Giant and nonreciprocal second harmonic generation from layered antiferromagnetism in bilayer CrI_3

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Abstract:

Layered antiferromagnetism is the spatial arrangement of ferromagnetic layers with antiferromagnetic interlayer coupling. Recently, the van der Waals magnet, chromium triiodide (CrI_3), emerged as the first layered antiferromagnetic insulator in its few-layer form[1], opening up ample opportunities for novel device functionalities[2-8]. Here, we discovered an emergent nonreciprocal second order nonlinear optical effect in bilayer CrI_3. The observed second harmonic generation (SHG) is giant: several orders of magnitude larger than known magnetization-induced SHG[9-12] and comparable to SHG in the best 2D nonlinear optical materials studied so far[13-16] (e.g. MoS_2). We showed that while the parent lattice of bilayer CrI_3 is centrosymmetric and thus does not contribute to the SHG signal, the observed nonreciprocal SHG originates purely from the layered antiferromagnetic order, which breaks both spatial inversion and time reversal symmetries. Furthermore, polarization-resolved measurements revealed the underlying C_{2h} crystallographic symmetry, and thus monoclinic stacking order in bilayer CrI_3, providing crucial structural information for the microscopic origin of layered antiferromagnetism[17-21]. Our results highlight SHG as a highly sensitive probe that can reveal subtle magnetic order and open novel nonlinear and nonreciprocal optical device possibilities based on 2D magnets.

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