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Layer- and Frequency-dependent Second-harmonic Generation from GaSe Atomic Crystals[1] YANHAO TANG, JOHN A. MCGUIRE, CHIH WEI LAI, Department of Physics and Astronomy, Michigan State University, KRISHNA C. MANDAL, Department of Electrical Engineering, University of South Carolina — GaSe is a layered semiconductor with an indirect bandgap at about 2.0 eV only ∼20 meV below the direct bandgap at room temperature. Atomically thin GaSe crystals are expected to exhibit an increasing bandgap. This can be probed through the strong nonlinear optical response of GaSe. We report optical second-harmonic generation (SHG) in reflection from GaSe atomic crystals of 1 to > 100 layers on a Si substrate with a 90 nm SiO_2 layer. Room-temperature measurements were performed with fundamental photon energies of 0.85 to 1.4 eV as well as at 1.58 eV. By accounting for multilayer interference, the layer-dependent SHG intensity data are fit to obtain the magnitude of the second-order nonlinear optical susceptibility, $\chi^{(2)}$. For samples thicker than ∼7 layers, we find $|\chi^{(2)}| = 80 \pm 18$ pm/V, consistent with reported values for bulk GaSe. For samples $\leq$6 layers, $|\chi^{(2)}|$ is reduced compared to that in thicker samples and shows a minimum in trilayer samples. The frequency-dependence of the SHG response suggests that this reduction of $|\chi^{(2)}|$ in the few-layer region is due to increase of the direct bandgap.

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