Abstract

Magnetism in low-dimension has garnered great interest in fundamental research as well as their phenomenological aspects, such as topological phases, electric-field manipulation, spin detection and spin injection [1-3]. Particularly, 2D magnets have drawn attention due to their ability of seamless integration of multi-layered heterostructure via tuning the strain, gating electrical properties [4]. These 2D magnets offer new physical significance in the context of spin fluctuations, which can be strongly enhanced [5-7]. With the advent of 2D magnets, many opportunities include such as control and modulation of magnetic, electronic and topological phases suitable for development of various nanoscale devices.

In pristine form, the atomically thin layered materials show limited functionalities. Instead, spin related phenomena is induced extrinsically for tuning its functionality in atomically thin layered vdW heterostructure *via* engineering defects such as vacancies, adatoms, grain or edge boundaries [8, 9], introducing magnetic dopants through intercalation or substitution [10], coupling with magnetic substrates [11]. However, it is a challenging task to create long-range magnetic ordering by externally inducing local moments using strong exchange interaction in these methods [12]. In this regard, origination of intrinsic magnetism in parent 2D lattice and designing vdW heterostructure with 2D magnets *via* interfacial phenomena will set the stage for realizing the underlying physics of spin related phenomena in technological front.

Reduced dimensionality and interfacial phenomena have often altered new exciting physical paradigms and recommends distinctive approach to precisely design an unusual device. To unveil exotic quantum phases, atomically thin two-dimensional (2D) vdW heterostructure, embraces control and tuning of various physical states by coupling with peripheral perturbation such as

pressure [13], photon [14], gating [15], Moire pattern [16] and proximity effect [11]. Among them, proximity effect is the imperative tools for creating interfacial phenomena by interacting with the neighbouring material. Proximity effect tunes the physical properties of pristine system by inducing spin-polarization and exchange coupling across the interface [17]. In this regard, realizing spin related interfacial phenomena via proximity effect in 2D vdW quantum systems will be promising for various technological advances in nanoelectronics. Spin induced phenomena via magnetic proximity effect (MPE) in 2D vdW heterostructure is now of particular area of interest due to long electron relaxation length, intrinsic magnetic ordering and spin orbit coupling (SOC). Therefore, realization of interfacial spin related phenomena *via* MPE is one of the current active research interests in the scientific community.

The present thesis mainly focuses on realization of spin related interfacial phenomena via magnetic proximity effect (MPE), which will administer electronic structure, magnetic properties and topological phases of 2D vdW heterostructure under the framework of *ab initio*-based simulations by respective Wannier-tight binding (WTB) method along with pertinent experimental cooperation. *Ab initio*-based density functional theory (DFT) is a legitimate tool for structure-property correlation simulation using pseudopotential wave basis set. Alongside DFT, WTB is a semi-empirical method primarily used to calculate electronic structure, surface states and single-particle Bloch electron states of the system. In this regard, DFT and WTB together provide an ultimate platform for comprehensive understanding of the properties of 2D vdW heterostructure systems. Few relevant facets of magnetic proximity induced spin related interfacial phenomena are briefly included in the following chapters of the thesis described below:

Chapter 1: This chapter briefly introduces the conventional techniques of extrinsically inducing local magnetic moments in non-magnetic systems. The limitation of this conventional technique can be overcome by introducing the spin-related interfacial phenomena via MPE to construct vdW quantum systems for modelling novel device prototype. At the end of the chapter, the motivation behind the work and objective of the thesis are presented.

Chapter 2: This chapter highlights the details of the computational methods used in this thesis to carry out the atomistic simulation. *Ab initio* based DFT, semi-empirical based Wannier-tight binding techniques are followed for fulfilling the objective of this thesis.

Chapter 3: This chapter shows MPE along with the external perturbative effect such as electric field in 2D vdW graphene (Gr)-Chromium tribromide ($CrBr_3$) heterostructure particularly for modelling single gate field effect transistor (FET) device prototype.

Chapter 4: This chapter mentions the MPE mediated topologically trivial and non-trivial phases in Gr-$CrBr_3$ vdW heterostructure for realizing valley contrasting quantum anomalous Hall effect (QAHE).

Chapter 5: This chapter summarizes the electrical conductivity on Gr-$CrBr_3$ vdW heterostructure system considering ferromagnetic (FM) configuration, which will provide an opportunity to realize the longitudinal and transverse thermoelectric effect.

Chapter 6: This chapter explains the proximity effects in 1T'WTe_2-$CrBr_3$ vdW heterostructure to realize the spin-polarization for application of spintronic device.

Chapter 7: This chapter displays the comprehensive conclusion extracted from all the chapters mentioned above discussing electronic, magnetic, topological, and transport properties in 2D vdW heterostructure via MPE. This chapter also have a particular emphasis on future prospects to explore more idea about the study in this thesis.

References:

[1] Park, J. -G. Opportunities and challenges of two-dimensional magnetic van der Waals materials: magnetic graphene? *Journal of Physics: Condensed Matter*, 28:301001, 2016.

[2] Roldán, R., Castellanos-Gomez, A., Cappelluti, E. and Guinea, F. Strain engineering in semiconducting two-dimensional crystals. *Journal of Physics: Condensed Matter*, 27:313201, 2015.

[3] Zhong, D., Seyler, K. L., Linpeng, X., Cheng, R., Sivadas, N., Huang, B., Schmidgall, E., Taniguchi, T., Watanabe, K., Mcguire, M. A., Yao, W., Xiao, D., Fu, K-M. C. and Xu, X. Van der Waals engineering of ferromagnetic semiconductor heterostructures for spin and valleytronics. *Science Advances*, 3:e1603113, 2017.

[4] Cortie, D. L., Causer, G. L., Rule, K. C., Fritzsche, H., Kreuzpaintner, W. and Klose, F. Two-dimensional magnets: forgotten history and recent progress towards spintronic applications. *Advanced Functional Materials*, 30:1901414, 2020.

[5] Huang, B. et al. Electrical control of 2D magnetism in bilayer CrI_3. *Nature Nanotechnology*, 13:544, 2018.

[6] Samarth, N. Magnetism in flatland. *Nature*, 546:216, 2017.

[7] Lado, J. L. & Fernández-Rossier, J. On the origin of magnetic anisotropy in two dimensional CrI_3. *2D Materials* 4:35002, 2017.

[8] Gonzalez-Herrero, H. et. al. Atomic-scale control of graphene magnetism by using hydrogen atoms. *Science*, 352:199, 2016.

[9] Uchoa, B., Kotov, V. N., Peres, N. M. R. & Neto, A. H. C. Localized magnetic states in graphene. *Physical Review Lett*er, 101:026805, 2008.

[10] Fan, M., Wu, J., Yuan, J., Deng, L., Zhong, N., He, L., Cui, J., Wang, Z., Behera, S. K., Zhang, C., Lai, J., Tour, M., Lou, J., Chu, C. W., Sun, D. and Ajayan, P. M. Doping nanoscale graphene domains improves magnetism in hexagonal boron nitride. *Advanced Materials*, 31:1805778, 2019.

[11] Hallal, A., Ibrahim, F., Yang, H., Roche, S. and Chshiev, M. Tailoring magnetic insulator proximity effects in graphene: first-principles calculations. *2D Materials*, 4:025074, 2017.

[12] Nair, R. R., Sepioni, M., Tsai, I. L., Lehtinen, O., Keinonen, J., Krasheninnikov, A. V., Thomson, T., Geim, A. K. and Grigorieva, I. V. Spin-half paramagnetism in graphene induced by point defects. *Nature Physics*, 8:199, 2012.

[13] Fulop, B. et al. New method of transport measurements on van der Waals heterostructures under pressure. *Journal of Applied Physics*, 130:064303, 2021.

[14] Lyons, T. P. et al. Interplay between spin proximity effect and charge-dependent exciton dynamics in $MoSe_2/CrBr_3$ van der Waals heterostructures. *Nature Communications*, 11:6021, 2020.

[15] Lazic, P., Belashchenko, K. D. and Zutic, I. Effective gating and tunable magnetic proximity effects in two-dimensional heterostructure. *Physical Review B*, 93:241401, 2016.

[16] Tong, Q., Chen, M. and Yao, W. Magnetic proximity effect in a van der Waals Moire superlattice. *Physical Review Applied*, 12:024031, 2019.

[17] Vasili, H. B. et al. Magnetoresistance in hybrid $Pt/CoFe_2O_4$ bilayers controlled by competing spin accumulation and interfacial chemical reconstruction. *ACS Applied Materials and Interfaces*, 10:12031, 2018.