Surface Electronic Structure Comparison of Fe-Intercalated and 2H-TaS_2

Dejia Kong[1], Sree Sourav Das[2], Jacob St. Martin[3], Peter Siegfried[4,5], Zhiqiang Mao[6,7], Seng Huat Lee[6,7], Ian Harrison[1], Nirmal Ghimire[8,9], Mona Zebarjadi[2], Zheng Gai[10]* and Petra Reinke[3]*

[1] Department of Chemistry, University of Virginia, Charlottesville, VA, 22903, USA

[2] Department of Electrical and Computer Engineering, University of Virginia, Charlottesville, VA, 22903, USA

[3] Department of Materials Science and Engineering, University of Virginia, Charlottesville, VA, 22903, USA

[4] Department of Physics and Astronomy, George Mason University, Fairfax, VA 22030, USA

[5] Quantum Science and Engineering Center, George Mason University, Fairfax, VA 22030, USA

[6] 2D Crystal Consortium, Materials Research Institute, The Pennsylvania State University, University Park, Pennsylvania 16802, USA

[7] Department of Physics, The Pennsylvania State University, University Park, Pennsylvania 16802, USA

[8] Department of Physics and Astronomy, University of Notre Dame, Notre Dame, IN 46556, USA

[9] Stravropoulos Center for Complex Quantum Matter, University of Notre Dame, Notre Dame, IN 46556, USA

[10] Center for Nanophase Materials and Sciences, Oak Ridge National Laboratory, Oak Ridge, TN, 37831, USA

Anisotropic ferromagnetic phases can be introduced to transitional metal dichalcogenide (TMD) TaS_2 through intercalating Fe in the van der Waals (vdW) gap. By deviating from the commensurate values (x = ¼ or ⅓), the crystalline structure as well as the magnetotransport properties of the TMD system can be tuned. For instance, $Fe_{1/4}TaS_2$ has a centrosymmetric 2 × 2 structure while $Fe_{1/3}TaS_2$ has a non-centrosymmetric $\sqrt{3} \times \sqrt{3}\, R30°$ supercell structure. The magnetic Curie temperature of Fe_xTaS_2 also exhibits a strong dependence on Fe concentration. We evaluate $Fe_{0.28}TaS_2$ and 2H-TaS_2 samples using STM/Spectroscopy (STM/S) and density functional theory (DFT) to investigate the real-space intercalant electronic structure comparatively and the potential phase segregation between the two commensurate compounds. $Fe_{0.28}TaS_2$ shows a $\sqrt{3} \times \sqrt{3}\, R30°$ supercell at 77 K, whereas 2H-TaS_2 displays no apparent supercell at the same temperature. Fe vacancy defects and clusters are discovered in the intercalated surface, and their surrounding local density of states (LDOS) shows non-trivial differences at energies compared to the pristine $Fe_{0.28}TaS_2$ area, which is related to Fe orbitals contributions based on the DFT calculations.

The STM work of this research was conducted at the Center for Nanophase Materials Sciences, ORNL, which is a DOE Office of Science User Facility.