Spin injection by spin–charge coupling in proximity induced magnetic graphene

Alexey A Kaverzin, Talieh S Ghiasi, Avalon H Dismukes, Xavier Roy and Bart J van Wees

1 Zernike Institute for Advanced Materials, University of Groningen, Groningen 9747 AG, The Netherlands
2 Department of Chemistry, Columbia University, New York, NY, United States of America
* Author to whom any correspondence should be addressed.

E-mail: a.kaverzin.rug@gmail.com

Keywords: magnetic graphene, spin-dependent conductivity, two-dimensional spin valve, spintronics, spin-dependent Seebeck effect

Supplementary material for this article is available online

Abstract

Within the field of spintronics major efforts are directed towards developing applications for spin-based transport devices made fully out of two-dimensional materials. In this work we present an experimental realization of a spin-valve device where the generation of the spin signal is exclusively attributed to the spin-dependent conductivity of the magnetic graphene resulting from the proximity of an interlayer antiferromagnet, chromium sulfide bromide (CrSBr). We clearly demonstrate that the usage of the conventional air-sensitive 3D magnetic contacts can be fully avoided when graphene/CrSBr heterostructures are employed. Moreover, apart from providing exceptionally long spin relaxation length, the usage of graphene for both generation and transport of the spin allows to automatically avoid the conductivity mismatch between the source and the channel circuits that has to be considered when using conventional low-resistive contacts. Our results address a necessary step in the engineering of spintronic circuitry out of layered materials and precede further developments in the area of complex spin-logic devices. Moreover, we introduce a fabrication procedure where we designed and implemented a recipe for the preparation of electrodes via a damage-free technique that offers an immediate advantage in the fields of air-sensitive and delicate organic materials.

Giant magnetoresistance effect [1, 2] and spin-transfer torque [3, 4] phenomena have already allowed for a breakthrough spin-based technology within the area of memory-related applications. Yet, the utilization of the spin degree of freedom within the scope of the semiconductor industry remains limited [5]. In order to progress further and make practical use of the spin transport functionality one has to advance substantially in every constituent of the spin transport devices. Fortunately, layered materials, being both rather versatile as a family [6–8] and easy to assemble into a heterostructure [9], offer a promising pathway to take in the view of both efficiency and size miniaturization and, thus, have become the main material choice for spintronic devices [10–12].

Yet graphene cannot offer the means for creation, manipulation and detection of the spins. Other two-dimensional (2D) materials, however, can supplement what graphene lacks: when it is combined in a single heterostructure with an appropriately chosen companion it attains a spin-to-charge coupling via the proximity effect that allows for an active generation and control of spin. Such possibility to combine the properties of different materials in a single structure has recently driven the booming interest in the van der Waals heterostructures [12, 15–17]. Of direct relevance for the spintronics are the reports that experimentally demonstrate spin Hall [18], Rashba–Edelstein [19–21], Zeeman spin Hall [22, 23], anomalous Hall [24–26] and spin-dependent Seebeck [26] effects, spin-dependent conductivity and other transport phenomena appeared/modified due to the present spin–orbit and/or exchange interactions induced in graphene [27–34].
Within this work we demonstrate the generation of the spin current exclusively by the graphene itself, which is possible when it is placed on top of a layered magnetic material such as chromium sulfide bromide (CrSBr). The CrSBr is an interlayer antiferromagnet with an inplane magnetic easy-axis [35, 36]. As it was demonstrated in [26], in a graphene/CrSBr heterostructure the large exchange shift of the band structure (estimated experimentally to be $\sim 20$ meV, figure 2(a)) results in a considerable difference between the conductivities for the carriers of opposite spin alignment, i.e. $\sigma_u \neq \sigma_d$. This directly implies a finite spin polarization of the graphene conductivity defined as $P_{Gr} = \frac{\sigma_u - \sigma_d}{\sigma_u + \sigma_d}$.

The finite $P_{Gr}$ grants graphene an active role in the generation and detection of the spin signal [37]. A charge current $I_c$ when passing through the graphene channel, generates an associated spin current $I_{PGr}$ and vice versa. The initial experiments were so far performed on devices that had only spin-polarized electrodes which also inevitably contributed to the spin injection/detection [26]. In contrast, here we prepare our stacks having both types of contacts (magnetic and non-magnetic) and demonstrate a distinct generation of the spin signal in graphene by using TiOx/Au electrodes only, without having the magnetic contacts involved in the spin current generation circuit. In this case the spin generation takes place exclusively within the graphene, which becomes a 2D source of the spin signal.

For the preparation of the Au contacts we developed a novel recipe that allows less mechanical stress exerted on graphene during the pick-up procedure. In addition, it avoids multiple direct spin-coating of graphene with poly(methyl methacrylate) (PMMA), which is known to introduce additional residues on the graphene surface. Our damage-free fabrication procedure has a great potential to be particularly beneficial for the materials that cannot withstand regular lithography-based preparation of metallic contacts while having similar flexibility and resolution. The newly developed part of the sample preparation is summarized in the schematics and sample images shown in figure 1 and is explained in details in the section 3. In short, we start with the exfoliation of hBN and fabrication of the Ti/Au contacts and gates (by e-beam lithography) on a water-soluble layer (Electra), as shown in panel (a). By spin-coating a PMMA layer on top and dissolving the Electra in water, we release the hBN–Ti–Au–PMMA from the SiO$_2$ substrate. The PMMA layer floating on water is picked up and brought on top of a polydimethylsiloxane (PDMS) stamp and then can be transferred on any targeted flake or substrate. In our case this hBN–Ti–Au–PMMA–PDMS structure was used to pick up a bilayer graphene flake and to transfer the resulting stack on top of a CrSBr flake. The resulting structure is shown in figure 1(b). Graphene and CrSBr flakes were exfoliated in advance on two separate Si/SiO$_2$ substrates. Panel (c) in figure 1 gives the optical image of the sample taken just before depositing the magnetic electrodes made of AlOx/Co.

Overall, our observations confirm the robustness and consistency of the devices based on graphene/CrSBr stacks and offer an evident experimental demonstration of the generation of the spin current distinctly within a 2D system without the usage of conventional ferromagnetic contacts. Importantly, when graphene is used as both source and transport channel for the spin signal, the conductivity mismatch between the impedances of the source and transport circuits is automatically avoided which is not the case for the commonly used metallic contacts. Moreover, the spin relaxation length in magnetic graphene is much larger than that in the conventional ferromagnetic materials thus suggesting it as a more efficient source of the spin signal. Finally, being atomically thin, graphene allows an effective modulation of its Fermi level which in turn is expected to result in an active control of the spin valve action by the electric gate. All these facts promote graphene/CrSBr based devices as a very promising system for realizing spin functionality in a fully 2D system where the spin action is controlled exclusively by electrical means.
of the currents flows through the channel with the lowest resistivity ($\rho_\text{inj}$ for the case shown in figure 2(a)). When the charge current is passed through the left part of the channel only, the created discontinuity of the spin current at the injection point generates a spin accumulation. From there, the spin accumulation decays exponentially with the characteristic length scale $\lambda$. Considering the circuit shown in figure 2(b), it is also apparent that the injection circuit is a Wheatstone bridge. Passing the current through it will result in a voltage difference (spin accumulation) appearing between the ‘up’ and ‘down’ channels when the bridge is unbalanced.

The described above spin generation mechanism is very similar to a conventional one when the charge current is passed between a ferromagnetic material and a non-magnetic one [38], yet it does not require an additional 3D ferromagnetic electrode. Similarity between these mechanisms is clear from our resistance model (figure 2(b)) and is also directly reflected in the appearance of the relevant terms in the expression for the associated non-local resistance $R_{\text{nl}}$ derived in [26]:

$$ R_{\text{nl}} = \frac{\lambda R_{\text{sq}}}{2W(1 - R^2_{\text{Gr}})} e^{-L/\lambda}(P_i - P_{\text{Gr}})(P_d - P_{\text{Gr}}). $$

(1)

Here $W$, $R_{\text{sq}}$ and $\lambda$ are width, square resistance and spin relaxation length of the channel, respectively. The formula is derived for the case when both injecting and detecting contacts have a finite $P_i$ and $P_d$. The first pair of the parenthesis represents the total injection efficiency of the circuit where $P_{\text{Gr}}$ and $P_i$ enter in a very similar fashion.

The second parenthesis in equation (1) represents the reciprocal process in the spin circuitry, i.e. spin detection. There are two components related to

**1. Results and discussion**

The graphene conductivity in the proximity of CrSBr becomes spin polarized due to the induced exchange splitting, figure 2(a). The presence of the exchange interaction implies a shift in energy between electron states polarized in ‘up’ and ‘down’ directions. We consider these ‘up’ and ‘down’ electrons as two separate carrier species of the current in our magnetic graphene which are dissociated from each other but can still communicate via spin relaxation processes. Schematically such two-channel model can be depicted in an electrical circuit as shown in the panel (b) of the figure 2, where $\rho_u$ and $\rho_d$ are corresponding resistivities in each of the two channels, $\rho_i$ (in units of $\Omega \cdot \text{m}$) is the resistivity that represents the connection between the two channels via the present spin relaxation processes. Each electrical connection to the channel has to couple to both ‘up’ and ‘down’ channels via the corresponding contact resistances. For the circuit shown in figure 2(b) detection of the spin signal (right side of the circuit) is realized with the use of the magnetic material (Co). This implies that the contact resistances that couple the contact to the two spin channels are not equal, i.e. $R_{\text{det}}^u \neq R_{\text{det}}^d$, and that the spin polarization of this contact resistance $P_{\text{det}}$ is nonzero. Contrary to the common implementation of the spin generation circuit, for the injection of the current here we use a non-magnetic electrode. In this case the spin polarization of the contact resistances $P_i$ is exactly zero since $R_{\text{inj}}^u = R_{\text{det}}^u = R_{\text{inj}}^d$. Nonetheless, since the spin polarization of the channel resistance is finite, the spin current is still generated by graphene itself.

The mechanism of generation of the spin accumulation in magnetic graphene can be understood as follows: in a homogeneous magnetic system the charge current is spin polarized, since a larger portion
the spin polarization of the magnetic graphene and to that of the magnetic detector electrode. The two associated detection mechanisms enter the equation in a similar way, which is also reflected by the resistance circuit.

In the experiment described here we inject the current via a non-magnetic electrode (figure 2(c)). Thus, \( P_1 = 0 \), which modifies the formula for the measured non-local signal as \( R_{\text{nl}} \propto (P_1 - P_{\text{GR}})(P_2 - P_{\text{GR}})|_{P_1 = 0} = P_{\text{GR}}(P_2 - P_3) \) resulting in two contributions. The first one is proportional to \( P_{\text{GR}} \) and gives a positive spin-related background which does not depend on the relative orientation of the magnetization of the top most layer of CrSBr, \( M_{\text{CrSBr}} \), with respect to the magnetization of the detector. The second term is proportional to the product \( P_{\text{GR}} \cdot P_3 \) and results in the two levels of the non-local resistance depending on the relative alignment of the magnetizations. This is exactly confirmed with the experiment shown in figure 2(d) where the non-local spin valve is measured as a voltage difference between cobalt contacts C4 and C1 when the AC current is supplied between the main gold injector A5 and a reference cobalt contact C6 (contacts are numbered in figure 1(c)). The measured voltage difference is normalized by the applied current which gives the non-local resistance \( R_{\text{nl}} \). The measurement is performed as a function of the external magnetic field \( B_y \) applied along the easy magnetic axis of both cobalt electrodes and CrSBr flake (\( y \)-axis, also referred to as crystallographic \( b \)-axis of CrSBr [35, 36]). The value of the measured non-local resistance is observed to have two clear levels as it is also expected from the derivation. In the case of a regular graphene channel on a SiO\(_2\) substrate the same measurement would show no modulation of the non-local resistance since the injection current via a gold contact has zero spin polarization. Yet, once graphene is in the proximity to a ferromagnetic substrate, the inherited exchange interaction results in graphene acting as a source of the spin accumulation detected by the magnetic contact.

Note that in a homogeneously proximitized graphene channel, when only non-magnetic contacts are used, the separation of the spin-associated signal from the commonly present background (due to a non-ideal Ohmic current) is not straightforward. In that case the non-local spin signal would be independent of the direction of the graphene magnetization since both injection and detection circuits would share exactly the same magnetization determined by the underlying CrSBr. Therefore, we use spin-sensitive magnetic electrodes in order to unambiguously identify the spin currents generated when non-magnetic TiO\(_x\)/Au contacts are employed in the injection circuit.

From figure 2(d) we observe that the parallel alignment of \( M_{\text{CrSBr}} \) and \( M_{\text{C4}} \) gives a lower non-local resistance than the anti-parallel alignment. Therefore, based on the derived relation \( R_{\text{nl}} \propto P_{\text{GR}}(P_2 - P_3) \) we conclude that \( P_{\text{GR}} \) is of the opposite sign compared to \( P_{\text{C4}} \). Under the same assumptions as described in [26] this may suggest that the graphene sample is hole doped which is in agreement with the dependence of graphene resistance on the applied \( V_{\text{G1}} \) (see SI, section 2). However, since we are not able to tune the Fermi level across the Dirac point in the graphene channel between contact A5 and C4 we cannot reliably determine the position of the Fermi level and, thus, the size of the induced exchange splitting. Nevertheless, assuming that spin polarizations of both graphene conductivity and cobalt contacts resistance are equal, we are able to estimate the spin relaxation length to be \( \sim 450 \text{ nm} \) (SI section 4).

Furthermore, our assumptions result in a rough estimate of the involved spin polarizations \( P_{\text{GR}} = -P_{\text{C4}} \approx 50\% \). The possible uncertainty in the estimation of \( P_{\text{GR}} \) can be due to several factors, e.g. inhomogeneity of the channel doping, non-equal spin polarization of the contact and graphene resistances, etc. In particular, the uncertainty in the spin-relaxation length estimate is the most probable reason for the rather large calculated spin polarization of graphene conductivity. Nevertheless, the obtained number still suggests a consistently high efficiency of the spin signal generation within the magnetic graphene, thus, implying that in our spin transport circuit we can substitute the commonly used magnetic contacts with the regular non-magnetic ones and still obtain a signal of a similar magnitude.

In order to explore the possibility to tune/change the sign of the spin polarization of graphene conductivity we added two top-gates G1 and G2 into the design of the device (figure 1(c)). In the full possible range of \([-10; 10]\) V applied to G1 we observe no significant change in the size of the switches yet there is a modulation of the background level (SI section 5, figure 4(d)) which is likely to be related to the change in the charge related background. Unfortunately, during the sweep of the gate G2, one of the side connections to the electrode A5 was lost and there was a significant change in the channel resistance covered by the hBN (SI section 2). After the sample change the same spin valve geometry as used for figure 2(d) still shows a characteristic spin valve measurement, yet the magnitude is decreased from \( \sim 30 \Omega \) to \( \sim 6.6 \Omega \) as seen in figure 3(b). From now on we will be discussing the sample in the altered state. Note that the estimation of the spin relaxation length is done for the changed state of the sample since the distance dependence of the signal was measured only after the sample changed. Therefore, the calculation of the spin polarization of graphene conductivity described earlier is done using the spin valve measurement shown in figure 3(b) where the size of the switch is 6.6 \( \Omega \).
Furthermore, we noticed that the exact switching behavior of $M_{CSB}$ direction is not always reproducible and does depend on a value of the magnetic field used for the alignment. This in fact may be expected considering that in our measurements we are mostly sensitive to the magnetization of the top most layer of the bulk CrSBr whereas the full magnetization behavior is determined by the anti-ferromagnetic interaction between the top most and its neighboring layers. In figure 3(b) we plot a spin valve measured with the same connections where for the positive range of the applied magnetic field the value of the $R_{sd}$ corresponds to an anti-parallel alignment. This implies that $M_{CSB}$ switched back towards positive direction of $y$-axis while the magnetic field was brought to zero after alignment at $-0.6$ T. As a consequence with further increase of the field only the switch of $M_{C4}$ is observed at $44$ mT after which $M_{CSB}$ and $M_{C4}$ remain in a parallel alignment.

Note that the switching field for the magnetizations of the used Co electrodes ($M_{C4}$, $M_{C3}$) is determined by their shape anisotropy and is below $100$ mT. This allows us to unambiguously identify the switches in the $R_{sd}$ associated with the switches of $M_{C4}$/$M_{C3}$. Thus, for all our samples we can always determine the alignment of the magnetization of the used Co electrode and that of the top most layer of CrSBr.

To complete the investigation of the induced spin accumulation we supplement spin valve measurements with the $R_{sd}$ dependence on the magnetic field applied perpendicular to the easy axis (along x-axis), figure 3(c). Black and red curves correspond to parallel and anti-parallel alignments between $M_{CSB}$ and $M_{C4}$, respectively. Applying the magnetic field perpendicular to the alignment of the injected spins usually results in a Hanle precession of the spins that takes place while they diffuse from the injector to the detector contact. Yet, in magnetic graphene the induced exchange field $B_{exch}$ is strong enough to destroy all the spin components except those that are (anti)parallel to the direction of $B_{exch}$ irrespective of relatively weak applied external field [26]. Under this condition spins are always aligned with the exchange field. The role of the external field is to change the directions of both the exchange field (parallel to $M_{CSB}$) and of the detecting electrode magnetization. Here and below we define the geometry where the non-local signal is measured with the magnetic field applied along x-axis as ‘Hanle’ geometry yet this is not a Hanle precession measurement as understood conventionally.

The magnetic detector is sensitive to the spin components that are (anti)parallel to its magnetization direction. Therefore, in this case the functional dependence of the change observed in $R_{sd}$ with applied $B_x$ is proportional to the cosine of the angle between $M_{CSB}$ and $M_{C4}$. Cobalt contacts are much softer magnetically compared to CrSBr along the x-axis. Based on superconducting quantum interference device (SQUID) measurements [35], within the magnetic field range of $B_x < 0.2$ T the direction of $M_{CSB}$ changes only by a few degrees while $M_{C4}$ becomes fully aligned with the field at $B_x \approx 0.2$ T. Therefore, irrespective of the initial alignment between $M_{C4}$ and $M_{CSB}$ at $B_x \approx 0.2$ T both black and red dotted curves merge and continue jointly at higher fields until the direction of $M_{CSB}$ saturates along the y-axis at $B_x \approx 1.4$ T. Above this value
both cobalt contact magnetization and injected spin direction coincide again and therefore the signal recovers its initial value at $B_x = 0$ T for the parallel alignment.

Together with the first harmonic response of the lock-in amplifier we collected the second harmonic that is commonly associated with the phenomena driven by the temperature gradient in the graphene channel induced by Joule heating. In figures 3(e) and (f) both the second harmonic spin valve and dependence of the non-local signal on $B_x$ are shown, measured at the same time as those given in panels (b) and (c). Similar to the first harmonic response there are two distinct levels of $R_{nl}^2$ depending on the relative alignment of $M_{CA}$ and $M_{CSB}$, as expected. These observations clearly identify the spin origin of the measured signal and confirm its attribution to the spin-dependent Seebeck effect which results from the induced exchange interaction as in [26]. Charge current generates a Joule heating which, due to the present spin-dependent Seebeck effect, results in a finite spin current and spin accumulation that is sensed by the ferromagnetic detector circuit. In [26] it was concluded that the sign of the second harmonic signal does not depend on the position of the Fermi level which means that the switching behavior has to be identical throughout different samples irrespective of the doping level. This is experimentally justified here since the sign of the switch is the same as that reported in [26], thus, further confirming the consistency of our interpretation of the results.

In figure 3(b) next to the main $\sim6.6\Omega$ switch we observe a much smaller one of $\sim1\Omega$ in size which occurs at $B_x = \pm0.38$ T (not visible in the second harmonic signal). In order to understand the origin of this additional switch, we compare the signal collected at the contacts pair C4-C1 with that collected at the pair C3-C1. Similar to C4, contact C3 is made out of cobalt but is placed further away at a distance of $1.9\mu$m from the Au injector A5 (center-to-center).

The corresponding spin valve (1st and 2nd harmonic) and the dependence on $B_x$ (1st harmonic) are displayed in figures 4(a)–(c). First of all, the two switches occurring at $B_y = -32$ mT and $B_y = -0.21$ T are clearly associated with spin-related signal as it fully complies with the expected behavior. Namely, the non-local resistance switches up at the moment of the switch of the magnetization of contact C3 and it switches down together with the switch of $M_{CSB}$. Furthermore, when comparing the pairs C4-C1 and C3-C1 we find that the size of the spin signal decreases substantially with the distance between the injector and detector electrodes. This is fully in accordance with the expected change of the spin signal due to the spin relaxation processes.

Significantly, the magnitude of the additional switch is almost the same as for pair C4-C1 clearly indicating minimal scaling with the distance. As seen, its size is comparable with the spin-associated switch which implies that not only the spin valve but also the dependence on $B_x$ should be largely affected by this additional contribution. Indeed, as seen in figure 4(c) the shape of the dependence is quite different from what is expected and observed for example for the pair C4-C1 (figure 3(c)). Specifically one could point out that at high enough field $B_x \gtrsim 2$ T, when all the magnetizations are assumed to be aligned with the field, the value of the non-local resistance does not saturate at the same level as $R_{nl}(B_x = 0)$. The distinction between the spin-related and the additional contributions is further accentuated by the apparent absence of the former one in the second harmonic spin valve, figure 3(b).

All these observations and particularly the absence of the signal scaling with the distance hint at the origin of the additional contribution to be associated with the charge transport through a parallel channel. In fact, CrSBr is a semiconductor that can have a finite conductivity due to the residual doping, although it is expected to be much lower.
than conductivity of graphene [35] (also see SI). The dependence of CrSBr resistance on the applied magnetic field (also in [35] and SI) further indicates that the behavior shown in figures 4(a) and (c) is likely to be a combination of the charge related contribution associated with the CrSBr magnetoresistance and the spin-related contribution originating from the spin transport in graphene. Charge transport in CrSBr is expected to be controlled by the position of the Fermi level that is tuned by applying a gate voltage. Therefore, we studied how the spin valve measurements on contacts pair C4-C1 changes when the voltage is applied on the local gate G1 within the range [−10; 10] V. In figure 4(d) the corresponding spin valves are plotted with an offset for clarity. Evidently the spin-associated signal does not change significantly under applying the gate voltage whereas the additional contribution does get suppressed at $V_{G1} = 10$ V. This can be understood assuming that by applying positive gate voltage we shift the Fermi level in CrSBr more into the band gap thus reducing its conductivity and blocking the unwanted parallel conduction channel. Thus, we conclude that the observed additional contribution to the measured signal is likely to be due to the finite resistivity of the CrSBr, yet we show that it is possible to distinguish it from the spin-related component by studying the dependence of it on the distance and/or gate voltage.

2. Conclusions

We have performed the non-local measurements in graphene/CrSBr heterostructure in both first and second harmonic and in both spin valve and Hanle geometries. We have demonstrated that by using exclusively a non-magnetic electrode we are able to create a finite spin accumulation inside the magnetic graphene. Moreover, the usage of graphene/CrSBr heterostructure for spin injection/transport offers other immediate advantages compared to conventional 3D magnetic electrodes. Firstly, the conductivity mismatch between the spin source and transport channel is automatically avoided which simplifies the optimization of the performance of the spintronic circuit. Secondly, a large spin relaxation length in graphene suggests higher efficiency of magnetic graphene as a source of the spin signal in comparison to the conventional ferromagnetic contacts. Finally, a large Seebeck coefficient of graphene ensures the presence of the spin-dependent Seebeck effect and offers even richer functionality with coupling the spin and heat currents. Overall, our findings confirm the graphene/CrSBr heterostructure as a robust platform for studying spin transport in a 2D magnetic channel. The distinct generation of spin signal within a 2D system when using nonmagnetic electrodes together with the potential tunability of the spin valve action by an electrical gate introduces graphene/CrSBr-based devices as a technologically relevant block for building fully 2D spintronic/spin-caloritronic devices. In addition, we have developed and implemented in the measured devices a novel damage-free recipe for the preparation of the contacts separately from the studied flake/material. Such recipe is of a great value for air sensitive as well as for organic materials.

3. Methods and sample fabrication

Devices D1–D3 were prepared starting with exfoliation of the necessary layered components, namely graphene, CrSBr and hBN. Graphene and CrSBr are exfoliated on top of doped Si/SiO$_2$ substrates. The details of the growth of CrSBr bulk crystals are given in [26, 35]. The flakes with appropriate thicknesses are identified by their optical contrast with respect to the SiO$_2$/Si substrate. For D1 (discussed in the manuscript) we used bilayer graphene and 20–40 nm thick CrSBr flakes. The hBN flake is exfoliated on a separate Si/SiO$_2$ substrate (SI, figure S4, step 1) that is preliminary covered with a water soluble conductive polymer Electra (AR-PC 5090.02, Allresist). Electra is spin-coated with the rate of 1000 rpm and baked afterwards at 95°C for 1 min on the hot plate. The thickness of the resulting Electra layer is about 200 nm. When an appropriate hBN flake of 20–50 nm thickness is selected (that is intended to be an insulator for the top gate), a 500 nm PMMA (4%, 950 K) layer is spin-coated on top of the Si/SiO$_2$/Electra/hBN substrate at a rate of 1000 rpm (SI, figure S4, step 2). A freshly covered substrate is baked on the hot plate at 180°C for 1 min. By means of e-beam lithography an appropriately designed structure is exposed in PMMA, including contacts and the top gate electrodes. After developing, the substrate is loaded into an e-beam evaporation setup where 0.5 nm of Ti and 90 nm of Au is deposited. Lift-off is done in acetone either at room or elevated temperature of 45°C and results in the structure schematically shown in figure 1(a) (top). We use here the fact that Electra does not get dissolved in either acetone nor developer solution and stays intact.

Resulting Si/SiO$_2$/Electra/hBN/Ti/Au is again covered with PMMA using the same coating parameters as described earlier (SI, figure S4, step 4). In the next step the sample is attached to a Scotch tape that has a 7 by 7 mm window centered at the hBN flake and then is immersed in water. Water dissolves Electra which leads to a gradual detachment of the Si/SiO$_2$ substrate that eventually sinks down while hydrophobic PMMA film stays floating on top of water together with the attached Scotch tape, figure 1(a) (middle, the Scotch tape is not shown in the schematics). The tape with the PMMA film is taken from water, dried in air and later put on top of a PDMS stamp with an Au/Ti/hBN structure.
facing outwards, figure 1(a) (bottom). At this stage the Ti layer is directly exposed to air and water and, thus, gets oxidized. Subsequently, the stamp with the PMMA film is used to pick up a chosen bilayer graphene flake from a separate Si/SiO2 substrate. The pick-up surface of the full stamp is flat since both hBN layer and TiOx/Au contacts are fully imbedded in the PMMA film. Graphene/hBN/Ti(TiOx)/Au/PMMA heterostructure is thereafter placed onto a targeted CrSBr flake. The final stack is shown in figure 1(b) where it is still covered with the PMMA layer. The same PMMA is used later on for the lithography and deposition of AlOx/Co contacts. Picture of device D1 after development and just before the deposition of AlOx/Co contacts is given in figure 1(c).

Our recipe offers several advantages from the perspective of fabrication of graphene-based samples that require different types of contacts simultaneously. First of all, before the pick-up of graphene flake, the hBN flake together with the Ti/Au contacts on Si/SiO2/Electra substrate is covered with a fully relaxed film of PMMA. This assures a smooth gap-less pick-up surface of the prepared mask. Second of all, the Ti/Au contacts are made in advance on a separate substrate, thus, fully avoiding the risks of any errors occurring during this procedure. Third of all, the graphene is covered by the polymer only once (during the pick-up). Normally, during the preparation of the graphene-based structure with the two different types of contacts the sample (including graphene) is covered by the polymer three times: once during the stacking (when it is done using the polycarbonate film) and two times for fabricating separately the two different contact types. Reducing the number of steps where graphene is covered by the polymer is expected to lower the amount of residues on its surface.

Overall we have measured three devices where we were able to see the switching behavior of the non-local resistance associated with the spin signal where non-magnetic contacts were used either as the current injector or as the voltage detector. Full characterization of the spin transport parameters was done only for the sample D1. The size of the observed spin signal was found to range from 50 mΩ for the samples D2-D3 to 30 Ω for the sample D1 indicating a large variation in the relevant sample parameters (spin relaxation length, distance between the injector/detector electrodes, spin polarization of conductivity of graphene and Co electrodes, etc.). Further details of the fabrication and measurements on the other samples are given in the supplementary information.

Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

Acknowledgments

We would like to thank T J Schouten, H Adema, A Joshua, H de Vries and J G Holstein for technical support. The presented research was funded by the Dutch Foundation for Fundamental Research on Matter (FOM) as a part of the Netherlands Organisation for Scientific Research (NWO), the European Union’s Horizon 2020 research and innovation program under Grant Agreement Nos. 785219 and 881603 (Graphene Flagship Core 2 and Core 3), NanoNed, the Zernike Institute for Advanced Materials, and the Spinoza Prize awarded in 2016 to B J van Wees by NWO. Synthesis, structural characterization and magnetic measurements received support as part of Programmable Quantum Materials, an Energy Frontier Research Center funded by the U.S. Department of Energy (DOE), Office of Science, Basic Energy Sciences (BES), under award DESC0019443. A D is supported by the NSF graduate research fellowship program (DGE 16-44869).

ORCID iD

Alexey A Kaverzin 🏃‍♂️ https://orcid.org/0000-0001-5057-5038

References

[1] Balibich M N, Broto J M, Fert A, van Dau F N, Petroff F, Etienne P, Creuzet G, Friederich A and Chazelas J 1988 Giant magnetoresistance of (001)Fe/(001)Cr magnetic superlattices Phys. Rev. Lett. 61 2472
[2] Binasch G, Grünberg P, Saurenbach F and Zinn W 1989 Enhanced magnetoresistance in layered magnetic structures with antiferromagnetic interlayer exchange Phys. Rev. B 39 4828
[3] Słonczewski J C et al 1996 Current-driven excitation of magnetic multilayers J. Magn. Magn. Mater. 159 11
[4] Myers E, Ralph D, Katine J, Louie R and Buhman R 1999 Current-induced switching of domains in magnetic multilayer devices Science 285 867–70
[5] Awschalom D D and Flatté M E 2007 Challenges for semiconductor spintronics Nat. Phys. 3 153–9
[6] Gong C et al 2017 Discovery of intrinsic ferromagnetism in two-dimensional van der Waals crystals Nature 546 263–9
[7] Burch K S, Mandrus D and Park J-G 2018 Magnetism in two-dimensional van der Waals materials Nature 563 47–52
[8] Gong C and Zhang X 2019 Two-dimensional magnetic crystals and emergent heterostructure devices Science 363 eaav4450
[9] Geim A K and Grigorieva I V 2013 Van der Waals heterostructures Nature 499 419–25
[10] Cardoso C, Soriano D, García-Martínez N and Fernández-Rossier J 2018 Van der Waals spin valves Phys. Rev. Lett. 121 067701
[11] Zhai X, Xu Z, Cui Q, Zhu Y, Yang H and Blanter Y M 2021 Electrically controllable van der Waals antiferromagnetic spin valve Phys. Rev. Appl. 16 014032
[12] Sierra J F, Fabian J, Kawakami R K, Roche S and Valenzuela S O 2021 Van der Waals heterostructures for spintronics and opto-spintronics Nat. Nanotechnol. 16 856–60
[13] Han W, Kawakami R K, Gmitra M and Fabian J 2014 Graphene spintronics Nat. Nanotechnol. 9 794–807 Review
[14] Drögeler M, Franzen C, Volmert F, Pohlmann T, Banszerus L, Wolter M, Watanabe K, Taniguchi T, Stampfer C and Beschoten B 2016 Spin lifetimes exceeding 12 ns in graphene nonlocal spin valve devices Nano Lett. 16 3533–9
[15] Garcia J H, Vila M, Cummings A W and Roche S 2018 Spin transport in graphene/transitions metal dichalcogenide heterostructures Chem. Soc. Rev. 47 3539–79
[16] Zollner K, Conita M and Fabian J 2020 Swapping exchange and spin-orbit coupling in 2D van der Waals heterostructures Phys. Rev. Lett. 125 196402
[17] Kurebayashi H, Garcia J H, Khan S, Sinova J and Roche S 2022 Magnetism, symmetry and spin transport in van der Waals layered systems Nat. Rev. Phys. 4 150–66
[18] Safer C, Inglá-Ayés J, Herling F, Garcia J H, Vila M, Ontoso N, Calvo M R, Roche S, Hueso L E and Casanova F 2019 Room-temperature spin Hall effect in graphene/MoS₂ van der Waals heterostructures Nano Lett. 19 1074–82
[19] Ghiasi T S, Kaverzin A A, Blah P J and van Wees B J 2019 Charge-to-spin conversion by the Rashba–Edelstein effect in two-dimensional van der Waals heterostuctures up to room temperature Nano Lett. 19 5959–66
[20] Benítez L A, Torres W S, Sierra J F, Timmermans M, Garcia J H, Roche S, Costache M V and Valenzuela S O 2020 Tunable room-temperature spin galvanic and spin Hall effects in van der Waals heterostructures Nat. Mater. 19 170–75
[21] Khokhriakov D, Hoque A M, Karpik B and Dash S P 2020 Gate-tunable spin-galvanic effect in graphene-topological insulator van der Waals heterostructures at room temperature Nat. Commun. 11 3657
[22] Wei P et al. 2016 Strong interfacial exchange field in the graphene/Exs heterostructure Nat. Mater. 15 711–6
[23] Behera S K, Bora M, Chowdhury S S P and Deb P 2019 Proximity effects in graphene and ferromagnetic CrBr₃ van der Waals heterostructures Phys. Chem. Chem. Phys. 21 25788–96
[24] Wang Z, Tang C, Sachs R, Barlas Y and Shi J 2015 Proximity-induced ferromagnetism in graphene revealed by the anomalous Hall effect Phys. Rev. Lett. 114 016603
[25] Tang C, Cheng B, Aldosary M, Wang Z, Jiang Z, Watanabe K, Taniguchi T, Bockrath M and Shi J 2018 Approaching quantum anomalous Hall effect in proximity-coupled YIG/graphene/h-BN sandwich structure APL. Materials 6 026401
[26] Ghiasi T S, Kaverzin A A, Dismukes A H, de Val D K, Roy X and van Wees B J 2021 Electrical and thermal generation of spin currents by magnetic bilayer graphene Nat. Nanotechnol. 16 788–94
[27] Ghiasi T S, Inglá-Ayés J, Kaverzin A A and van Wees B J 2017 Large proximity-induced spin lifetime anisotropy in transition-metal dichalcogenide/graphene heterostructures Nano Lett. 17 7528–32
[28] Benítez L A, Sierra J F, Torres W S, Arrighi A, Bonell F, Costache M V and Valenzuela S O 2018 Strongly anisotropic spin relaxation in graphene–transition metal dichalcogenide heterostructures at room temperature Nat. Phys. 14 303
[29] Zühlmann S, Cummings A W, Garcia J H, Kedves M, Watanabe K, Taniguchi T, Schönemberger C and Makk P 2018 Large spin relaxation anisotropy and valley-Zeeman spin-orbit coupling in WSe₂/Gr/hBN heterostructures Phys. Rev. B 97 075434
[30] Inglá-Ayés J, Herling F, Fabian J, Haeso L E and Casanova F 2021 Electrical control of valley-Zeeman spin-orbit-coupling-induced spin precession at room temperature Phys. Rev. Lett. 127 047202
[31] Leutenantsmeyer J C, Kaverzin A A, Wojtaszek M and van Wees B J 2016 Proximity induced room temperature ferromagnetism in graphene probed with spin currents 2D Mater. 4 014001
[32] Singh S, Katoch J, Zhu T, Meng K-Y, Liu T, Brangham J T, Yang F, Flatté M E and Kawakami R K 2017 Strong modulation of spin currents in bilayer graphene by static and fluctuating proximity exchange fields Phys. Rev. Lett. 118 187201
[33] Karpik B et al. 2020 Magnetic proximity in a van der Waals heterostructure of magnetic insulator and graphene 2D Mater. 7 015026
[34] Wu Y et al. 2020 Large exchange splitting in monolayer graphene magnetized by an antiferromagnet Nat. Electron. 3 604–11
[35] Telford E J et al. 2020 Layered antiferromagnetism induces large negative magnetoresistance in the van der Waals semiconductor CrBr₃ Adv. Mater. 32 2003240
[36] Lee K, Dismukes A H, Telford E J, Wiscons R A, Wang J, Xu X, Nuckolls C, Dean C R, Roy X and Zhu X 2021 Magnetic order and symmetry in the 2D semiconductor CrBr₃ Nano Lett. 21 3511–7
[37] Zayets V 2012 Spin and charge transport in materials with spin-dependent conductivity Phys. Rev. B 86 174415
[38] Johnson M and Silsbee R H 1985 Interfacial charge-spin coupling: injection and detection of spin magnetization in metals Phys. Rev. Lett. 55 1790