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Second-Harmonic Generation in a Phase-Match Free Nonlinear 2D Crystal MERVIN ZHAO, ZILIANG YE, YU YE, HANYU ZHU, YUAN WANG, XIANG ZHANG, Univ of California - Berkeley — The second harmonic generation (SHG) produced from two-dimensional atomic crystals have been utilized to great effect in studying the grain boundaries and electronic structure of such crystals. However, the SHG in many transition metal dichalcogenides (TMDCs) only occur in odd numbered layers due to their noncentrosymmetric nature, limiting the applicability of their SHG. Here, we probe the SHG from the bulk noncentrosymmetric molybdenum disulfide (MoS2). Whereas the commonly studied 2H crystal phase's antiparallel dipoles in adjacent layers give an oscillatory SH response, the parallel dipoles of each atomic layer in the 3R phase constructively interfere to amplify the second harmonic intensity. Due to this interference, we observed the phase-match free condition yielding a quadratic dependence between the intensity and layer number. Additionally, we probed the layer evolution of the A and B excitonic transitions in 3R-MoS2 using SHG spectroscopy. We find exciton splittings distinct from 2H-MoS2, resulting from the different interlayer interactions of the two polytypes.

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