

## Second Harmonic Generation in Janus MoSSe Monolayer and Stacked Bulk with Vertical Asymmetry

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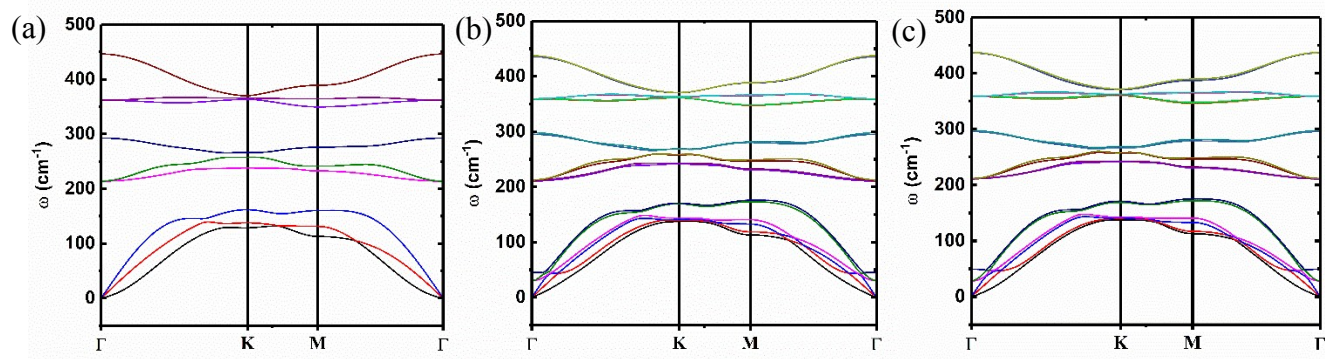


Figure S1 The Phonon Dispersion of bulk Janus MoSSe stacked patterns (a) AA, (b) AB, (c) AC

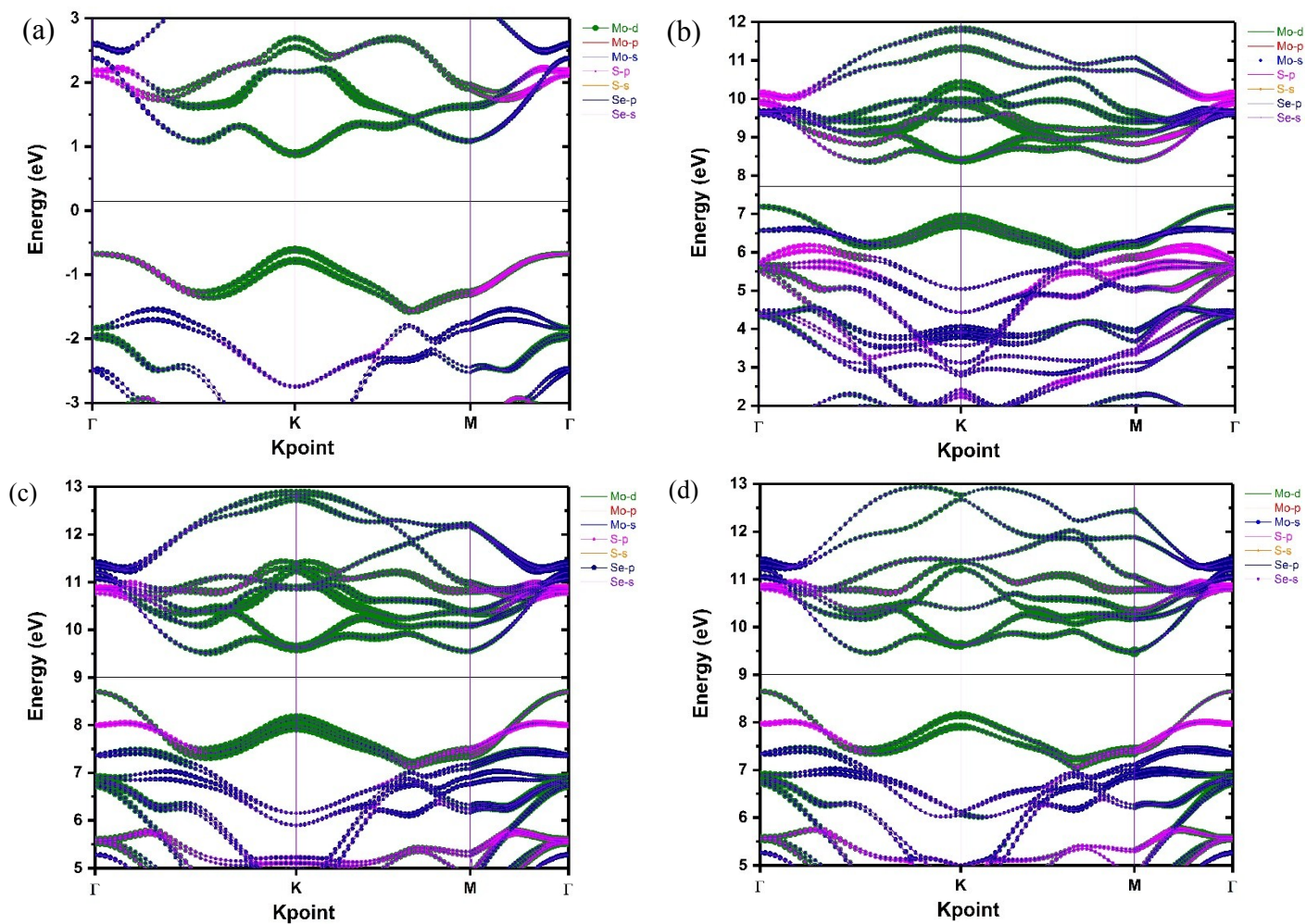


Figure S2. Projected band structures for (a) monolayer, (b) AA stack, (c) AB stack and (d) AC stack bulk Janus MoSSe. The Fermi energy for each system is labeled as the solid line across the K path

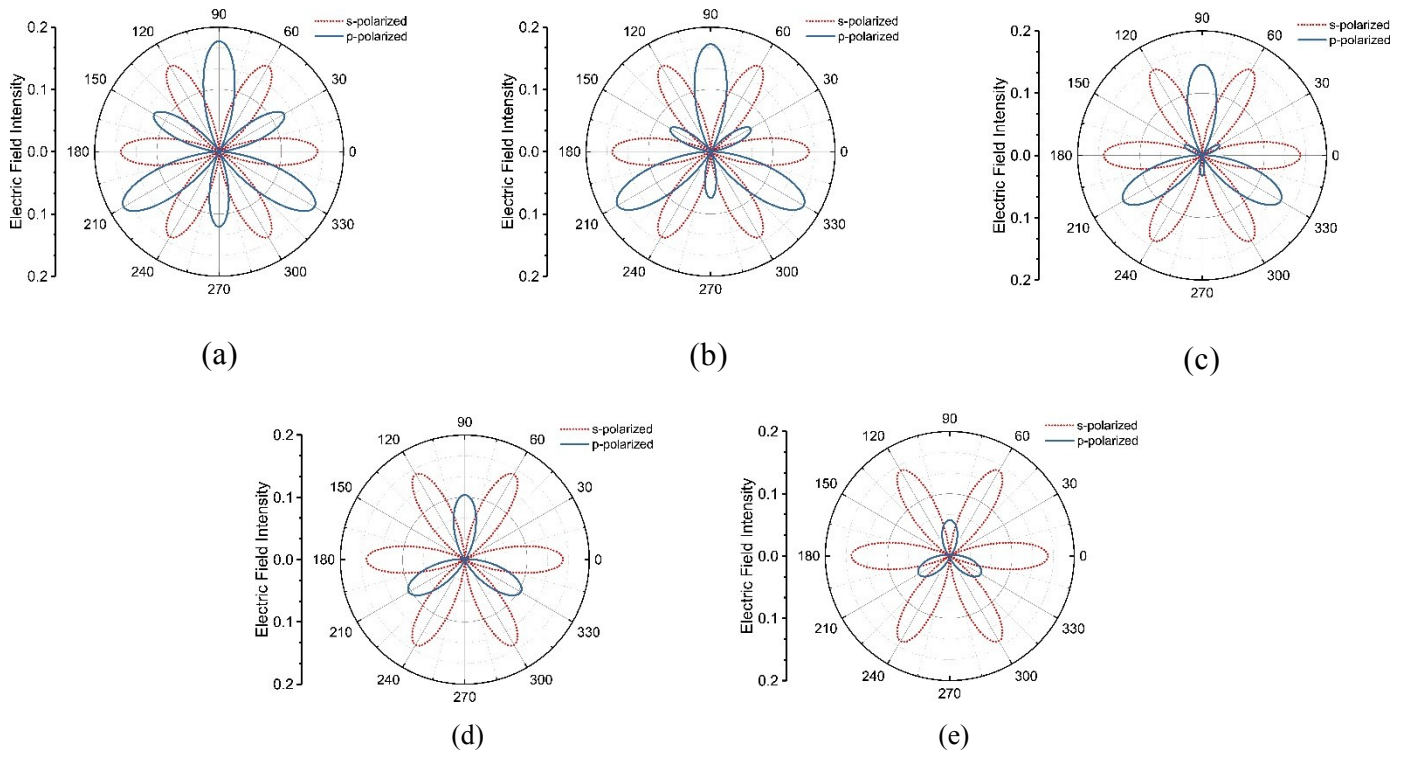


Figure S3. The polarization diagram for different incident angle (a) 15° (b) 30° (c) 45° (d) 60° and (e) 75° in the monolayer Janus MoSSe system

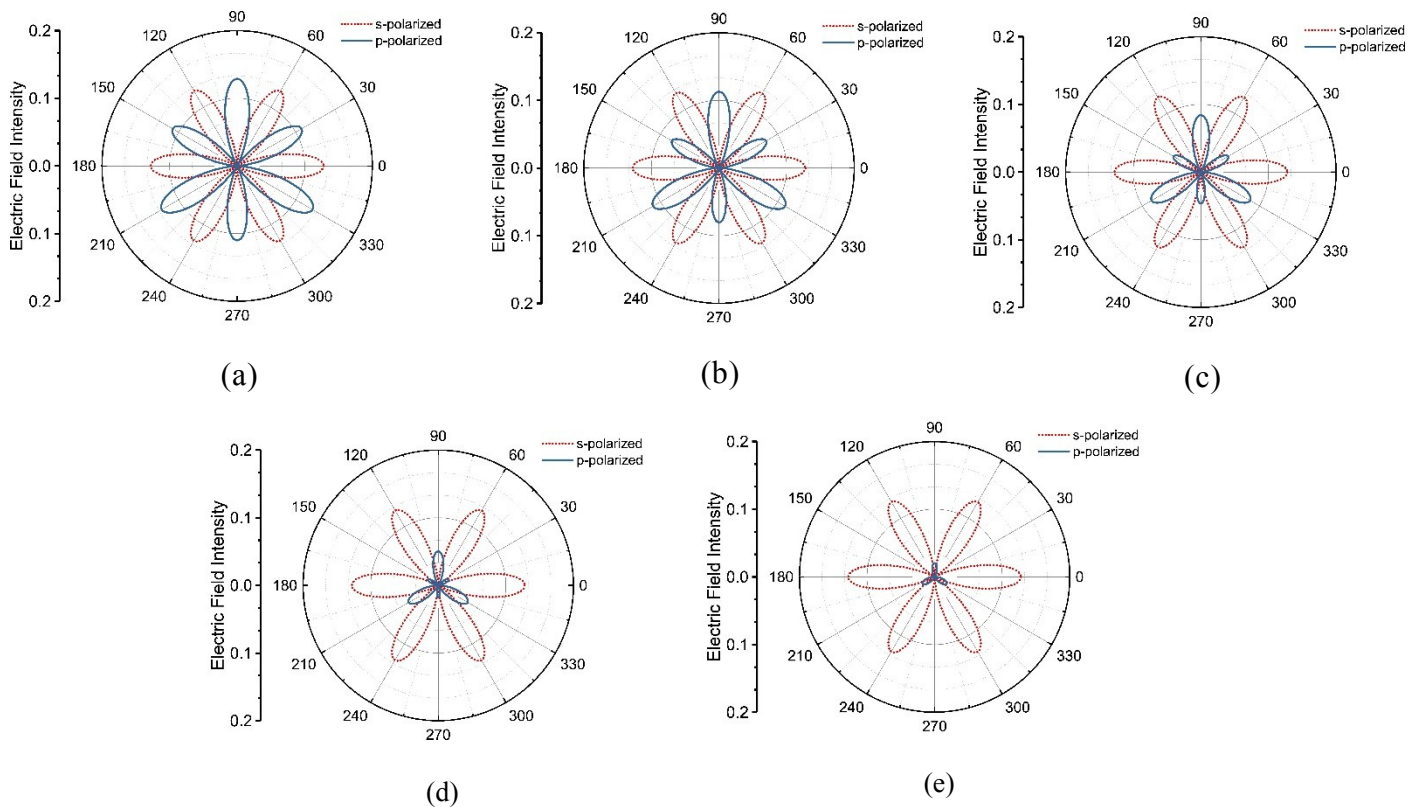


Figure S4. The polarization diagram for different incident angle (a) 15° (b) 30° (c) 45° (d) 60° and (e) 75° in the AA stacked bulk Janus MoSSe system

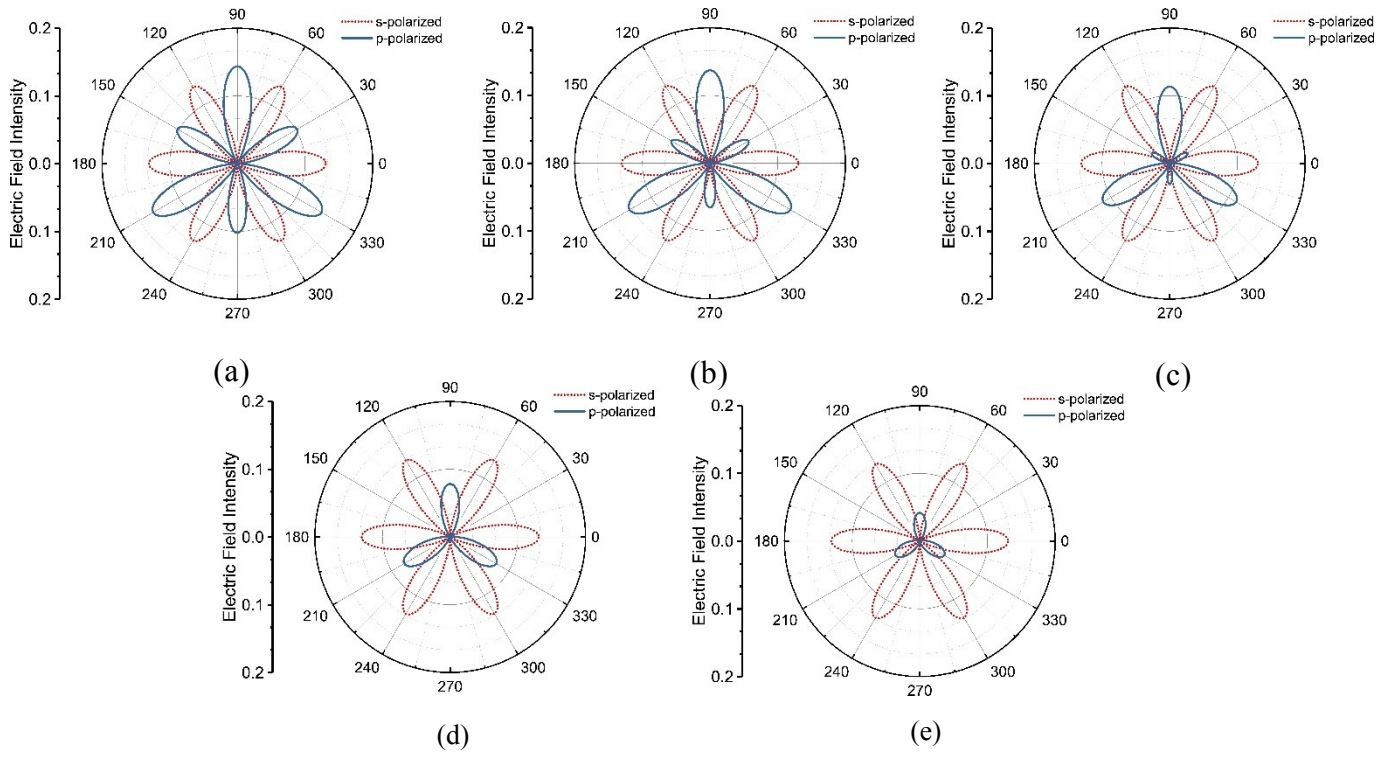


Fig. S5. The polarization diagram for different incident angle (a)  $15^\circ$  (b)  $30^\circ$  (c)  $45^\circ$  (d)  $60^\circ$  and (e)  $75^\circ$  in the AB stacked bulk Janus MoSSe system