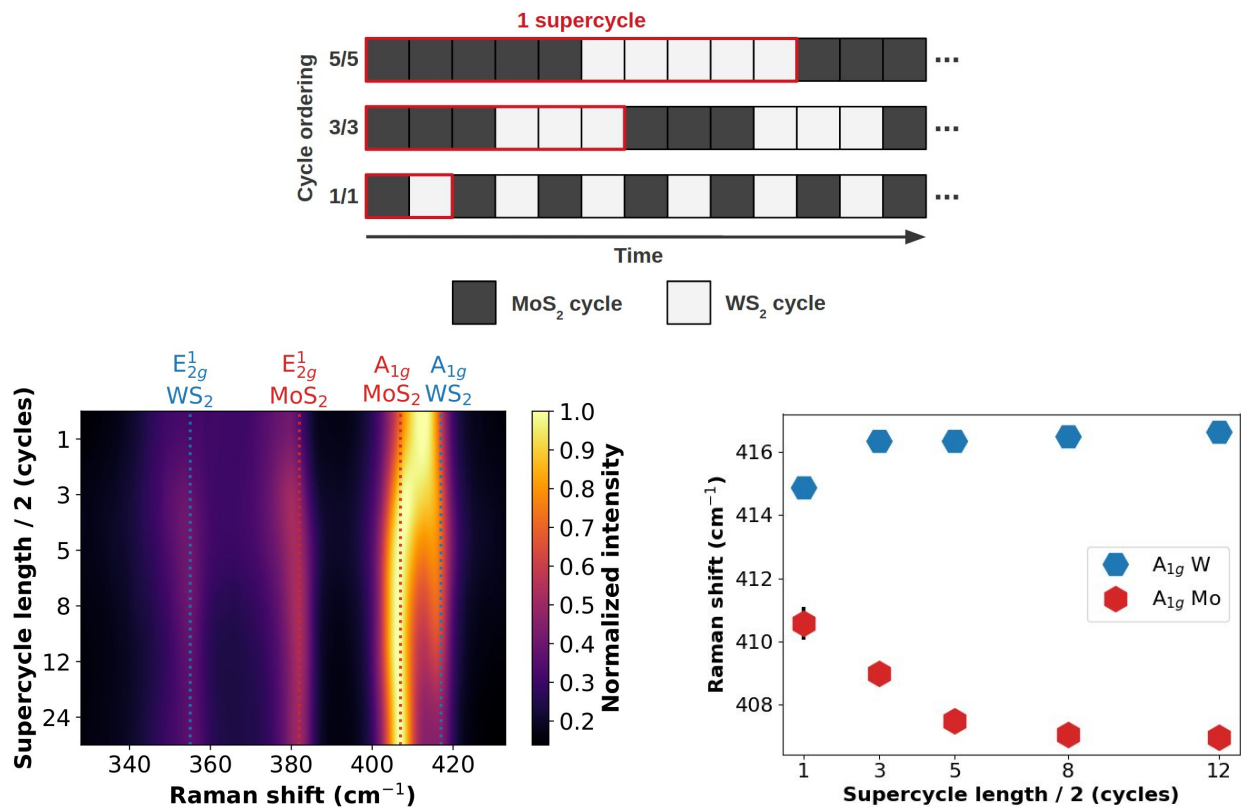


**Figure 1** Excellent control over the alloy ratio  $x$  of the  $\text{Mo}_{1-x}\text{W}_x\text{S}_2$  films as evidenced by XPS measurement (right) is achieved by manipulating the relative number of  $\text{MoS}_2$  and  $\text{WS}_2$  ALD cycles within a supercycle (left). The alloy ratio measured by XPS follows the rule of mixtures, indicating a well-behaved alloying process.



**Figure 2** The atomic ordering of the  $\text{Mo}_{1-x}\text{W}_x\text{S}_2$  is controlled independently of their alloy ratio by changing the ordering of the ALD cycles (top). Bottom: the ordering parameter shifts the lattice vibrational frequencies as evidenced by Raman spectroscopy (left). Specifically, the  $\text{MoS}_2$ -like and  $\text{WS}_2$ -like  $A_{1g}$  vibrational frequencies converge as the alloys are more well-mixed (right).