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Edge Nonlinear Optics on a MoS2 Atomic Monolayer XIAOBO YIN, ZILIANG YE, University of California Berkeley, DANIEL CHENET, Columbia University, YU YE, KEVIN O'BRIEN, University of California Berkeley, JAMES HONE, Columbia University, XIANG ZHANG, University of California Berkeley — The structural discontinuity at an interface yields significant electric dipolar contributions to the surface optical nonlinearity, making the highly surface- and molecular-specific second-harmonic spectroscopy an indispensable tool for non-invasive study of surface sciences. Not only does it measure dipolar width across interfaces, but it also probes real-time dynamics of surface, such as atomic reconstructions, charge transfer and molecular conformational transitions. Here we study experimentally the second-order nonlinear optics on the one-dimensional edges of hexagonal molybdenum disulfide (MoS2) atomic membranes. The broken inversion symmetry of the atomically thin monolayer shows strong second-harmonic generation (SHG), in stark contrast to the centrosymmetric bulk material which is immune to the second order nonlinear processes. The destructive interference and annihilation of nonlinear waves from neighboring atomic membranes not only reveals the few-atom-wide line defects that stitch different crystal grains together but also allows the rapid mapping of crystal grains and grain boundaries over large areas which typically requires a cumbersome diffraction-filtered dark-field transmission electron microscope (TEM). More interestingly, this unique optical imaging technique enables the nonlinear optical detection of the electronic edge state at the atomic edges of two-dimensional crystals where the translational symmetry is broken. The observed edge resonance of SHG clearly indicates the electronic structure variation at the atomic edges that have been long suspected to be the active sites for electrocatalytic hydrogen evolutions.

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