Abstract

Since the discovery of monolayer graphene, the study of atomically-thin layered materials has grown as one of the leading issues in condensed matter physics. Electrons confined in a two-dimensional atomic layer show distinct behavior from those in three-dimensional bulk crystals. The archetypal example is the Dirac electrons in graphene, whose energy spectrum exhibits a point node at the Fermi level and linear momentum dependence near the node. The peculiar nodal structure leads to interesting transport phenomena, such as the anomalous integer quantum Hall effect and the Klein tunneling. In the past decade, however, tremendous efforts have been devoted to the quest for layered materials beyond graphene. One direction is to investigate the effect of strong spin-orbit coupling (SOC). For instance, stimulated by a theoretical proposal that the SOC gaps out the Dirac node and changes the system into a $\mathbb{Z}_2$ topological insulator, similar honeycomb monolayers of heavier elements with larger SOC, such as Si, Ge, and Sn, have been studied. Another direction is to explore the effect of electron interactions. Recent findings of magnetism in few-layer transition metal (TM) compounds with $3d$ electrons, e.g., TM trichalcogenides and TM trihalides, opened up a new field, materials science of atomically-thin magnets, where strong electron interactions of TMs play a crucial role. These advances on the SOC and electron interactions have been gained almost independently, and their cross section has not been discussed intensively thus far. Synergy between the SOC and electron interactions has recently attracted much attention as a clue for realizing topological states of matter. It will be very intriguing to investigate the synergetic effects in two-dimensional electronic states in layered materials.

In this thesis, by using *ab initio* calculations and tight-binding analyses, we theoretically investigate the layered TM compounds with $4d$ and $5d$ electrons, where the synergy between the SOC and electron interactions can be expected in their two-dimensional electronic states. We focus on the honeycomb-layered structure commonly seen in a variety of layered TM compounds, such as TM trichalcogenides and TM trihalides. In the systems, the cubic crystalline electric field from the ligands splits the d levels into energetically higher e_g and lower t_{2g} manifolds. Considering a wide range of layered TM compounds with e_g and t_{2g} electrons, we theoretically explore new electronic and magnetic properties arising from the cooperation between the SOC and electronic interactions.

First, we investigate the electronic band structure and possible magnetic properties of TM trichalcogenides with relevant e_g electrons. We find by using *ab initio* calculations that the electronic band structures of monolayer TM trichalcogenides with group 10 TM elements exhibit multiple Dirac nodes near the half-filled level

in the e_g manifold. The multiple Dirac nodes originate from a hidden honeycomb superstructure composed of the third-neighbor transfers arising from the strong hybridization between the e_g orbitals and the ligand p orbitals. In addition, we show that the Dirac nodes are gapped out by the interplay between the SOC and trigonal lattice distortions, and hence, the Dirac gap can be tuned by applying tensile strain as well as chemical substitutions. Furthermore, we also show that electron interactions and carrier doping can turn the multiple Dirac semimetal into the quantum anomalous Hall insulator with a high Chern number. We also extend our analysis to the bilayer and bulk systems, where the peculiar band structures appear as the vestiges of the multiple Dirac nodes in the monolayer systems.

Next, motivated by the finding of multiple Dirac nodes, we perform a systematic analysis of the topology of e_g band structures in the monolayer honeycomb systems. From the tight-binding analysis, we find that the ratio between nearest- and third-neighbor transfers strongly depends on the trigonal distortion of the ligand octahedra. Analyzing a tight-binding model with the nearest- and third-neighbor transfers, we find a variety of band crossings at the half-filled level of the e_g orbitals, which include multiple Dirac nodes, semi-Dirac point nodes, quadratic band crossings, and a line node. These band crossings are also gapped out by the interplay between the SOC and trigonal lattice distortions, and the system turns into various topological phases distinguished by the spin Chern numbers. Our results provide a comprehensive understanding of the existing *ab initio* results. Furthermore, we also perform *ab initio* calculations for candidate materials to demonstrate the flexible band engineering found in the tight-binding results.

Finally, we investigate the interplay between the SOC and electron interactions in the t_{2g} electrons on the similar honeycomb-layered structures. We focus on the possibility of peculiar anisotropic exchange interactions, called the Kitaev interactions, which have recently attracted much attention as a source of quantum spin liquids. We theoretically propose a new scenario to realize the Kitaev interactions by introducing polar asymmetry in the crystalline structure. By using the second-order perturbation theory for a multiorbital Hubbard model, we find that the asymmetric superexchange process in the polar structure gives rise to unconventional antiferromagnetic-type Kitaev interactions. Moreover, we propose by using *ab initio* calculations that Ru trihalides with multiple anions potentially exhibit the antiferromagnetic Kitaev interactions following our scenario.

Our results enlighten several new directions in the study of $4d$ and $5d$ honeycomb-layered TM compounds. Our findings for the e_g-orbital systems suggest that the e_g-orbital manifold offers an intriguing playground for the band topology as the e_g-orbital transfers can be flexibly tuned by lattice distortions as well as chemical substitutions. On the other hand, our results for the t_{2g}-orbital systems could contribute to materials science of exotic magnetism such as quantum spin liquids. We believe that the results in this thesis stimulate further theoretical and experimental exploration of atomically-thin layered materials.