Title:	FIRST- PRINCIPLES STUDIES OF TRANSIYION METAL CHALCOGENIDES AND OXIDE HETEROSTRUCTURES FOR SPINTRONIC APPLICATIONS
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Abstract: Spintronics focuses on active control and manipulation of the spin degree of free- dom of electrons to get beyond the limitations of the semiconductor-based electronic industry. New device concepts that use an electric field or spin-polarized current result in quick and energy-efficient magnetic switching, in contrast to magnetic field-driven magnetic switching. In spintronic devices, spin polarization is controlled either by mag- netic layers or via spin-orbit coupling (SOC). The era of spintronics began with the dis- covery of giant magnetoresistance (GMR). Certain spintronics-based technologies, such as spin-transfer-torque magnetoresistive random-access memory (STT-MRAM), have been put into use on a commercial scale. The efficient use of these spintronic devices, however, still has certain challenges to overcome. Therefore, it is imperative to discover and design innovative materials featuring outstanding and distinctive spin-based mech- anisms. 2D magnetic materials in transition metal chalcogenides (TMC) provide unique physical paradigms and encourage the development of cutting-edge spintronic devices. The features of 2D TMC span a broad range, including topological phases, semimet- als, half-metals, and Mott insulators, to mention a few. Beyond ferromagnetism, there are other magnetic states such as antiferromagnets, and quantum spin liquids. Sim- ilarly, atomically tailored oxide heterostructures create a powerful platform offering whole new opportunities for electronics and spintronics. They manifest phenomena en- compassing the Rashba physics, magnetic ordering, and enhancement of SOC among others owing to the reconstruction of the spin, orbital, lattice, and charge states at the interfaces. It is possible to create oxide-based magnetic tunnel junctions (MTJ), and interface-based magnetoelectric spintronic devices using multiferroic oxides. Addition- ally, spin-orbit-based mechanisms in oxide heterostructures provide a new spintronics dimension out of which the Rashba physics emerges as a strong candidate. It is widely acknowledged that first-principles density functional theory (DFT) is an efficient method for designing functional materials and elucidating underlying mecha- nisms of experimental phenomena. This thesis focuses on using state-of-the-art first- principles calculations based on DFT to unveil the electronic, spintronic, and topolog- ical characteristics in materials picked from the strongly correlated class of TMC and oxide heterointerfaces. The aim is to identify multifunctional materials with intrinsic magnetic ordering and high SOC-driven exotic phenomena in conjunction with the low dimensionality for future low-power-consumption quantum electronics and spintronics. The thesis consists of five chapters and has been organized in the following frame- work:Chapter 1 begins with a succinct backdrop of the study. The next section of this chapter provides an overview of strongly correlated materials, including Mott-Hubbard and charge-transfer insulators. After that, it provides a quick review of 2D magnetic TMC and oxide heterostructures, two classes of strongly correlated materials. We have also included a brief history of magnetism in solids and the exchange interactions that mediate the magnetic ordering in order to provide some understanding of magnetism in solid-state systems. The concept of SOC has then been explained, with emphasis placed on the Rashba effect and its emergence in oxide heterostructures, followed by a short description of SOC-driven topological states, particularly the quantum anomalous Hall effect (QAHE). Towards the end of chapter 1, the theoretical background of various computational methods useful in understanding the calculation of results presented in this thesis has been provided. Chapter 2 infers to the ab initio study of electronic and ground state magnetic properties in bulk and layered transition metal chalcogenides. In this context, the first section of this chapter investigates the electronic structure and magnetic ordering in tri- clinic $CuSeO_3$. The electronic structure suggests that $CuSeO_3$ in the triclinic phase is a charge-transfer insulator. We find the non-collinear antiferromagnetic configuration as the ground-state magnetic ordering. This can be attributed to the hybridization of Cu-d and O-p orbitals, giving rise to Cu-O-Cu superexchange interactions. We observe a spiral spin texture in the reciprocal space with a finite out-of-plane spin component. Spintronics of such non-collinear antiferromagnetic charge transfer insulating states is a relatively new and rapidly developing field of physics. In the following section of this chapter, the vdW layered transition metal ternary chalcogenide CoAsS with strong electron correlations is explored for its potential in spintronics. The dynamically stable CoAsS monolayer via its rich magnetic and electronic phase diagram has been demon- strated as a quasi-2D magnetic material. Because of the broken time-reversal symmetry and SOC, a topological state with a non-trivial Chern number is revealed at the critical Coulomb parameter using Kubo’s approach. These results make the CoAsS monolayer very appealing for low-power-consumption nanoelectronics and spintronics. Chapter 3 focuses on the emergent phenomena in perovskite oxide heterostruc- tures based on polar perovskite oxide LaVO 3 (LVO). In this regard, the first section of the chapter explores the electronic properties in the heterostructure between a Mott in- sulator LVO and a band insulator $SrTiO_3$ (STO) with two distinct orientations (001) and (111). The electronic structure confirms that this heterointerface comprising two bulk insulators is n-type metallic in both orientations. Because of the peculiar orbital

recon- struction at the two interfaces, we notice the difference in orbital occupation as well as orbital degeneracies. Furthermore, the Fermi surface reveals fourfold/sixfold sym- xiimetry as well as the presence/absence of open orbits in the (001)/(111) orientations. Thus, unique orbital-occupation and degeneracies due to orbital reconstruction as well as symmetry of Fermi surface highlights the important role played by crystal field in determining the electronic properties in LVO/STO heterostructure. In the subsequent section of this chapter, we have examined the (001) and (111) surfaces of polar per- ovskite oxide KTaO 3 (KTO) terminated with the Ta atoms for electronic and spintronic properties. We observed the presence of 2DEG with parabolic bands near the Fermi level at both the surfaces of KTO. The distribution of 2DEG is primarily contained in the surface layers of KTO in the 5d orbitals of Ta atoms. Due to the significant SOC of Ta atoms and the breaking of inversion symmetry at the surfaces, the Rashba spin splitting is visible in the bands. The spin texture on the Fermi surface and constant energy contours corroborate the 2D nature of the Rashba spin splitting on the (001) surface, While a complicated spin structure is seen on the surface of (111)-KTO. The final section of this chapter is devoted to the emergent properties in polar-polar het- erostructure of perovskite oxides LVO and KTO depending on the crystal orientation. The charge transfer from the surface layers to the interfacial region due to the electronic reconstruction mechanism results in high carrier density 2DEG. Momentum-dependent Rashba spin splitting is seen in the electronic bands originating from the symmetry- breaking electric field. We demonstrate the orbital dependence of the Rashba effect in this heterostructure on account of orbital reconstruction at the interface. Spin-splitting is larger at the crossing regions of the d xy and d xz/yz sub-bands. Further, the spin texture of the Rashba-split bands highlights the complexity of the orientation-dependence in the LVO/KTO heterostructure. Chapter 4 is based on a theoretical understanding of high-mobility, conducting, and spin-polarized 2DEG at the EuO/TaO 2 interface of a ferromagnetic insulator EuO and non-magnetic polar perovskite KTO. The presence of proximity-induced Zeeman ex- change splitting and the Rashba spin-orbit field in the EuO/KTO superlattice warrants a thorough examination of the system's detailed electronic band structure to look for the possibility of non-trivial quantum phenomena. 2DEG is confined on the KTO side par-ticularly at the interfacial TaO 2 layer in the d xy orbitals of Ta due to orbital reconstruc- tion as confirmed by distributions of spin and charge density. The substantial exchange splitting (0.73 eV) between the two spin channels of interfacial Ta-5d xy orbitals due to the proximity effect of ferromagnetic EuO results in a spin-polarized 2DEG at the interface. The band crossing points between the majority and minority spin bands open a gap when SOC is turned on. The large spikes of the Berry curvature at the momenta of SO-induced gaps and integral Chern number demonstrate the non-trivial band topol- ogy. We observe the Rashba-type spin texture of the bands having the SO-induced gap xiiishowing single spin winding of electron spin with opposite chirality without noticing the conventional Rashba splitting in the bands. The magnetic easy axis is perpendicular to the interfacial plane. Hence our findings suggest that EuO/KTO system has potential in low-power quantum electronics, spintronics, and spin-orbitronics. The final chapter, chapter 5, concludes this thesis by summarising the key findings of the different studies presented in the individual chapters. It also provides a prospec- tive future research outlook by steering the characteristics of various 2D materials and probing further emergent phenomena at the interface of oxide heterostructures for spin- tronic applications.

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