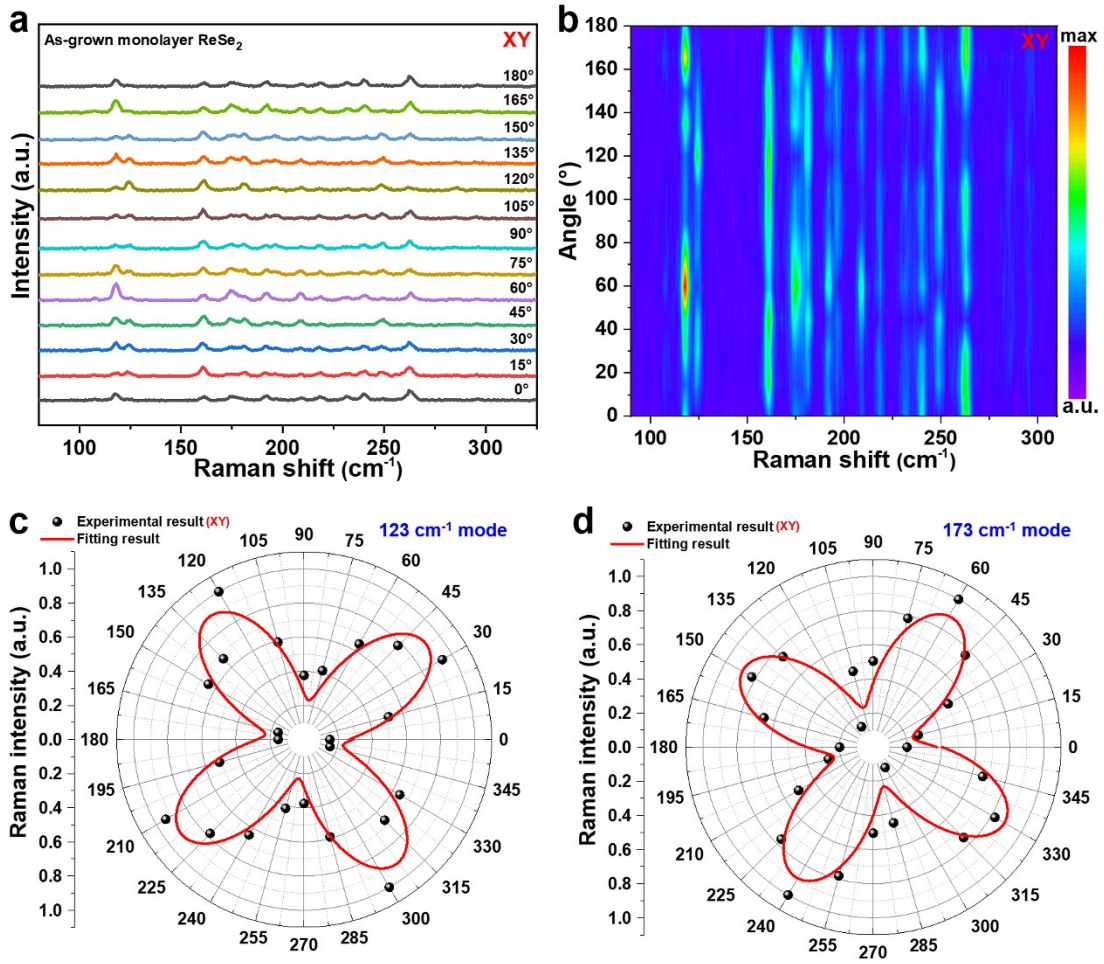


**Fig. S4** (a) Angle-resolved polarized Raman spectra of the as-grown monolayer $ReSe_2$ under crossed configurations. (b) The Raman intensity color map corresponding to (a). (c) and (d) The normalized Raman intensities of $ReSe_2$ 123 $cm^{-1}$ and 173 $cm^{-1}$ modes as a function of rotation angle.

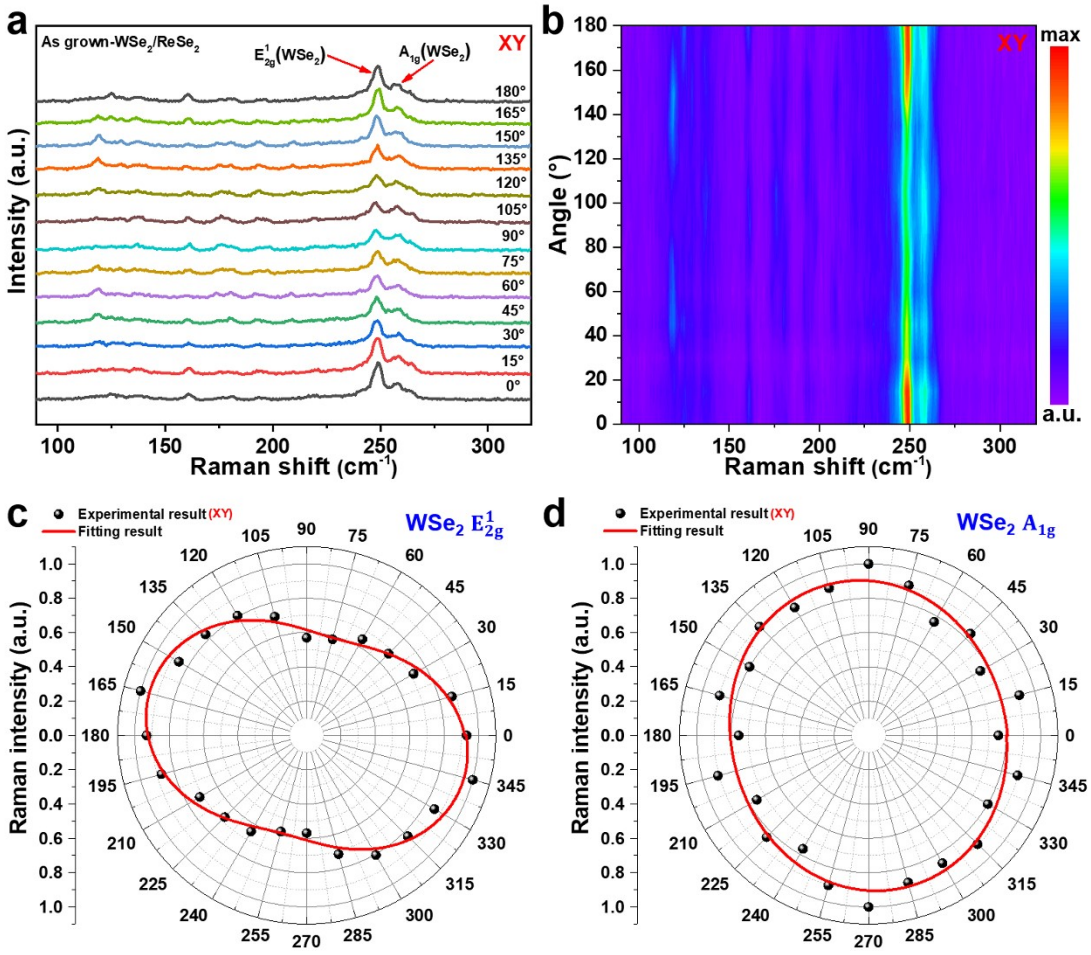


**Fig. S5** (a) Angle-resolved polarized Raman spectra of the as-grown $WSe_2/ReSe_2$ vertical heterostructure under crossed configuration. (b) The Raman intensity color map corresponding to (a). (c) and (d) The normalized Raman intensities of $WSe_2$ $E_{2g}^{1}$ and $A_{1g}$ modes as a function of rotation angle.

| Calculated in ref. 25<br>Peak position ($cm^{-1}$) | Measured in ref. 25<br>Peak position ($cm^{-1}$) | This work<br>Peak position ($cm^{-1}$) |
|---|---|---|
| 103.6 | 110.0 | 106.2 |
| 118.0 | 116.7 | 116.4 |
| 123.1 | - | - |
| 125.9 | 123.8 | 123.0 |
| 162.5 | 158.2 | 159.7 |
| 175.6 | 171.0 | 173.1 |
| 179.4 | 179.0 | 176.5 |
| 182.6 | - | 179.8 |
| 194.9 | 190.0 | 190.3 |
| 197.7 | 194.0 | 195.1 |
| 206.8 | 207.7 | 207.2 |
| 219.9 | 217.2 | 216.7 |
| 235.1 | 231.8 | 229.8 |
| 242.4 | 239.0 | 238.7 |
| 251.7 | 247.1 | 247.4 |
| 265.8 | 260.4 | 261.2 |
| 287.9 | 283.6 | 283.6 |
| 298.4 | 293.9 | 294.5 |

**Table S1** The fitted frequencies of the 17 Raman modes measured in the as-grown monolayer $ReSe_2$, and the contrast with the calculated values.