Asymmetric Magnetic Proximity Interactions in Ferromagnet/Semiconductor van der Waals Heterostructures

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Magnetic proximity interactions (MPIs) between atomically-thin semiconductors and two-dimensional magnets provide a means to manipulate spin and valley degrees of freedom in nonmagnetic monolayers, without the use of applied magnetic fields. In such van der Waals (vdW) heterostructures, MPIs originate in the nanometer-scale coupling between the spin-dependent electronic wavefunctions in the two materials, and typically their overall effect is regarded as an effective magnetic field acting on the semiconductor monolayer. Here we demonstrate that this picture, while appealing, is incomplete: The effects of MPIs in vdW heterostructures can be *markedly asymmetric*, in contrast to that from an applied magnetic field [1]. Valley-resolved optical reflection spectroscopy of $MoSe_2/CrBr_3$ vdW structures reveals strikingly different energy shifts in the K and K' valleys of the $MoSe_2$, due to ferromagnetism in the $CrBr_3$ layer. Strong asymmetry is observed at both the A- and B-exciton resonances. Density-functional calculations indicate that valley-asymmetric MPIs depend sensitively on the spin-dependent hybridization of overlapping bands, and as such are likely a general feature of such hybrid vdW structures. These studies suggest routes to selectively control *specific* spin and valley states in monolayer semiconductors.

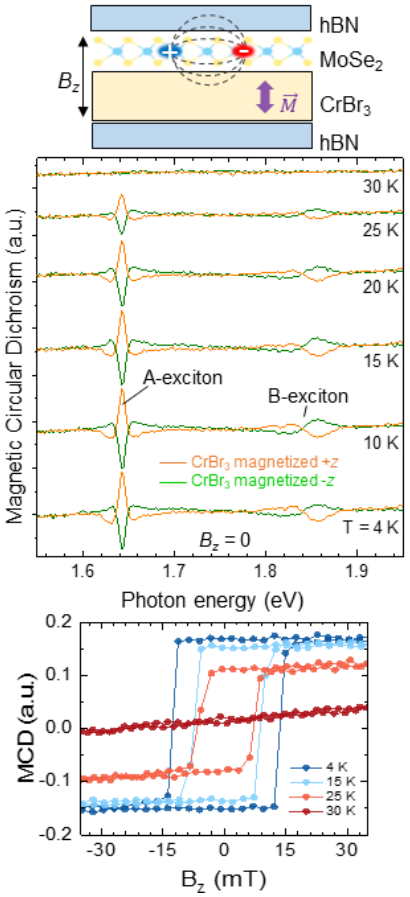

Fig. 1 $MoSe_2/CrBr_3$ structure. MCD at B_z=0 shows a strong magnetic response from the (non-magnetic) $MoSe_2$ below T_c~28K, indicating MPIs. Bottom: Hysteresis of MCD.

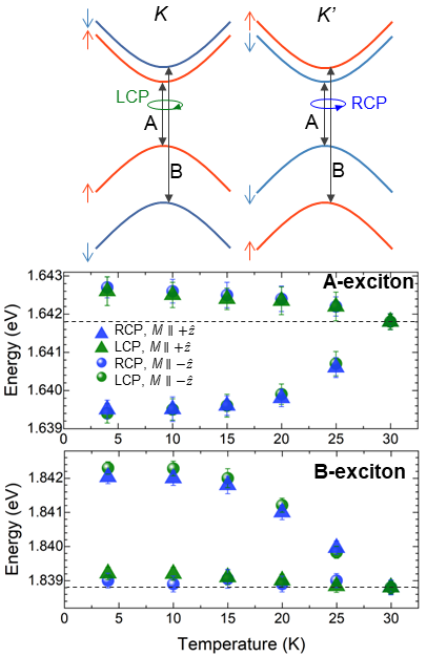

Fig. 2 Optical transitions of spin-up/down bands in K/K' valleys shift *asymmetrically* due to MPIs (in contrast to effect of real magnetic fields).

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