Enhanced Tunable Second Harmonic Generation from Twistable Interfaces and Vertical Superlattices in Boron Nitride Homostructures

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Broken symmetries induce strong even-order nonlinear optical responses in materials and at interfaces. Unlike conventional covalently bonded nonlinear crystals, van der Waals (vdW) heterostructures feature layers that can be stacked at arbitrary angles, giving complete control over the presence or lack of inversion symmetry at a crystal interface. Here we report highly tunable second harmonic generation (SHG) from nanomechanically rotatable stacks of bulk hexagonal boron nitride (BN) crystals, and introduce the term twistoptics to describe studies of optical properties in twistable vdW systems. By suppressing residual bulk effects, we observe SHG intensity modulated by a factor of more than 50, and polarization patterns determined by moiré interface symmetry. Finally, we demonstrate greatly enhanced conversion efficiency in vdW vertical superlattice structures where multiple BN crystals are joined by symmetry-broken interfaces. Our study paves the way for compact twistoptics architectures aimed at efficient tunable frequency-conversion, and demonstrates SHG as a robust probe of buried vdW interfaces.