Title: XAFS study on the ferromagnetism in Mn-doped MoS_2 nanosheets

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Introduction: Two dimensional van der Waals crystals with long-range ferromagnetic ordering has attracted much attention, combined with their outstanding electronic and optical properties, could lead to innovation magnetic, magneto-electric and magneto-optic application. Reversible tuning magnetic properties at room-temperature in 2D materials will promote the development of spintronics, logic and memory operations and other quantum information device, Therefore, how to render the inherently nonmagnetic nature of normal MoS_2 nanosheets with robust ferromagnetism and how to tune their magnetic behaviors in a reversible and controllable way are important issues for expanding the applications of MoS_2 nanosheets in spintronics. Fortunately, Mn-doped MoS_2 nanosheets was substitutionally obtained by using the supercritical hydrothermal reaction, which provide a good prototype material to study manipulating the magnetic properties of MoS_2 nanosheets. To be able to air-stable tune the magnetic behavior of Mn-doped MoS_2 nanosheets in a reversible and controllable way, we using benzyl viologen (BV) as the surface charge transfer donor, and demonstrate an alternative way of reversible tuning ferromagnetic behavior in Mn-doped MoS_2 nanosheets.

Experimental methods: The Mn substitutionally dope MoS_2 nanosheets was achieved by a sulfur-rich supercritical hydrothermal reaction environment. The Mo K-edge and Mn K-edge X-ray absorption near-edge (XANES) spectra were measured at the 4W7B beamline of the Beijing Synchrotron Radiation Facility (BSRF). The Mn L-edge, S L-edge and Mo M-edge X-ray absorption near-edge structure (XANES) spectra were collected at the BL12B beamline of NSRL. The UPS and XPS were measured at the BL09U beamline of NSRL. All the acquired XAFS data were processed by the IFEFFIT software package.

Results and discussion: The substitutional Mn doping is convincingly evidenced by Mn K-edge extended X-ray absorption fine structure (EXAFS) and high-angle annular dark-field scanning transmission electron microscopy. The Mn-doped MoS2 nanosheets show robust intrinsic ferromagnetic response with a saturation magnetic moment of 0.05 at room temperature. Mn L-edge and S L-edge X-ray absorption near-edge structure (XANES) and first-principles calculations clarifies that the critical role of Mn 3d electronic states in mediating the magnetic interactions in MoS_2 nanosheets.

Conclusion: In summary, we have successfully doped Mn ions into MoS_2 nanosheets and obtained robust ferromagnetism at room-temperature with a magnetic moment of 0.05 emu/g. More interestingly, the ferromagnetism in the as-synthesized nanosheets could be reversibly tuned by molecular charge transfer through adsorption of BV molecules. Based on the experimental characterizations and first-principles calculations, the enhanced ferromagnetism can be attributed to the extra electrons incorporated by BV molecules which adjust the antiferromagnetism coupling between the localized 3d states of Mn and the delocalized neighboring S p states to ferromagnetism.