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E-mail: stephan.roche@icn2.cat**Keywords:** spintronics, dimensional, materials, van der Waals, heterostructures

Abstract

We briefly summarize more than fifteen years of intense research in 2D materials (2DM)-based spintronics, which has led to an in-depth understanding of fundamental spin transport mechanisms, novel functionalities in magnetic tunnel junctions and spin orbit torque devices, and the formidable and unprecedented capability of proximity effects to make graphene a spin active material. Although the portfolio of functional 2DM-based devices and related heterostructures is continuously increasing, we outline key technological challenges that are still impeding practical spintronic applications in spin-logics and non-volatile memory technologies. We conclude by mentioning current and future directions which will maintain the momentum of the field of ultracompact spintronics based on 2DM and van der Waals heterostructures.

1. Progress over more than a decade

In 2007, Tombros *et al* [1] reported the first spin valve behavior and nonlocal spin transport measurement in graphene supported onto silicon oxide, with an estimated spin diffusion length of about $2\ \mu\text{m}$ at room temperature. From such finding, it became clear that graphene was the ideal material to convey spin information over ultralong distances at technologically relevant temperatures [2, 3]. A roadmap was established in 2015 [4] pointing towards the needs to understand the origin and nature of spin transport and relaxation, the use of proximity effect to make graphene magnetic or to manipulate the spin degree of freedom by using strong spin-orbit coupling materials, as well as to design functional devices such as spin field effect transistors and switches, spin sensors, or spin-active components in memory technologies.

After years of research and progress, the foundations of spin transport in 2DM and van der Waals heterostructures have been established, including the role of internal degrees of freedom such as pseudospin in spin relaxation [5–8], or the universal

spin diffusion length in polycrystalline graphene of varying grain morphologies [9, 10]. Importantly, as anticipated by Yang and co-workers [11, 12], the role of proximity effects in modifying the spin-dependent properties of graphene has become cornerstone. As a matter of fact, spin-transport measurements in bilayer graphene have evidenced a strong spin-charge coupling due to a large induced exchange interaction by the proximity of an interlayer antiferromagnet (CrSBr) [13], with exchange splitting in order of 20 meV (corresponding to exchanging field in order of 170 Tesla), while the magnetized graphene persists up to the Néel temperature of CrSBr ($T_N \sim 132\ \text{K}$). Progress has been made on growing high temperature 2D magnets with scalable method such as molecular beam epitaxy technique, but still efforts are required to achieve high-quality and large scale materials [14–16]. Heterostructures of graphene and chromium trihalide magnetic insulators (CrI_3 , CrBr_3 , and CrCl_3) have been found to manifest unprecedented gate tunability. The graphene becomes highly hole-doped due to charge transfer from the nearby magnetic insulator which can be modulated upon switching the magnetic states of the nearest CrI_3 layers [17].

On the other hand, interfacing graphene with strong spin–orbit coupling materials such as transition metal dichalcogenides has been shown not only to manifest in giant spin transport anisotropy, related to peculiar spin textures induced by proximity effects [18–20], but also provides the necessary (room temperature) charge-to-spin conversion efficiency for generating pure spin currents in the spin Hall effect regime [21–25] or demonstration of spin field effect transistor [26, 27] and multifunctional spin logic gates [28]. The large quantity of 2DMs allows for an almost infinite number of possible stacking and combinations, also harnessing the formation of moiré superlattices and the knob of twist angle as a resource to fine-tune spin–orbit coupling parameters for a desired spin device functionality [29–31].

The search for suited 2DMs combination to enhance spin transfer capability, as for instance manifested through the spin torque effect, has also experienced substantial progress. On the theoretical side, advances have revealed novel features unique to 2DMs [32–35], whereas experiments have started to produce significant torque efficiencies, which will serve as basis for further development [36–39]. Layered materials such as WTe_2 [40–43] or TaIrTe_4 [44, 45] with reduced symmetries are also generating unconventional spin–orbit torques.

2. Key challenges

A perspective of the potential advantages brought by graphene, 2DM and related van der Waals heterostructures to improve non-volatile magnetic random-access memories (MRAMs), such as spin-transfer torque MRAM and next-generation spin–orbit torque MRAM was published in 2022 [46]. MRAM is emerging for enabling low-power technologies, which are expected to spread over large markets from embedded memories to the Internet of Things.

Aspects such as the fundamental properties of atomically smooth interfaces, the reduced material intermixing, the crystal symmetries and the proximity effects were described as pivotal drivers for disruptive MRAM at advanced technology nodes. However, improving the technology readiness level is facing many technical challenges, some concerning all applications using 2DMs. First, despite IMEC’s successful co-integration of large-scale WS_2 (two monolayer thick) with magnetic materials (in a MRAM stack) in CMOS-compatible fab environments [46], optimizing engineering processes remains time-consuming and resources-intensive tasks. This is not easily affordable, even within the frame of a large scale project such as the Graphene Flagship.

Second, the diversity of potential 2DMs to be implemented is so large that a preliminary benchmarking is necessary to streamline material developments efforts by focusing on the most promising candidates. This is crucial, for instance, in significantly reducing the drive current required to switch a magnetic material within the MRAM stack. To achieve this, there is a demand for intensive, realistic simulations to identify the most suitable material assembly, and experimental verification on a broad set of parameters (compounds, thicknesses, thermal budgets) is necessary.

On the technology side, the large-scale synthesis and transfer of 2DMs remain two hurdles for further progress. As many advances have been realized in the mastering of catalytic vapor deposition growth techniques, the transfer from metallic to insulating or magnetic substrates comprise serious technical difficulties. The preservation of the integrity of as-grown materials during the transfer process is crucial. Measurements performed on WS_2 -based stack, were found to maintain the MRAM operation. However, the device did not display a significant gain in switching while the torque effect could not be detected [46].

3. Next steps and future directions

Beyond MRAM, other incipient applications are being explored [30]. Spin logic and multiplexer devices using graphene have been explored, using drift currents, the gate dependence of spin lifetimes or spin accumulation. Spin communication and interconnects over long distances is also possible, circumventing the capacitive coupling of the charge-based counterparts, a challenge being also tackled with the transfer of magnons in 2D magnets [47–49]. Combining functionalities of 2D materials, can open the way of novel magneto-optic devices, for instance by leveraging excitonic transitions in 2D (magnetic) semiconductors [50–53], as demonstrated in graphene-transition dichalcogenides heterostructures [54, 55].

Among the vast class of novel magnetic materials that have emerged during recent years [56], one notes the discovery of altermagnets [57, 58], which are materials (such as RuO_2 and MnTe) with collinear antiferromagnetic order possessing no net magnetization, but exhibiting spin split bands with alternating spin polarization both in real and reciprocal spaces. These materials (especially their two-dimensional forms [58]) enlarge the portfolio of enabling structures that could be combined with other 2DMs and help develop disruptive technologies of ultralow power and fast magnetization switching. Similarly, multiferroic van der Waals heterostructures

(such as $\text{FeCl}_2/\text{Sc}_2\text{CO}_2$) present interesting perspective for novel types of nonvolatile electrically switchable spintronic devices [59–61].

On the theoretical side, to accelerate the search for upper performance of given metrics (such as the spin–orbit torque efficiency), more modelling and simulation efforts are required. Such efforts should be supported by artificial intelligence techniques to elaborate automatically structural and energetic models of the many possible heterostructures under consideration. Furthermore, the recourse to methodologies able to cope with interface disorder and structural imperfections are mostly desired to describe the reality of real materials. The building of models shall retain the *ab-initio* accuracy while reducing computational costs through machine learning techniques, whereas linear scaling transport methodologies provide scaling capability of simulation of multi-million atomic (and disordered) structures [62].

Other directions which are emerging include topological spintronics, which finds its origin in the use of novel resources such as the orbital Hall effect and related orbitronics effects [34, 63, 64], nontrivial spin textures [65, 66] and the use of 2D magnetic materials to design ultra-compact spin active building blocks [30, 67, 68], which could further improve performances at single device level.

The feedback between experimental observations and advanced modelling is fundamental to progress the field and eventually demonstrate superior performances of 2DMs and van der Waals heterostructures in practical spintronic devices. This will be key to further stimulate new efforts in technology platforms or in large scale industries, already engaged into non-volatile memory technology business.

Data availability statement

No new data were created or analyzed in this study.

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