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Magnetic two-dimensional systems

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ABSTRACT

Two-dimensional (2D) systems have considerably strengthened their position as one of the premier candidates to become the key material for the proposed spintronics technology, in which computational logic, communications and information storage are all processed by the electron spin. In this article, some of the most representative 2D materials including ferromagnetic metals (FMs) and diluted magnetic semiconductor (DMSs) in their thin film form, magnetic topological insulators (TIs), magnetic graphene and magnetic transition metal dichalcogenides (TMDs) are reviewed for their recent research progresses. FM thin films have spontaneous magnetization and usually high Curie temperature (T_c), though this can be strongly altered when bonded with semiconductors (SCs). DMS and magnetic TIs have the advantage of easy integration with the existing SC-based technologies, but less robust magnetism. Magnetic ordering in graphene and TMDs are even more fragile and limited to cryogenic temperatures so far, but they are particularly interesting topics due to the desired long spin lifetime as well as the outstanding mechanical and optical properties of these materials.

1. Introduction

The possibility of providing ultimate logic bit by electron's spin rather than or in addition to its charge has claimed exciting new horizons in physics, material science and electronic engineering. For spin-electronics or *spintronics* purposes, the materials carrying out the mission must be spin polarized, such as magnetic metals with the broken symmetry between spin down and spin up states near the Fermi level (E_F). Historically the spin arrangement has long been investigated within the context of conventional FM and their alloys, while the study of spin generation, relaxation, and spin-orbit coupling in non-magnetic materials has taken off rather recently with the advent of spintronics, and it is here that many novel 2D materials and systems can find their greatest potentials in both science and technology.

The earliest studies of 2D magnetic phenomena can be tracked back to discussions of the interfacial magnetism of FM/SC heterojunctions [1–18]. In this regard, a large portion of the research work over the last two decades was stimulated by the idea of creating a spin field effect transistor (SFET) [19], in which the transport of the electron spins is confined in a high mobility 2D electron gas (2DEG) channel and can be manipulated by the application of a gate voltage. An ideal spin-injected SC would demonstrate high spin polarization, operate at room temperature (RT) and be both robust and easily fabricated for potential high throughput needs. Various FM/SC heterostructures have been hotly investigated since it was demonstrated that substantial spin accumulation and diffusion occurs at the FM/SC interface [20]. The generation of high-spin-polarization current is essential for the SFET concept and therefore the half metallic materials (such as Heusler alloys, magnetite, and chromium dioxide) have been integrated into the hybrid systems [21–23]. Although an full electrical semiconductor spin field effect transistor has recently been demonstrated, all be it at 300 mK and requiring quantum ballistic point contacts [24]. Hybrid systems with ferromagnetic source as sink of spin and an easily gate tuned channel through a high mobility 2D material may offer a route to the SFET at RT.

Van der Waals materials such as graphene, layered TMDs, copper oxides, and iron pnictides with properties dominated by their 2D structural units have become the new focus of 2D magnetism research for the recent few years. The success of single-layer graphene has shown that it is not only possible to exfoliate stable, single-atom or single-polyhedral-thick 2D materials from bulk van der Waals solids, but also that these materials can exhibit fascinating and technologically useful properties. In graphene's

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band structure, the linear dispersion at the *k* point gives rise to novel physical phenomena, such as the anomalous quantum Hall effect (AQHE), and has opened up a new category of "Dirac" physics. Stimulated by the rise of graphene, other 2D crystals including layered hexagonal-boron nitride (h-BN) and TMDs were subsequently demonstrated. The more recently discovered TIs, featured with an insulating bulk gap and gapless Dirac-like band dispersion surface states, are new members of the 2D family. Unlike that of graphene, the electron states of TIs surface are strongly spin–orbit coupled and immune to time-reversal-invariant disorders. Magnetism is not common for the elements consisting Van der Waals materials; however, spin polarization in these materials can still be induced by defects, edge states, magnetic dopants and/or via the proximity to an adjunct magnetic source *etc*. Compared to FM, they generally have less robust magnetism, but significantly longer spin lifetime or spin coherence length, which are eagerly desired in spintronic engineering. Furthermore, their capacity of integration with FM offers a promising direction toward the development of hybrid devices that can perform logic, communications, and storage within the same material technology.

In this article, a selection of magnetic 2D systems will be introduced, following their historical path of emergence. This includes FM and DMS in their thin film form, magnetic TIs, magnetic graphene and magnetic TMDs, as illustrated in Fig. 1. The very recent advances of the study of these materials will be reviewed. This article does in no way try to give a complete overview of the emerging 2D systems, as many of them are still under fast developing till this day; neither is it intended to give deep theoretical descriptions of why the many fascinating phenomena occur in them. The purpose of this article is, instead, to present some of the most important findings of these material systems and to highlight a few hotly debated topics of the contemporary magnetic 2D systems research.

2. Ferromagnetic metal thin films

As the simplest form of magnetic 2D system, transition metal thin films and their alloys have been most thoroughly exploited in the history, as they are relatively easy to grow epitaxially on III–V SCs such as GaAs, InAs and InGaAs *etc*. with relatively small lattice mismatch [1–18]. The magnetic properties of these nanoscale FM thin films are closely linked with two general questions, namely (*i*) how the magnetic ordering changes with reduced dimensionality and (*ii*) how the magnetic ordering changes due to electronic bonding at the interface with the SC substrate.

The FM ultrathin films have been found to form metastable phases under certain conditions. While the bulk Co is hcp structure, the bcc Co on the GaAs(1 0 0) was demonstrated by Prinz [2,4,25] in 1985 and since then many experiments on Co/GaAs were reported with inconsistency, making this issue rather complex [1–6]. Using high-resolution transmission electron microscopy (TEM), Gu et al. [1] demonstrated hcp-structured epi-Co on GaAs and Mangan et al. [2] observed the coexistence of bcc and hcp phases. Idzerda et al. [3] confirmed the bcc structure of Co on GaAs(1 1 0) using extended X-ray absorption fine structure. Theoretical studies suggest that bcc Co is not a metastable phase but a forced structure originating from imperfections [4,5]. Calculations show bulk bcc Co can have a magnetic moment as large as 1.7 μ_B/atom [6]. The tendency to form into bcc stacking, which doesn't exist in nature, has also been found in epitaxial Ni thin films on GaAs. This was firstly demonstrated by Tang et al. [7] as early as 1986 by growing Ni on GaAs(0 0 1) at RT. The presence of bcc phase was observed up to 2.5 nm. This study was late on reproduced by Jiang et al. [8] who alternatively did the growth at 170 K and obtained a thicker bcc Ni, i.e. 3.5 nm. The bcc Ni/GaAs (1 0 0) as having a Curie temperature (T_c) of 456 K and a magnetic moment of 0.52 μ_B/atom, reveals a remarkably different electronic structure to that of fcc Ni and crucially a positive cubic anisotropy of +4.0 × 10^5 ergs/cm^3, as opposed to -5.7×10^4 ergs/cm^3 for the naturally occurring fcc Ni.

The interdiffusion between the FM and the SC atoms also play an important role in determining the magnetic properties of the ultrathin FM films. Theoretical calculations suggest that a bulk bcc Co can carry a magnetic moment as large as 1.7 μ_B/atom [6,9], while experimental reports are always below this value. By careful analyses of the RHEED patterns, Monchesky and Unguris [10] demonstrated a ferromagnetically dead layer associated with the formation of interfacial Co$_2$GaAs for Co thickness less than 3.4 monolayers (MLs) and an abrupt in-plane spin-reorientation transition reorients the magnetization along the [001] direction at 7 ML. It should be noted that all these boundaries discussed above are not absolute values but strongly depend on the specific sample deposition conditions, such as the surface atomic configuration of the substrate and the growth temperature *etc*. For example, passivating layers such as S and Sb have been used to reduce the chemical interaction at the Co/GaAs interface and the latter gives a factor of 2.3 enhancement of the magnetic moment compared to the film deposited on bare GaAs(1 1 0) substrate [3]. Characterized by X-ray photoelectron spectroscopy (XPS), the presence of the As peak in the 6-nm Ni film reveals the occurrence of As diffusion into the Ni layer destroying the magnetic properties of the fcc Ni film and leading to a 20% reduction of the magnetization compared to the bulk value [11]. In the study of the evolution of interface properties of the electrodeposited Ni film upon annealing, a significant increase of As out-diffusion has been observed for annealing temperatures up to 623 K accompanying a rise in the Schottky barrier height, which has been attributed to the Ni-Ga-As compound formation [12].

The third reason is the reduced thickness. It is well known that FM materials follow the so-called island growth geometry at low coverage. In other words, FM atoms at low thicknesses, typically less than a few MLs, can be too diffused to intensively interact with one another. For example, many researchers have reported on high quality epitaxial growth of Fe on GaAs, among which there exist the long lasting debate over the presence of magnetic dead layer at the Fe/GaAs interface [13]. This detrimental effect used to be attributed to the formation of antiferromagnetic Fe$_2$As [14] and half-magnetized Fe$_3$Ga$_{2x}$As$_x$ in the vicinity of the interface, until Xu et al. [15] demonstrated the evolution of the magnetic phase of Fe/GaAs corresponding to the growth morphology. This result was further confirmed with unambiguous X-ray magnetic circularly dichroism (XMCD) measurement of the Fe/GaAs(1 0 0) interface [16].

Direct experimental demonstration of the magnetic state of epitaxial FM/SC interface down to the atomic scale remains a

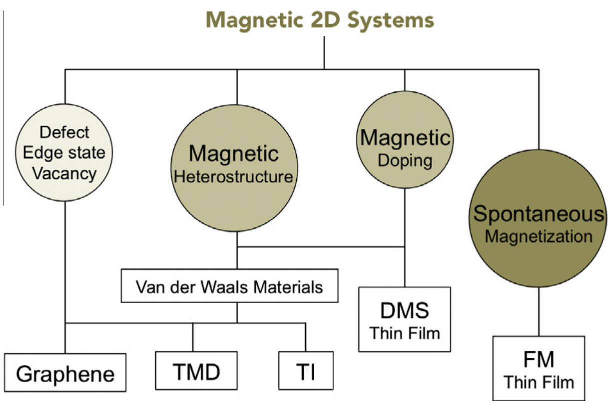

Fig. 1. Schematic illustration of the magnetic 2D systems included in this review.