Giant valley-Zeeman coupling in the NbS_2 surface layer of $V_{1/3}NbS_2$

Phil D C King[1]

[1] *School of Physics and Astronomy, University of St Andrews, United Kingdom*
pdk6@st-andrews.ac.uk

One of the most striking properties of the transition-metal dichalcogenides (TMDs) is a locking of their quasiparticle spin to a valley pseudospin. This results from strong spin-orbit coupling and is enabled by global or local inversion symmetry breaking [1,2], providing a route to stabilise novel physical properties such as Ising superconductivity [3,4], and potentially facilitating new computing schemes via `valleytronics' [1]. Tuning of the resulting spin splittings is thus strongly desired. To date, this has been achieved via externally-applied magnetic fields [5] and by proximity coupling in van der Waals heterostructures [6], but only modest changes of the intrinsic spin-orbit splitting have been realised. Here, we investigate a monolayer-like NbS_2 layer at the surface of the V-intercalated TMD $V_{1/3}NbS_2$ using spatially- and angle-resolved photoemission spectroscopy (μ-ARPES) [7]. Our measurements and corresponding density functional theory calculations reveal that the bulk magnetic order induces a giant valley-selective Ising coupling exceeding 50 meV in the surface NbS_2 layer, equivalent to application of a ~250 T magnetic field. This energy scale is of comparable magnitude to the intrinsic spin–orbit splittings, and indicates how coupling of local magnetic moments to itinerant states of a transition metal dichalcogenide surface layer provides a powerful route to controlling their valley–spin splittings.

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