Depth-dependent magnetic properties in the ferromagnetic van der Waals semiconductor VI_3

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Two-dimensional van der Waals (vdW) magnetic semiconductors display emergent chemical and physical properties arising from both intra-layer and inter-layer interaction, both providing a versatile toolkit for studying quantum phenomena in hetero-structures and few layers systems, as well as holding promise for novel quantum spintronic functionalities [1,2]. Among 3d transition-metal vdW, Vanadium tri-iodide has recently attracted significant attention, as due to a) the presence of both structural and magnetic transitions as a function of temperature, suggesting a relevant role of magneto-elastic interactions and b) the interplay of dimensionality with relevant interactions, such as spin–orbit coupling (SOC), where a crossover of 3D vs. 2D electronic properties is expected [1]. However, detailed experimental information on ground state electronic properties as well as on spin/orbital degrees of freedom are still lacking, mainly due to its extreme air sensitivity and challenging chemical environment. Here we present chemical and layer sensitive X-ray electron spectroscopies results supported by model calculation, where we report via PhotoElectron Spectroscopy (RESPES and ARPES) a complete characterization of the electronic ground states in Vanadium tri-iodide, showing that orbital filling drives the stabilization of a quasi-Mott insulating state at the surface, with strong influence of dimensionality effects [3]. Moreover, Temperature-dependent X-ray Absorption Spectroscopy (XAS) and X-ray Magnetic Circular Dichroism (XMCD) experiments clearly reveal a reduced dimensionality of magnetic order due to electronic correlations, providing evidence of both an unquenched orbital magnetic moment (up to 0.66(7) μB/V atom) in the ferromagnetic state and (an instability of the orbital moment in the proximity of the spin reorientation transition [4,5]. Our results have direct implications in band engineering and layer-dependent properties of 2D systems, suggesting VI_3 as a candidate for the study of orbital quantum effects in spintronics.

Keywords [optional]: Quantum Spintronics, van der Walls 2D magnetism

[1]M. Gibertini, et al., Nature Nanotechnology 14, 408 (2019).

[2]B. Huang et al. Nature 546, 270 (2017).

[3]A. De Vita et al. Nano Letters 17, 7034—7041 (2022).

[4]A. De Vita et al. Nano Letters 5, 1487–1493. (2021)

[5]R. Sant et al. J. Phys.: Condens. Matter 35 405601 (2023)