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Title: Magnetic, Electronic, and Structural Coupling effects in the 2D Ising Ferromagnetic Insulator VI_3

Abstract: Van der Waals magnets have recently emerged as a tunable platform for exploring a rich variety of layer-dependent magnetic phenomena arising from magnetic ground states and responses that depend upon both the number of layers and how those layers are stacked. Here, we report that atomically thin vanadium triiodide (VI_3) is an Ising ferromagnet with coupled magnetic, electronic, and structural properties. Employing reflective magnetic circular dichroism (RMCD) measurements, we found that the signal exhibits an abrupt drop when the thickness is reduced from two layers down to one. We identified that this observation is originated from the vanishing of the optical resonance owing to the absence of interlayer coupling in the monolayer. In contrast to known magnets, the Curie temperature shows an anomalous increase as layer number decreases, reaching a maximum of about 60 K in monolayers. This is about 20% higher than that of the parent bulk crystals. Polarization-resolved second harmonic generation (SHG) measurements of exfoliated flakes reveal a magnetism-independent SHG signal with quasi threefold rotation symmetry in multilayers of varying thicknesses down to trilayer that is absent in monolayer, bilayer, and pristine bulk crystals. These measurements suggest that exfoliation induces a chiral layer stacking arrangement, which modifies interlayer magnetic coupling leading to the observed layer-dependent magnetic properties. Our results show that VI_3 is a versatile system for future studies of correlated physics through engineering of its layer stacking and twist degrees of freedom.