Chapter 1

Introduction

1.1 Magnetism in low-dimensions

Atomically thin two-dimensional (2D) materials rarely show exciting features as we require them in pristine form. Instead, they offer new physical paradigm when these pristine materials were essentially modified by forming van der Waals (vdW) heterostructure. These modifications were achieved for 2D vdW crystals via rapid fabrication, which negotiates the controlled levels of chemical, physical, optical and electrical properties having applications from transistors and energy storage to environmental remediation [1–3]. Till date, the modifications in two-dimensional crystals like semiconductor, superconductor, semimetal, metals, topological insulators, the magnetic semiconductor is mostly seen via doping, introducing magnetic species or inducing defect, coupling with substrates, where extrinsically the impurities were introduced to amend and modify their properties [4–7]. These modifications suggest a new path to realize intrinsic magnetic behaviour in vdW heterostructure, where dynamics of spin are predicted to be significantly enhanced [8, 9]. Extrinsically introduced dopants or defects in 2D vdW crystals have been commonly employed to alter the surface and interfacial phenomena leading to produce exciting outcomes. Pristine materials with low, moderate and high concentration of dopants or defects act like an insulator, superconductors and traditional metals, respectively [10]. Combining 2D materials with dopants or defects with different functionalities has led to the formation of new and unique

M Bora and P Deb, *Magnetic proximity effect in two-dimensional van der Waals heterostructure.* J. Phys. Mater., 4:034014, 2021 heterostructures for investigating novel physical phenomena for developing low-power nanoelectronic devices.

Doping in usual semiconductors like hexagonal boron nitride (h-BN) by graphene quantum dots (GQDs) can infer unique physicochemical properties. The insertion of GQDs also enhances the crystal structure of h-BN, characteristics of semiconductor and bonding among carbon (C)–nitrogen (N), carbon (C)–boron (B) at the interface leads to manipulation of spin and also induces ferromagnetism in h-BN [11]. Essentially, these can be beneficial objectives to introduce a smooth incorporation for nanoelectronics, spin-logics, energy storages. Magnetic impurities such as Mn, Fe, Co and Ni-doped 2D layered materials provide controlled exchange interaction by altering the perpendicular magnetic anisotropy and Curie temperature, T_C [12], also unveil the new techniques to check the direction of magnetization [13]. Coupling of 2D materials with the magnetic substrate is a promising system for inducing magnetism by modulating external perturbative effects like light [14], gating [15] and electric field [16].

However, even with a successful realization and modifications of extrinsically induced phenomena in 2D atomic crystals, they have certain restrictions in the context of doping concentration, which results in disordered spins and notably reduces the mobility of electrons in 2D layered crystals [17]. Also, low solubility of dopant or defect concentration challenges the growth and agglomerates to nanocluster which complicates the determination of physical and chemical properties [18]. Moreover, externally induced magnetic moments in layered atomic crystals have been realized by doping, defects and coupling with magnetic substrates. In this scheme, it is challenging to induce long-range interaction between extrinsic injections of local spins via robust exchange interaction [5] and 2D materials coupling with magnetic substrate hinders the development of advanced spin related device by inhibiting vdW heterostructure. However, after successfully examining the novel properties in vdW heterostructure by vacancies, adatoms, doping, defects, coupling with magnetic species or substrates [4–7], it is important to reconsider 2D vdW heterostructure and analyse long-range spin correlation.

To overcome the limitations of extrinsically induced phenomena, an extraordinary path for designing materials has emerged from proximity effects. Proximity effect is the most crucial tools required for manipulating spintronics [19, 20], superconductor [21, 22], topologically non-trivial phenomena [23, 24] and excitonic [23]. This effect is highly flexible to interfacial phenomena when two or more dissimilar materials are integrated and brought near to each other because of proximity effect. The culmination of MPE tends to modify functionalities and difference in the exchange interaction near the interface [25, 26]. Moreover, proximity effect broadly transforms a wide range of materials, where the effective Hamiltonian of the interface between the two monolayers has different functionalities than that of the two individual monolayers. The first instinct about proximity effect was realized from superconducting materials, known for 86 years [27]. The properties of superconducting materials can penetrate from one region into a usual neighbouring region, which may not have superconducting nature. Similarly, MPE was realized almost three decades ago by Zuckermann [28]. He explained theoretically that a thin film comprising weak intrinsic ferromagnetic behaviour brought in close vicinity to a thick film of a paramagnetic material and has observed non-zero Curie temperature. The method that was employed to understand MPE is by solving the Landau–Ginzburg theory of phase transition. The following equation scrutinize as

$$M(\vec{r}) = \int d^3r' \, U(\vec{r})\chi T(\vec{r}-\vec{r}';0)M(\vec{r}') - U(\vec{r})M^3(\vec{r})\sum_{\omega} G_p^0(\omega) \quad (1.1)$$

Here, $M(\vec{r})$ depicts local magnetization at $\vec{r}$, $U(\vec{r})$ represents Hubbard exchange term, $\chi T(\vec{r};0)$ is the static $\vec{r}$ dependent magnetic susceptibility of non-interacting conduction electrons and $G_p^0(\omega)$ displays the momentum propagator p of conduction electrons. However, this approach was appropriate for superconducting material and not reliable for magnetic interfaces. Therefore, the exact theory was realized by Bergmann [29] in 1978 explained by experimentally depositing a thin film of Ni, Co and Fe over a paramagnetic metallic substrate (Pb_3Bi). This interpretation implied that the magnetization value in the initial few FM layers nearest to the PM is considerably different from the bulk counterpart. Therefore, Bergmann's experimental measurement was relevant for understanding the fundamental problem of the relation among magnetism and dimensionality, also verifies the existence of MPE. Till date, a few studies have been explored using dopants or defects limiting its functionality. In this regard, a synergy needs to be established between the optimized dimensionality with combining capabilities among the layers leading to vdW heterostructure for various applications in spintronics and valleytronics

For employing proximity effect as an essential tool to alter the physical quantities by designing new interface with enhanced functionality, it is important to consider advanced high-throughput computational simulation and mathematical algorithms to develop and understand the physical insights of interfacial phenomena in vdW heterostructures. Inspite of other advanced technique such as quantum Monte Carlo (QMC) calculation [30], density functional theory (DFT) still persists as a powerhouse for investigating huge and more complicated systems exhibiting interfacial phenomena with pronounced atomic relaxations for graphene, 2D ferromagnet, dichalcogenides and related vdW heterostructure systems. Computationally, solving quantum mechanical phenomena by scaling down atomic size restricts their applications to more complex systems such as vdW heterostructures are in nanometer range. The computational techniques solve quantum mechanical phenomena such as DFT [31, 32] and Wannier tight-binding Hamiltonian method (WTBH) under the formulation of Wannier basis sets [33] are favored to elucidate electronic, topological, magnetic, and transport properties in layered quantum systems by altering computational feasibility and transferability with consecutive evolution of chemical accuracy [34]. In this regard, DFT is quite satisfactory *ab initio*-based method for calculating structure-property correlation using pseudopotential plane wave basis whereas WTBH method considers Wannier basis sets in real space. As a consequence, DFT and WTBH together form an ideal platform to realize interfacial phenomena in complex and real 2D systems. In this regard, advances in scaling down upto the monolayer limit and heterostructures in view of proximity effects, resembles to modify broad variety of 2D vdW materials which has the possibility to surpass the restrictions of doping, defects, coupling with substrates, adatoms, grain boundaries, edge effects and chemical functionalization. Meanwhile, the MPEs involve transfer of an ordered state to the nearest neighbour region, where the electronic structure was not strongly affected initially at pristine form. Later, this terminology has pertained more extensively which includes proximity-induced spin–orbit interaction (SOI) or topologically non-trivial phases [35, 36]. Figure 1.1 gives an overview of the diverse area for interfacial engineering via MPE:

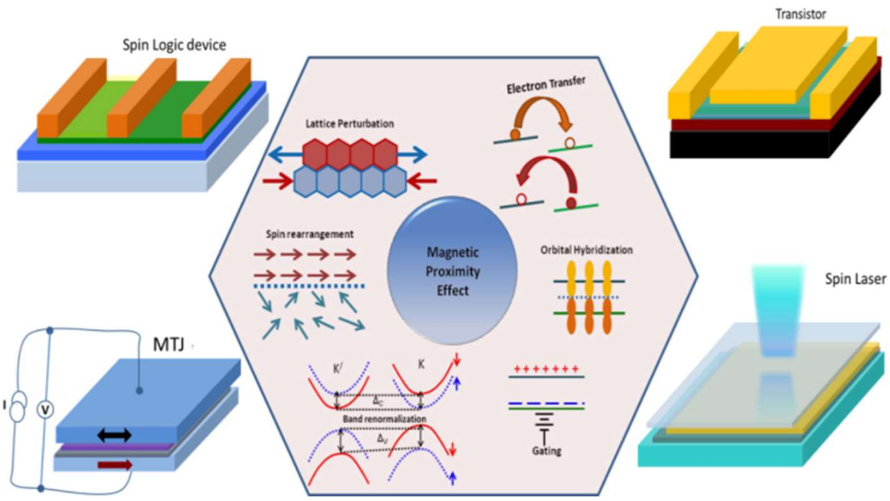

Figure 1.1: Displaying the scheme of MPE which can be realized, by keeping adjacent material in close vicinity via different mechanisms acquiring different functionalities applicable in designing various spin-electronic devices.

(a) The lattice perturbation in the interface changes the stacking configuration from in-plane to out-of-plane for tuning the magnetic behaviour in layered vdW heterostructure.

(b) The electron transfer in the interface varies the concentration of electrons and orbital nature in atomically thin vdW nanomaterials, leading to the variation in the electronic property.

(c) Lattice symmetry breaking tunes the orbital physics and subsequently the rearrangement of spin is induced in the interface due to MPE.

(d) The orbital hybridization in the interface due to MPE modifies the resultant magnetic property by influencing the electronic properties and orbital character of vdW heterostructure.

(e) The band structure in the interface may renormalize when one layer is kept in close proximity with a material of matching lattice constant. Band renormalization in atomically thin vdW heterostructure can be induced by the dielectric screening of neighbour material.

(f) The electrical control via gating plays an important role when 2D magnet will be in close vicinity with the nonmagnetic material, where interfacial polarization is intrinsically related to the vdW heterostructure.

1.2 van der Waals quantum systems

1.2.1 Graphene

An allotrope of carbon is known as graphene consist of an array of single-atomic layer organized in a honeycomb lattice structure. The individual atom in graphene sheet is bonded to three nearest neighbour atoms strongly connected by σ-bond providing one electron to valence band, which expands over the entire sheet. In this regard, the electrons present in graphene are fully confined in 2D plane irrespective of bulk 3D systems. The electronic bands of graphene disclose the linear dispersion relation between conduction and valence bands connecting at high-symmetry points in Brillouin zone as shown in figure 1.2. In contrary, the conventional bulk materials exhibit quadratic dispersion relation, with overlapping states between conduction band minimum and valence band maximum. The electronic properties can be easily tuned by gating, which shifts the Fermi level at charge neutrality extrema.

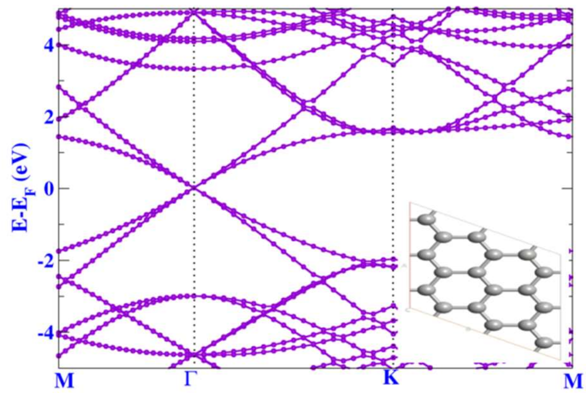

Figure 1.2: Displays the electronic band structure for pristine monolayer graphene.

1.2.2 Chromium tribromide ($CrBr_3$)

Chromium tribromide ($CrBr_3$) is a 2D vdW material comprising a semiconducting nature with a finite band gap of 0.3-2.1 eV. In monolayer limit, the intrinsic origin of $CrBr_3$ exhibit long range magnetic ordering with a Curie temperature (T_C)~37K. The layers of $CrBr_3$ are stacked vertically via weak vdW force, which can further be fabricated and exfoliate into atomically thin sheet. $CrBr_3$ is also identified as Mott insulator [37], where the contribution of d-d orbital provides a Coulombic energy U to investigate the energy gap [38]. The atomic configuration of $CrBr_3$ is displayed in figure 1.3.

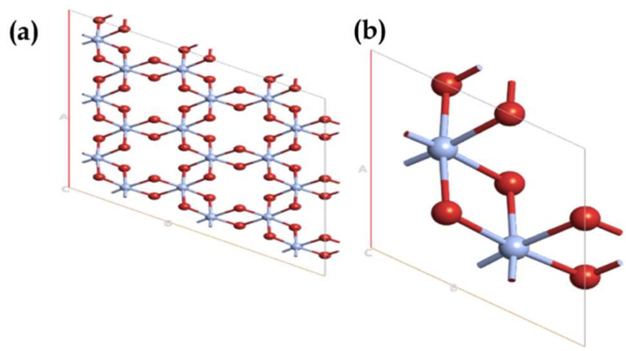

Figure 1.3: Displays the atomic configuration of pristine monolayer $CrBr_3$. (a) 2x2x1 supercell (b) unit cell of the monolayer system. The blue and red spheres depict chromium (Cr) and bromine (Br) atoms, respectively.

1.2.3 Transition metal dichalcogenides (TMDs)

Transition metal dichalcogenides (TMDs) are another class of 2D materials with crystal formula MC_2, where M and C denotes metal and chalcogen atoms, respectively [39]. TMDs are three atomic layer thick with intralayer covalent bonding and interlayer bonded by weak vdW forces. Moreover, few TMDs contain metallic as well as Weyl semimetal type nature such as 1T′-WTe_2 (Tungsten Di-telluride). It exhibits excellent magnetoresistive behaviour and quantum spin Hall (QSH) at low temperature [40]. The prefixes 2H and 1T′ refers to the stacking pattern of transition metal atoms exaggerate on top of one another, while 1T′ distorted phase predicts most experimentally favoured condition and protects inversion symmetry. The crystal structure (as a inset in figure 1.4) and electronic band structure of monolayer 1T′-WTe_2 is shown in figure 1.4.

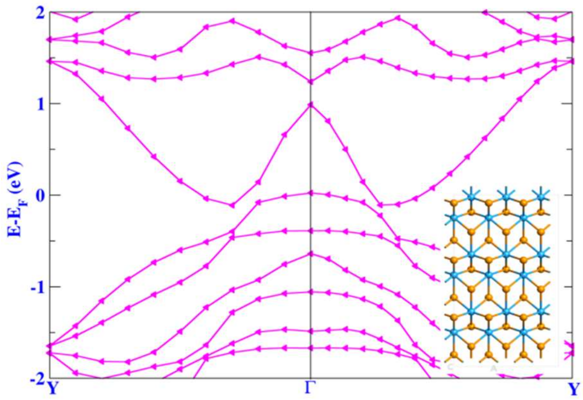

Figure 1.4: Displays the electronic band structure of pristine monolayer 1T′-WTe_2. The inset shows 1T′-WTe_2 3x3x1 supercell of monolayer system. The orange and blue spheres represent tungsten (W) and tellurium (Te), respectively.

1.3 From Bulk to proximitized material

In general, the dimension in bulk structures widely attribute to large characteristic scales, where time-reversal symmetry naturally breaks in proximitized materials. In the bulk system, the phase transition appears at fixed temperature, while 1D material illustrates long-range behaviour, which is observed at absolute temperature. Being at the periphery between these two end points, the condition in atomically thin van der Waals crystals such as graphene, h-BN, transition metal dichalcogenides (TMDCs), magnetic semiconductors (chromium trihalides, $FePS_3$, and $MnPS_3$) [41, 42] is extremely complicated. Therefore, it can destroy the intrinsic magnetism by strong thermal perturbation, even short-range MPE surpass their macroscopic length scale which vigorously modifies the transport and optical properties. For example, gapless and massless pristine graphene exhibiting a linear dispersion near K point in the Dirac cone, provides negligible spin–orbit coupling (SOC) and spin-unpolarized density of states (DOS). Thereafter, proximity effects profoundly alter the characteristics of neighbouring material such that it can acquire spin-polarization [43], SOC [44, 45], magnetism [46], or superconductivity [47]. Sandwiching with magnetic insulators will lead to an ideal combination of vdW heterostructures, which can be advantageous for exchange interaction at the interface because of atomically active interfacial registry. In addition, proximity with magnets alters the electronic structure of atomically thin interfacial registry and can proceed for spin-selective transmission across the interface. Therefore, scaling down of nanostructures and modified version of interfaces can ardently amend the properties of 2D atomic crystals through proximity effects.

However, in this thesis we have highlighted the anatomy of proximity effect along with external perturbative effect (SOC, electric field, gating, strain, etc) and their intriguing properties for their application in spintronics, valleytronics and magnetoresistance, which provides many other tantalizing opportunities.

Thereof, proximity effect can be an exciting platform to model novel exotic quantum phenomena, revealing the universal properties of heterosystems impervious to disorganization of spin and local fluctuations which lead to the applications such as magnetic storage using magnetic skyrmions [48], or quantum computing [49]. The proximity effect basically relies on microscopic mechanism of vdW heterostructures and will be discussed in the next section.

1.3.1 Direct exchange coupling

The direct exchange phenomena result in the electrostatic screening from neighbouring atoms. It generates an overlapping electron wave function and exchange antisymmetric fermions. The electrons act as fermions, the wave functions of two-electron exhibit spatial as well as spin ordinant that should be antisymmetric as shown in figure 1.5. If we consider the bonding (antibonding) conditions exhibiting smaller eigen energy, the spatial part will be symmetric (antisymmetric) and spin will be antiparallel (parallel), which guide towards ferromagnetic (antiferromagnetic) exchange interaction [50]. The interacting behaviour of electrons promotes symmetric or antisymmetric overlap of electron wavefunction having lower energy related to spatial component and achieves the ferromagnetic or antiferromagnetic ground state of the system.

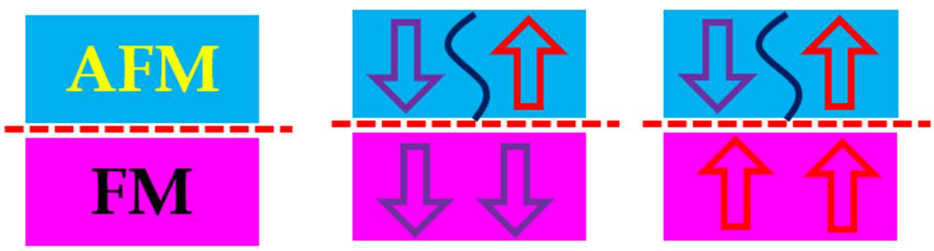

Figure 1.5: Direct exchange effect in FM/AFM bilayer explaining the mechanism of proximity effect.

1.3.2 Charge transfer

While designing vdW heterostructure *via* interfacial phenomena, charge transfer occurs at the interface to transform electronic and structural behaviour of the system [51]. As a consequence of such interfacial phenomena, the chemical potential get neutralized across the edge leading to difference in work function value. Moreover, charge disparity, dissimilar chemical potential and band renormalization influence charge transfer at interface, configures 2D electron gas and electron reconstruction. The redistribution of charges (i.e., spin and orbital) occurs at the surface and interface of atoms leading to interfacial magnetism. Therefore, charge transfer mechanism is the key mechanism to manipulate spin, orbital and electron contribution across the interface at proximitized materials.

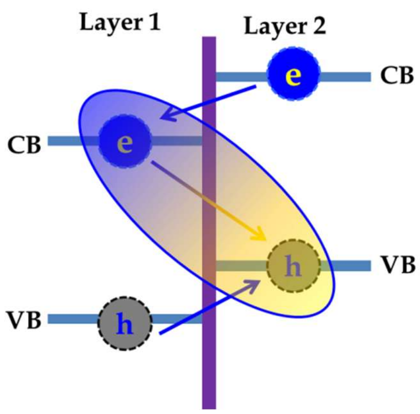

Figure 1.6: Charge transfer mechanism across the interface from one layer to adjacent layer via proximity integration.

1.4 Induced magnetism in non-magnetic layered materials

Since the discovery of graphene, efforts to generate magnetism in non-magnetic 2D materials have continued to gain enormous attention very quickly in proximitized materials. One of the conventional approaches is introducing defects or dopants or adding adatoms, coupling with metallic magnetic substrates or 3D materials [4–6]. Creating defects, dopants or coupling with magnetic substrates can extrinsically develop localized spin moments generated from unpaired electrons, which has the possibility of orbital hybridization at the interface through conduction electron in atomically thin vdW materials. However, efforts to arrange these electronic spins in an ordered fashion will be a challenging task in material preparation. In this line, realization of long-range magnetic ordering in close proximity with non-magnetic layered material is still a subject of research interest [52]. Flat band ferromagnetism has been introduced *via* induction of defects in the zigzag periphery of graphene nanoribbons or grain boundaries of atomically thin layered vdW heterostructure [53]. Such defects lead to inaccurate electronic band which signifies the large amount of DOSs in a small energy regime and generates stoner instability, tending to ferromagnetic phase. Moreover, these chemically dynamic defects are exposed to the penetration of exotic species with which long-range ferromagnetism cannot be acquired [54]. The ferromagnetic ordering in bilayer graphene was predicted by electronic band structure under the existence of electrical biasing [55] and doped gallium selenide [56].

The proximity effect is a scheme to transform the pristine system via interfacial effect through adjacent material with functionalities such as superconducting, magnetoresistance, magnetic and topologically non-trivial. In this regard, effective Hamiltonian is sensitive due to proximitized layer β and acquires different functionality from those of α, β and γ layers as shown in figure 1.7. A pristine graphene monolayer has been transferred above YIG [57] or coupled free standing above WS_2 [16] can manifest AHE and enhances the strength of SOC. Most importantly, a dephasing of spin can be observed because of spatiotemporal perturbation of interfacial exchange interaction [58], which focuses on understanding the spin related transport phenomena in proximity systems.

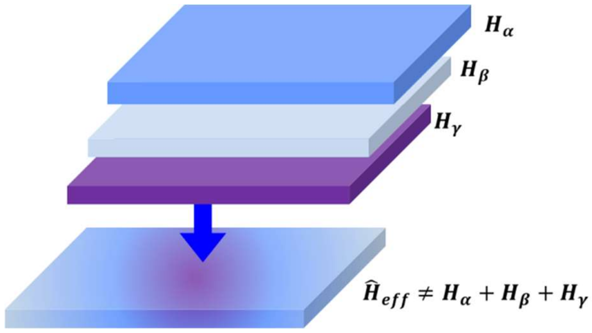

Figure 1.7: Proximitized layer β in close vicinity with layers α and γ as a function of effective and individual Hamiltonians of the layers represented as $\hat{H}_{eff}$, H_α, H_β and H_γ.

1.5 Interfacial Engineering using proximity effect

The advantage of utilizing magnetic 2D nanostructures as a constituent of vdW heterostructures can basically fulfil two important goals. One is to extend the performance of vdW heterostructures suitable for carrying out experiments and to develop novel device applications. Additional way is to engineer standard interface evolving from proximity to nearby 2D vdW system as a tool to manipulate the magnetic behaviour of the magnetic layer. 2D materials realize high spin and valley polarization, which offers exciting opportunities for the development of efficient spintronics and valleytronics. Sandwiching 2D spintronic or valleytronic material via MPE with magnetic semiconductor or insulator can provide a plausible procedure to acquire spin or valley-polarized heterostructure. Moreover, coupling with magnetic insulators leads to seamless integration and interplay of interfacial exchange interaction at the atomically sharp interface. The main aim to employ spin degrees of freedom essentially require spin-dependent phenomena at proximitized material, which is not present initially in bulk counterparts, such as spin-up and spin-down electrons (with respect to the direction of magnetization or an externally applied electric or magnetic field) that are no longer identical. Various heterostructure systems have been realized with MPE to tune the spin and valley-polarization phenomena, which includes EuS on graphene [59], WSe_2 on EuS [60], graphene on CrI_3 [61] and graphene on h-BN [62]. MPE has been realized till date with various combined heterostructure such as ferromagnetic substrate-superconductor [63], nonmagnetic bilayer heterostructure [64], and the nonmagnet-ferromagnetic substrate [42] shown schematically in figure 1.7. However, coupling with magnetic conducting substrate limits the design in developing spin-switches via clearly short-circuiting the neighbour layer material. Moreover, the concept of spin-injection and topological behaviour from one layer to another neighbouring layer via proximity effect is presently a fascinating and emerging area of research. Thereafter, 2D layered nonmagnetic material is introduced and linked with two ferromagnetic layers for realizing the behaviour of vdW spin-valve, which have a different outlook from lateral heterostructure spin-valves, where the substantial section of the nonmagnetic layered system is not in correspondence with the ferromagnetic electrode. The relative orientations of spin in vdW spin-valves can show a robust impression on in-plane conductance predominant by spin proximity effect [65]. Topological phases in a 2D layered system can attract enormous attention due to the exciting physics and application prospect in spintronics and valleytronics [35, 66]. In case of quantum spin Hall effect, topological edge states are inoculated by time-reversal symmetry, while the existence of exchange coupling can break time-reversal symmetry leading to quantum anomalous Hall effect (QAHE) [67]. QAHE phases have been realized in graphene and graphene on Ising antiferromagnet $MnPSe_3$ via proximity effect. Topologically trivial states are realized by combining uniform and staggered spin–orbit interaction exhibit proximitized graphene edge states visualizing the magnetization orientation-dependent QAHE phases [68]. The proximity exchange coupling can incorporate stronger and enhanced effects [14] for realizing Zeeman coupling by introducing an extrinsically applied large magnetic field, without significantly varying the band structure. Therefore, the short-range behaviour of proximity effects leads to large exchange coupling within the 2D layer which is in close contact with magnetic material [69]. More attempts have been made using chemical vapour deposition of Bi_2Te_3 on $Cr_2Ge_2Te_6$ [70] and MBE growth of $Cr_2Ge_2Te_6$ on $(Bi, Sb)_2Te_3$ [71]. Another cascade of focus toward QAHE was recently toggled by the experimental realization in proximity-coupled YIG/graphene/h-BN heterostructure system [72]. Apart from SOC, other property can be approachable to graphene via proximity effect which can be relevant in spintronics.

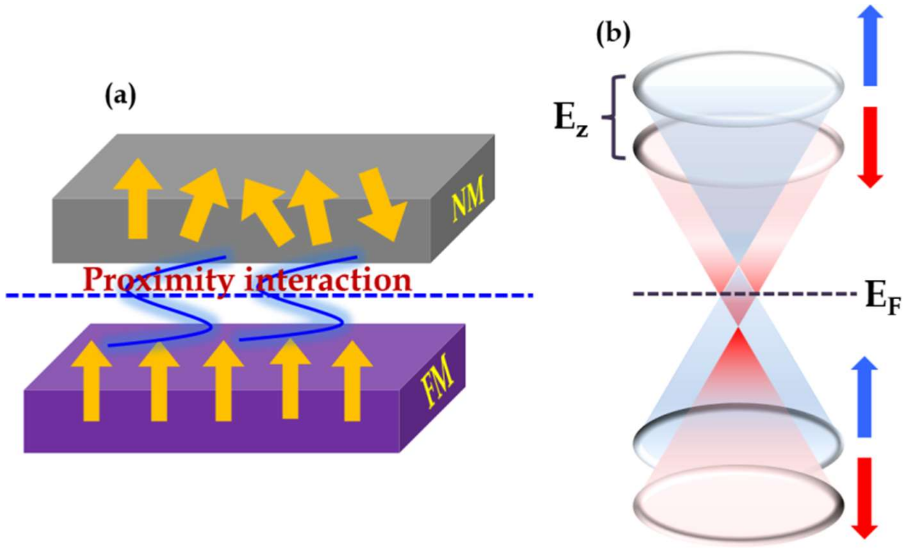

Figure 1.8: (a) Proximitized heterostructure of non-magnet (NM) and ferromagnet (FM). (b) Zeeman energy splitting of electronic bands (Dirac Cone) and Fermi level (E_F) due to seamless proximity integration. The blue and red arrows represent spin-up and down states, respectively.

1.6 Applications related to proximity effect

On altering spin and charge related phenomena of materials utilizing MPE the evolving applications can mostly scrutinize based on spintronics, valleytronics, magnonics, Spin-orbitronics. However, MPE obeys two important motivations i.e., one is to scrutinize the opportunities which can accompany or restore other strategies for designing the nanoelectronic device and other is to explore different systems, where the presence of inherent properties utilizing proximity effect could validate novel applications. Moreover, proximity effect allows us to reimagine process information as well as introduce low-power spintronics but also how to combine delicately non-labile memory and logic devices. Thereafter, penetration of spin current can be improved by proximity effects comprising a novel range of spin-based switching devices.

1.6.1 Magnetic Tunnel junction (MTJ)

The magnetic tunnel junction (MTJ) is the most important architecture for designing novel spin-electronic device [73, 74]. The most prominent advantage in all type of vdW MTJs is the breadth of the barrier, which enables the whole area of tunnelling. In contrary, non-uniform MTJs realize the tunnelling current preferentially through thinner barrier regions, which is an exponential parameter across the breadth of the barrier. However, various vdW MTJs has been realized on the basis of $Fe_{0.25}TaS_2$-Ta_2O_5-$Fe_{0.25}TaS_2$ [73], graphite-CrI_3-graphite [75], Fe_3GeTe_2-BN-Fe_3GeTe_2 [76], and graphite-$CrBr_3$-graphite [77]. A huge tunnelling magnetoresistance has been observed on graphite-CrI_3-graphite heterostructure with maximum magnetoresistance representing 19 000% at 2 K [78], 550% at 300 mK [79], 10 000% at 10 K [75] and 1000 000% at 1.4 K [80]. The importance of this kind of MTJ was considered to utilize various scattering mechanisms for tunnelling of electrons over the alternate polarization of spin in CrI_3 layers. Moreover, those MTJs were understood from magnon-assisted tunnelling procedure is realized by coupling with magnetic substrates logically by microwave. The excited magnons can propagate into the adjacent 2D materials which differ from traditional electron- or phonon-assisted tunnelling procedure without necessarily considering the conducting electron. A prominent spin-valve effect has been realized in Fe_3GeTe_2/h-BN/Fe_3GeTe_2 junctions because of the variation in coercive field from the two electrodes acquiring non-consistent demagnetization factor having a different structure. The basic proof-of-concept shows a promising focus towards proximitized materials for high-efficacy in spin-electronics, magnonics or magnetoresistance as shown in figure 1.9.

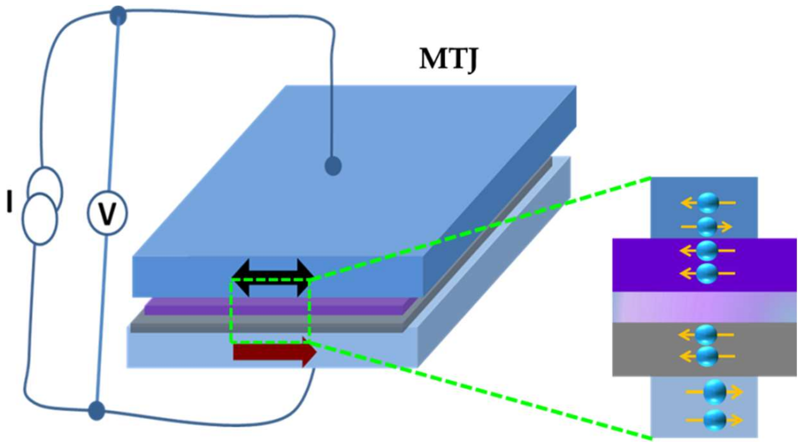

Figure 1.9: Schematic representation of a magnetic tunnel junction device.

1.6.2 Spin-lasers

Lasers are predominant in day-to-day life with their applications in optical storage, printing, optical sensor, display systems [81]. However, to attain population inversion for lasing, a huge mobility of electron is required, which guides to realize shorter spin relaxation times along with a shorter spin diffusion length [82]. With the incorporation of magnetic species in spin lasers, MPE could be introduced as electrically toggle sources of spin-polarized carrier as well as to overcome the utmost need for inducing magnetization via spin pumping on nonmagnet. The feasibility in vdW-based spin lasers with prudent spin-dependent properties has been realized by experimental evidence of lasing identical structures, which permit a very low lasing threshold [83]. Moreover, for realizing proximity effect in vertical geometric lasers could be advantageous for vdW heterojunction for elucidating modified properties analogous to lateral counterparts [84]. The figure 1.10 displays the schematic picture of spin-laser.

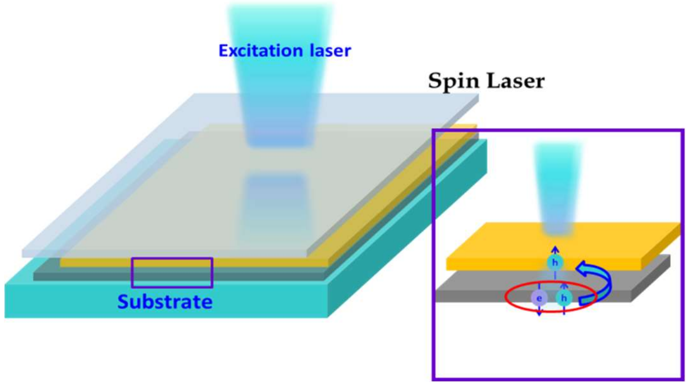

Figure 1.10: Displaying the device prototype of spin laser via proximity effect. The excitation laser is used to manipulate the spin in presence of proximity effect and making it feasible for optoelectronic applications.

1.6.3 Spin-logics

Spin logic is a device that mostly employs magnetoresistive properties in which lower or higher electrical resistance states can be modified by orienting the magnetization value of each layer in parallel or antiparallel, respectively. The feasible utilization of spin, instead of charge as an internal variable in developing devices for processing and storing information, has been discussed broadly [85, 86] because it has the ability to permit low-power spintronic device. However, beyond the development of magnetic hard drives, a fundamental challenge remains to incorporate such non-liable ferromagnetic material as a purpose of ordered amalgamation of built-in memory and spin logic [87]. This interesting anticipation provides a focus to conquer the intrinsic restrictions on widely engaged logic circuits on the basis of von Neumann architecture. The plan of such a logic circuit depends on the development of central processing units integrated via a transmission channel to memory. The conventional examples such as the internet protocol addresses using network routers are analogous with a list of patterns for finding a match. Introduction of such conventional logic devices may be restricted to scalability issues, making them complicated for large issue of scavenging that are essential for fulfilling contemporary tasks [88]. However, the spin in lateral spin-valves introduce magnetologic gates (MLGs) [89] which has been successively expanded it to ferromagnet-graphene heterojunctions [90], for smooth incorporation of memory and logic utilizing assembly. The possibility for realizing spin logic has given thrust during the demonstration at an ambient temperature of the MLG built on graphene [91]. The tunability of large SOC is not possible in pristine graphene materials, but can be realized by stacking graphene in close proximity with materials exhibiting spin–orbit phenomena.

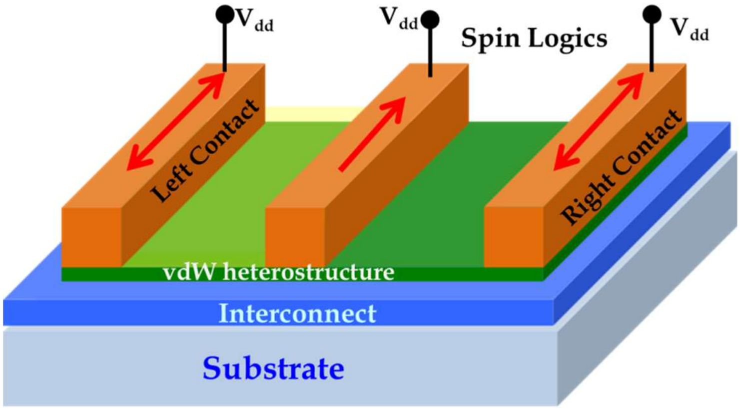

Figure 1.11: Schematic illustration of device prototype of spin logics via MPE.

1.6.4 Field effect Transistors (FET)

The transistor application related to spin instead of charge is known as spin transistors which could unveil high potentiality in non-labile device applications. However, the realization is still a fundamental challenge. The external perturbative control on 2D vdW heterostructure is of gaining enormous attention for fundamental key reasons and its potential device application like field-effect transistor [62]. In the past two decades, thin films comprising of conducting materials, gate manipulated magnetism has been established, firstly by doping or defects created in semiconductor and later followed by traditional ferromagnetic substrates [92]. Moreover, the electric nfield manipulation has been recently realized in bilayer CrI_3 [8, 93].

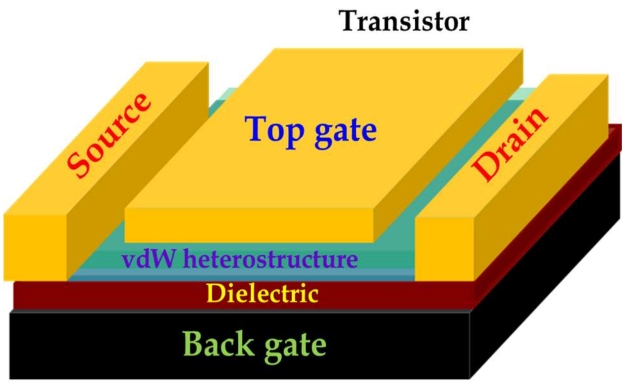

Figure 1.12: A device prototype of field effect transistor (FET) using MPE. The vdW heterostructure is sandwiched between top and back gate connected to two leads named as source and drain.

1.7 Objectives of the thesis

As mentioned in previous section, many efforts have been focused in understanding interfacial phenomena extrinsically by engineering defects, dopants, adatoms and coupling with 3D substrates. Such interfacial effect with extrinsic perturbation has limitations in maintaining distinct order parameter, resulting in short-circuits of adjacent layer. In this regard, designing vdW heterostructure by proximity effect overcomes the restricted shortcomings and tender customizable functionalities in vdW heterostructure system. To overcome these shortcomings of maintaining long-range ordering and short circuiting in device front, the following objectives are framed for the thesis.

(i) Theoretical modelling to achieve proximity induced effects in the developed 2D vdW heterostructures through Quasi-Newtonian algorithm under the DFT framework.

(ii) Detailed calculation to realize the impact of magnetic proximity effect is-

(a) Electronic structure (i.e., DOS, bands with projection in momentum space, charge density, spin density, spin-polarized effect etc.)

(b) Topological phases (i.e., SOC, Berry curvature, topological invariant, trivial and non-trivial phase, WCC) using DFT and WTBH method.

(c) Magnetic property (magnetic moment and magnetic ordering) under the framework of DFT simulation.

(d) Transmission coefficient calculation (ballistic transport under NEGF formalism, thermoelectrics (electrical conductivity), spin-polarized conductance) using DFT and BoltzTrap2 code.

(iii) Variation of electric field along with proximity effect for tuning the spin related phenomena of modelled vdW heterostructure for nanoscale devices.

(iv) Theoretically designing bilayer heterostructure comprising Weyl semimetal and ferromagnet semiconductor under DFT framework by transforming the semimetal-semiconductor characteristic to half-metallic character, which establishes magnetic proximity effect.

(v) Tunability of magnetic proximity effect with the variation of external stimulus such as electric field and quantum conductance in transmission channel of vdW heterostructure.

References

[1] Talukdar, M., Behera, S. K., Bhattacharya, K. and Deb, P. Surface modified mesoporous g-C_3N_4@$FeNi_3$ as prompt and proficient magnetic adsorbent for crude oil recovery. *Applied Surface Science*, 473:275, 2019.

[2] Talukdar, M., Behera, S. K. and Deb, P. Graphitic carbon nitride decorated with FeNi3 nanoparticles for flexible planar micro-supercapacitor with ultrahigh energy density and quantum storage capacity. *Dalton Transactions*, 48:12137, 2019.

[3] Talukdar, M., Nath, O. and Deb, P. Enhancing barrier properties of biodegradable film by reinforcing with 2D heterostructure. *Applied Surface Science*, 541:148464, 2021.

[4] Alvarez, C. J., Dau, M. T., Alain, M., Vergnaud, C., Poche, H. L., Pochet, P., Jamet, M. and Okuno, H. Impact of a van der Waals interface on intrinsic and extrinsic defects in an MoSe2 monolayer. *Nanotechnology*, 29:425706, 2018.

[5] Chen, J., Shan, Y., Wang, Q., Zhu, J. and Liu, R. P-type laser-doped $WSe_2/MoTe_2$ van der Waals heterostructure photodetector. *Nanotechnology*, 31:295201, 2020.

[6] McCreary, K. M., Swartz, A. G., Han, W., Fabian, J. and Kawakami, R. K. Magnetic moment formation in Graphene detected by scattering of Pure spin currents. *Physical Review Letters*, 109:186604, 2012.

[7] Sun, Y., Zhuo, Z., Wu, X. and Yang, J. Room-temperature ferromagnetism in two-dimensional Fe2Si nanosheet with enhanced spin-polarization ratio. *Nano Letters*, 17:2771, 2017.

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

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

[10] Liu, H., Liu, Y. and Zhu, D. Chemical doping of graphene. *Journal of Material Chemistry*, 21:3335, 2011.

[11] Fan, M. et al. Doping Nanoscale graphene domains improves magnetism in Hexagonal Boron nitride. *Advanced Materials*, 31:1805778, 2019.

[12] Li, B., Xing, T., Zhong, M., Huang, L., Lei, N., Zhang, J., Li, J. and Wei, Z. A two-dimensional Fe-doped SnS_2 magnetic semiconductor. *Nature Communications*, 8:1958, 2017.

[13] Chernyshov, A., Overby, M., Liu, X., Furdyna, J. K., Lyanda-Geller, Y. and Rokhinson, L. P. Evidence for reversible control of magnetization in a ferromagnetic material by means of spin-orbit magnetic field. *Nature Physics*, 5:656, 2009.

[14] Srivastava, A., Sidler, M., Allain, A. V., Lembke, D. S., Kis, A. and Imamoglu, A. Valley Zeeman effect in elementary optical excitations of monolayer WSe_2. *Nature Physics*, 11:141, 2015.

[15] Song, H. -D., Zhu, P. -F., Yang, X., Qin, M., Ren, Z., Duan, C. -G., Han, G., Liao, Z. -M. and Yu, D. Electrical control of magnetic proximity effect in a graphene/multiferroic heterostructure. *Applied Physics Letters*, 113:183101, 2018.

[16] Omar, S. and Van Wees, B. J. Spin transport in high-mobility graphene on WS_2 substrate with electric-field tunable proximity spin-orbit interaction. *Physical Review B*, 97:045414, 2018.

[17] Dietl, T. and Ohno, H. Dilute ferromagnetic semiconductors: Physics and spintronic structures. *Review Modern Physics*, 86:187, 2014.

[18] Koyam, Y., Arai, H., Tanaka, I., Uchimoto, Y. and Ogumi, Z. First principles study of dopant solubility and defect chemistry in $LiCoO_2$. *Journal of Material Chemistry A*, 2:11235, 2014.

[19] Korenev, V. L., Akimov, I. A., Zaitsev, S. V., Sapega, V. F., Langer, L., Yakovlev, D. R., Danilov, Y. A. and Bayer, M. Dynamic spin polarization by orientation-dependent separation in a ferromagnet–semiconductor hybrid. *Nature Communications*, 3:959, 2012.

[20] Hirohata, A., Yamada, K., Nakatani, Y., Prejbeanu, I. –L., Diény, B., Pirro, P. and Hillebrands, B. Review on spintronics: Principles and device applications. *Journal of Magnetism and Magnetic Materials*, 509:166711, 2020.

[21] Buzdin, A. I. Proximity effects in superconductor-ferromagnet heterostructures. *Review Modern Physics*, 77:935, 2005.

[22] Ovsyannikov, G. A., Constantinian, K. Y., Demidov, V. V. and Khaydukov, Y. N. Magnetic proximity effect and superconducting triplet correlations at the cuprate superconductor and oxide spin valve interface. *Low Temperature Physics*, 42:873, 2016.

[23] Scharf, B., Xu, G., Matos-Abiague, A. and Žuti´c, I. Magnetic proximity effects in transition-metal dichalcogenides: converting Excitons. *Physical Review Letter* 119:127403, 2017

[24] Lee, C., Katmis, F., Jarillo-Herrero, P., Moodera, J. S. and Gedik, N. Direct measurement of proximity-induced magnetism at the interface between a topological insulator and a ferromagnet. *Nature Communications*, 7:12014, 2016.

[25] Vasili, H. B., Gamino, M., G`azquez, J., S´anchez, F., Valvidares, M., Gargiani, P., Pellegrin, E. and Fontcuberta, J. 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.

[26] Phuc, H. V., Hieu, N. N., Hoi, B. D. and Nguyen, C. V. Interlayer coupling and electric field tunable electronic properties and Schottky barrier in a graphene/bilayer-GaSe van der Waals heterostructure. *Physical Chemistry Chemical Physics*, 20:17899, 2018.

[27] Holm, R. The Electric Tunnel Effect across Thin Insulator Films in Contacts. *Journal of Applied Physics*, 22:569, 1951.

[28] Zuckermann, M. J. The proximity effect for weak itinerant ferromagnets. *Solid State Communication*, 12:745–7, 1973.

[29] Bergman, G. Transition from Pauli Paramagnetism to band ferromagnetism in very thin Ni films. *Physical Review Letters*, 41:264, 1978.

[30] Ceperley, D. and Alder, B. Quantum Monte carlo. *Science*, 231:555, 1986.

[31] Kohn, W. and Sham, L. J. Self-consistent equations including exchange and correlation effects. *Physical Review*, 14:A1133, 1965.

[32] Hohenberg, P. and Kohn, W. Inhomogeneous electron gas. *Physical Review*, 136:B864, 1964.

[33] Garrity, K. F. and Choudhary, K. Database of Wannier tight-binding Hamiltonians using high-throughput density functional theory. *Scientific Data*, 8:106, 2021.

[34] Grimme, S., Antony, J., Ehrlich, S., and Krieg, H. A consistent and accurate ab initio parametrization of density functional dispersion correction (dft-d) for the 94 elements h-pu. *The Journal of Chemical Physics*, 132(15):154104, 2010.

[35] Han, W., Kawakami, R. K., Gmitra, M. and Fabian, J. Graphene spintronics. *Nature Nanotechnology*, 9(10):794, 2014.

[36] Alicea, J. New directions in the pursuit of Majorana fermions in solid state systems. *Reports on Progress in Physics*, 75:7, 2012.

[37] Fumega, A. O., Blanco-Canosa, S., Babu-Vasili, H., Gargiani, P., Li, H., Zhou, J. -S., Rivadulla, F. and Pardo, V. Electronic structure and magnetic exchange interactions of Cr-based van der Waals ferromagnets. A comparative study between $CrBr_3$ and $Cr_2Ge_2Te_6$. *Journal of Material Chemistry C*, 8:13582, 2020.

[38] Baral, D., Fu, Z., Zadorozhnyi, A. S., Dulal, R., Wang, W., Shrestha, N., Erugu, U., Tang, J., Dahnovsky, Y., Tian, J. and Chien, T. Small energy gap revealed in CrBr3 by scanning tunnelling spectroscopy. *Physical Chemistry Chemical Physics*, 23:3225, 2021.

[39] Manzeli, S., Ovchinnikov, D., Pasquier, D., Yazyev, O. V. and Kis, A. 2D transition metal dichalcogenides. *Nature Review Materials*, 2(8):17033, 2017.

[40] Tang, S. et al. Quantum spin Hall state in monolayer 1T′-WTe_2. *Nature Physics*, 13:683, 2017.

[41] Hu, G. et al. Antisymmetric magnetoresistance in a van der Waals antiferromagnetic/ferromagnetic layered $MnPS_3$/Fe_3GeTe_2 stacking heterostructure. *ACS Nano*, 14(9):12037, 2020.

[42] Lyons, T. P., Gillard, D., Molina-Sanchez, A., Misra, A., Withers, F., Keatley, P. S., Kozikov, A., Taniguchi, T., Watanabe, K., Novoselov, K. S., Fernandez-Rossier, J. and Tartakovskii, A. I. Interplay between spin proximity effect and charge-dependent exciton dynamics in $MoSe_2/CrBr_3$ van der Waals heterostructures. *Nature Communications*, 11:6021, 2020.

[43] Lazic, P., Sipahi, G. M., Kawakami, R. K. and Žuti´c, I. Graphene spintronics: Spin injection and proximity effects from first principles. *Physical Review B*, 90:085429, 2014.

[44] Seoane, J. et al. Cerebrospinal fluid-derived circulating tumour DNA better represents the genomic alterations of brain tumours than plasma. *Nature Communications*, 6:8839, 2015.

[45] Savero Torres, W., Sierra, J. F., Benítez, L. A., Bonell, F., Costache, M. V. and Valenzuela, S. O. Spin precession and spin Hall effect in monolayer graphene/Pt nanostructures. *2D Materials*, 4:4, 2017.

[46] Zhang, W., Wong, P. K. J., Zhu, R. and Wee, A. T. S. Van der Waals magnets: Wonder building blocks for two-dimensional spintronics? *InfoMat*, 1:479–95, 2019.

[47] Cao, Y., Fatemi, V., Fang, S., Watanabe, K., Taniguchi, T., Kaxiras, E. and Jarillo-Herrero, P. Unconventional superconductivity in magic-angle graphene superlattices. *Nature*, 556:43, 2018.

[48] Hsu, P-J., Kubetzka, A., Finco, A., Romming, N., Bergmann, K. V. and Wiesendanger, R. Electric-field-driven switching of individual magnetic skyrmions. *Nature Nanotechnology,* 12:123–6, 2017.

[49] Bhattacharyya, B., Awana, V. P. S., Senguttuvan, T. D., Ojha, V. N. and Husale, S. Proximity-induced supercurrent through topological insulator-based nanowires for quantum computation studies. *Scientific Reports*, 8:17237, 2018.

[50] Simon, S. H. The oxford solid state basics, Oxford University press, Oxford, UK 2013.

[51] Chakhalian, J., Freeland, J. W., Millis, A. J., Panagopoulos, C. and Rondinelli, J. M. Colloquium: Emergent properties in plane view: Strong correlations at oxide interfaces. *Review of Modern Physics, 86*:1189, 2014.

[52] Sepioni, M., Nair, R. R., Tsai, I. L., Geim, A. K. and Grigorieva., I. V. Revealing common artifacts due to ferromagnetic inclusions in highly oriented pyrolytic graphite. *Europhysics Letters*, 97:47001, 2012.

[53] Magda, G. Z., Jin, X., Hagyamasi, I., Vancso, P., Osvath, Z., Nemes-Incze, P., Hwang, C., Biro, L. P. and Tapaszto, L. Room-temperature magnetic order on zigzag edges of narrow graphene nanoribbons. *Nature*, 514:608, 2014.

[54] Makarov, T. L. et al. Edge state magnetism in zigzag-interfaced graphene via spin susceptibility measurements. *Scientific Reports*, 5:13382, 2015.

[55] Sharpe, A. L., Fox, E. J., Barnard, A. W., Finney, J., Watanabe, K., Taniguchi, T., Kastner, M. A. and Goldhaber-Gordon, D. Emergent ferromagnetism near three-quarters filling in twisted bilayer graphene. *Science*, 365:605–8, 2019.

[56] Cao, T., Li, Z. and Louie, S. G. Tunable Magnetism and Half-Metallicity in Hole-Doped Monolayer GaSe. *Physical Review Letters*, 114:236602, 2015.

[57] Wang, Z., Tang, C., Sachs, R., Barlas, Y. and Shi, J. Proximity-Induced Ferromagnetism in Graphene Revealed by the Anomalous Hall Effect. *Physical Review Letters*, 114:016603, 2015.

[58] Singh, S., Katoch, J., Zhu, T., Meng, K-Y., Liu, T., Brangham, J. T., Yang, F., Flatté, M. E. and Kawakami, R. K. Strong Modulation of Spin Currents in Bilayer Graphene by Static and Fluctuating Proximity Exchange Fields. *Physical Review Letters*, 118:187201, 2017.

[59] 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.

[60] Norden, T., Zhao, C., Zhang, P., Sabirianov, R., Petrou, A. and Zeng, H. Giant valley splitting in monolayer WS_2 by magnetic proximity effect. *Nature Communication*, 10:4163, 2019.

[61] Farooq, M. U. and Hong, J. Switchable valley splitting by external electric field effect in graphene/CrI_3 heterostructures. *npj 2D Materials and Application*, 3:3, 2019.

[62] Behera, S. K. and Deb, P. Controlling the bandgap in graphene/h-BN heterostructures to realize electron mobility for high performing FETs. *RSC Advances*, 7:31393, 2017.

[63] Savostin, E. O. and Pertsev, N. A. Superconducting straintronics via the proximity effect in superconductor–ferromagnet nanostructures. *Nanoscale*, 12:648–57, 2020.

[64] Island, J. O. et al. Spin–orbit-driven band inversion in bilayer graphene by the van der Waals proximity effect. *Nature*, 571:85, 2019.

[65] Cardoso, C., Soriano, D., Garcia-Martinez, N. A. and Fernandez-Rossier, J. Van der Waals spin valves. *Physical Review Letters*, 121:067701, 2018.

[66] Ren, Y., Qiao, Z. and Niu, Q. Topological phases in two-dimensional materials: a review. *Reports on Progress in Physics*, 79:066501, 2016.

[67] Qiao, Z., Yang, S. A., Feng, W., Tse, W. K., Ding, J., Yao, Y., Wang, J. and Niu, Q. Quantum anomalous Hall effect in graphene from Rashba and exchange effects. *Physical Review B*, 82:161414(R), 2010.

[68] Hogl, P., Frank, T., Zollner, K., Kochan, D., Gmitra, M. and Fabian, J. Quantum Anomalous Hall Effects in Graphene from Proximity-Induced

Uniform and Staggered Spin-Orbit and Exchange Coupling. *Physical Review Letters*, 124:136403, 2020.

[69] Zollner, K., Junior, P. E. F. and Fabian, J. Proximity exchange effects in MoSe2 and WSe2 heterostructures with CrI3: Twist angle, layer, and gate dependence. *Physical Review B*, 100:085128, 2019.

[70] Algeria, L. D., Ji, H., Yao, N., Clarke, J. J., Cava, R. J. and Petta, J. R. Large anomalous Hall effect in ferromagnetic insulator-topological insulator heterostructures. *Applied Physics Letter*, 105:053512, 2014.

[71] Mogi, M., Tsukazaki, A., Kaneko, Y., Yoshimi, R., Takahashi, K. S., Kawasaki, M. and Tokura, Y. Ferromagnetic insulator Cr2Ge2Te6 thin films with perpendicular remanence. *APL Materials*, 6:0911904, 2018.

[72] Tang, C., Cheng, B., Aldosary, M., Wang, Z., Jiang, Z., Watanabe, K., Taniguchi, T., Bockrath, M. and Shi, J. Approaching quantum anomalous Hall effect in proximity-coupled YIG/graphene/h-BN sandwich structure. *APL Materials*, 6:026401, 2018.

[73] Arai, M., Moriya, R., Yabuki, N., Masubuchi, S., Ueno, K. and Machida, T. Construction of van der Waals magnetic tunnel junction using ferromagnetic layered dichalcogenide. *Applied Physics Letters*, 107:103107, 2015.

[74] Tong, Q., Chen, M. and Yao, W. Magnetic Proximity Effect in a van der Waals Moiré Superlattice. *Physical Review Applied*, 12:024301, 2019.

[75] Wang, Z., Gutiérrez-Lezama, I., Ubrig, N., Kroner, M., Gibertini, M., Taniguchi, T., Watanabe, K., Imamo˘glu, A., Giannini, E. and Morpurgo, A. F. Very large tunneling magnetoresistance in layered magnetic semiconductor CrI_3. *Nature Communications*, 9:2516, 2018.

[76] Wang, Z., Sapkota, D., Taniguchi, T., Watanabe, K., Mandrus, D. and Morpurgo, A. F. Tunneling Spin Valves Based on Fe3GeTe2/hBN/Fe3GeTe2 van der Waals Heterostructures. *Nano Letters*, 18:4303–8, 2018.

[77] Ghazaryan, D. et al. Magnon-assisted tunnelling in van der Waals heterostructures based on $CrBr_3$. *Nature Electronics*, 1:344–9, 2018.

[78] Song, T. et al. Giant tunneling magnetoresistance in spin-filter van der Waals heterostructures. *Science*, 360:1214–8, 2018.

[79] Klein, D. R. et al. Probing magnetism in 2D van der Waals crystalline insulators via electron tunnelling. *Science*, 360:1218–22, 2018.

[80] Kim, H. H., Yang, B., Patel, T., Sfigakis, F., Li, C., Tian, S., Lei, H. and Tsen, A. W. One Million Percent Tunnel Magnetoresistance in a Magnetic van der Waals Heterostructure. *Nano Letters*, 18:4885–90, 2018.

[81] Ferreira, M. F. S. et al. Roadmap on optical sensors. *Journal of Optics*, 19:083001, 2017.

[82] Chun, A. L. An extended membrane. *Nature Nanotechnology*, 9:750, 2014.

[83] Lee, J., Bearden, S., Wasner, E. and Žuti´c, I. Spin-lasers: From threshold reduction to large-signal analysis. *Applied Physics Letters*, 105:042411, 2014.

[84] Miao, J., Xu, Z., Li, Q., Bowman, A., Zhang, S., Hu, W., Zhou, Z. and Wang, C. Photothermal Effect Induced Negative Photoconductivity and High Responsivity in Flexible Black Phosphorus Transistors. *ACS Nano*, 11:10472, 2017.

[85] Karpiak, B. et al. Magnetic proximity in a van der Waals heterostructure of magnetic insulator and graphene. *2D Materials*, 7:015026, 2020.

[86] Hoque, A. M., Khokhriakov, D., Karpiak, B. and Dash, S. P. Charge-spin conversion in layered semimetal TaTe2 and spin injection in van der Waals heterostructures. *Physical Review Research*, 2:033204, 2020.

[87] Zutic, I., Fabian, J. and Sarma, S. D. Spintronics: Fundamentals and applications. *Review of Modern Physics*, 76:323, 2004.

[88] Dery, H., Wu, H., Ciftcioglu, B., Huang, M., Song, Y., Kawakami, R., Shi, J., Krivorotov, I., Zutic, I. and Sham, L. J. Nanospintronics based on magnetologic gates. *IEEE Transactions Electron Devices*, 59:259, 2012.

[89] Wen, H., Dery, H., Amamou, W., Zhu, T., Lin, Z., Shi, J., Žuti´c, I., Krivorotov, I., Sham, L. and Kawakami, R. K. Experimental demonstration of XOR operation in graphene magnetologic gates at room temperature. *Physical Review Applied*, 5:044003, 2016.

[90] Behin-Aein, B., Datta, D., Salahuddin, S. and Datta, S. Proposal for an all-spin logic device with built-in memory. *Nature Nanotechnology*, 5:266, 2010.

[91] Yu. L. et al. Graphene/MoS2 hybrid technology for large-scale two-dimensional electronics. Nano Letters, 14:3055–63, 2014.

[92] Li, W., Shi, J., Zhang, K. H. L. and MacManus-Driscoll, J. L. Defects in complex oxide thin films for electronics and energy applications: challenges and opportunities. *Materials Horizons*, 7:2832–59, 2020.

[93] Jiang, S., Shan, J. and Mak, K. F. Electric-field switching of two-dimensional van der Waals magnets. *Nature Materials*, 17:406–10, 2018.