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Spin-orbit splitting engineered broadband resonant second harmonic generation of artificially stacked heterostructures

Noncentrosymmetric transitional metal dichalcogenides (TMDCs) and their 3R-phase vertical heterostructures (HSs) provide an ideal platform for studying atomic-scale nonlinear optics, especially second harmonic generation (SHG).[1,2] TMDC monolayers with tunable energy gaps can be artificially stacked not only to enhance the SHG efficiency but also to broaden the spectral range for the exciton resonance.[2] Besides, investigating 2D interfacial phenomena in vertical HSs of two distinct materials are arguably intriguing in the aspect of linear and nonlinear optics. We synthesized 3R-stacked homo-bilayer, hetero-bilayer structures, comprised of monolayers of MoS_2 and its alloy $MoS_{2x}Se_{2(x-1)}$, and studied their broadband SHG properties in the distinct coupling regimes. Photoluminescence analysis on all the vertical HSs showed clear intralayer (A- and B-excitonic) transitions from each constituent layer and interlayer exciton transitions, thereby confirming the excellent optical quality of the HS system. Especially, wavelength-dependent SHG measurements on the hetero-bilayer unveiled up to 4 times stronger SHG response over the spectral range of 550 nm to 780 nm. Our proof-of-concept study indicates that the spectral range for efficient SHG can be engineered by controlling the Se concentration in the $MoS_{2x}Se_{2(x-1)}$ layers in well-aligned vertical HS systems, which can tune the spin-orbit-split A- and B-excitons as well as the bandgap of each constituting layer. The strengthening and widening effects of SHG are not simply interpreted as the superposition of resonant SHG across the A- and B-exciton levels from the constituent layers. Nonetheless, our results demonstrate the feasibility of artificial strong second-order nonlinear optical materials working over a broad spectral range by combining MoS_2 with different $MoS_{2(1-x)}Se_{2x}$ alloys.

References

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Figures

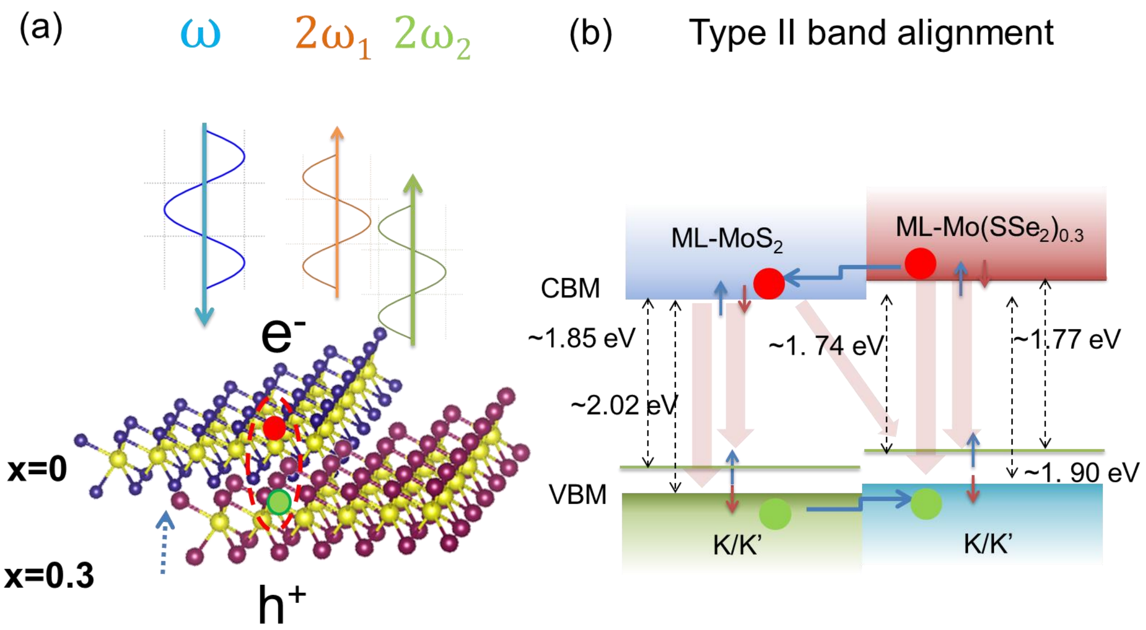

Figure: (a) Schematic for second harmonic generation in a strongly coupled vertical heterostructure comprised of monolayer (ML) MoS_2 and $MoS_{2(x-1)}Se_{2x}$, where x is the selenium doping concentration (b) Illustration showing their relative type II band alignment. Conduction band minima and valence band maxima are denoted by CBM and VBM, respectively. Both intralayer A/B-exciton transition and interlayer exciton transition are indicated by pink arrows.