## Raman spectroscopy study of atomically thin molybdenum ditelluride

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We exfoliate atomically thin crystals of  $MoTe_2$  and investigate their phonon properties by Raman spectroscopy.  $MoTe_2$  were synthesized through chemical vapor transport. Atomically thin  $MoTe_2$  films were exfoliated mechanically from the bulk crystals onto SiO<sub>2</sub> (Fig. 1A). The Raman spectroscopy was performed at room temperature with an excitation of 532 nm. The Raman spectra of atomically thin  $MoTe_2$  show prominent peaks of the in-plane  $E^1_{2g}$  mode at ~ 234 cm<sup>-1</sup> and the out-of-plane  $A_{1g}$  mode at 171 cm<sup>-1</sup> (Fig.1B). Similar to the other dichalcogenides, the  $E^1_{2g}$  mode upshifts while the  $A_{1g}$  mode downshifts with decreasing thickness. Additionally, we observe a strong peak in atomically thin  $MoTe_2$ , which is unobservable for bulk. The peak intensity becomes significant with decreasing thickness, but the peak vanishes in a single-layer crystal (Fig. 1B). Our density functional theory calculations reveal that the peak can be assigned as a Raman inactive mode of the out-of-plane  $B^1_{2g}$  mode. The activation of the  $B^1_{2g}$  mode in the atomic layers is likely due to translation symmetry breaking along the c-axis direction.



Figure 1: (A) A typical optical image of atomically thin  $MoTe_2$  on  $SiO_2$ . (B) Raman spectra of atomically thin  $MoTe_2$ .