Spin transport in multilayer graphene away from the charge neutrality point

Xin He^a, Yan Wen^a, Chenhui Zhang^a, Peng Li^{a,b}, Dongxing Zheng^a, Aitian Chen^a, Aurelien Manchon^a, Xixiang Zhang^{a,*}

^aPhysical Science and Engineering Division, King Abdullah University of Science and Technology (KAUST), Thuwal 23955-6900, Saudi Arabia

^bInstitute for Quantum Computing, Department of Electrical and Computer Engineering, University of Waterloo, Waterloo N2L, Canada

ABSTRACT

Graphene is considered as a promising material in spintronics due to its long spin relaxation time and long spin relaxation length. However, its spin transport properties have been studied at low carrier density only, beyond which much is still unknown. In this study, we explore the spin transport and spin precession properties in multilayer graphene at high carrier density using ionic liquid gating. We find that the spin relaxation time is directly proportional to the momentum relaxation time, indicating that the Elliott-Yafet mechanism still dominates the spin relaxation in multilayer graphene away from the charge neutrality point.

Keywords: Multilayer graphene, Ionic liquid gating, Spin transport, Elliott-Yafet mechanism

1. Introduction

Due to the high charge-carrier mobility [1], weak spin-orbit coupling [2, 3], and efficiently gate-tunable transport properties [4], graphene has become a prospective material for spintronics [5-7]. When the study of spintronics in graphene just rose, the spin-relaxation time and spin-relaxation length were measured to be 170 ps and 2 μm [8], respectively, which are much higher than those in the traditional materials [9], yet much lower than the theoretical predictions [10, 11]. The poor quality of the tunneling layer [12], the polymer residues during the fabrication process [13], and the presence of scattering centers in the substrate [14, 15] are considered to be the main factors that degrade the spin transport properties of graphene. Correspondingly, several measures have been proposed to address these problems, such as the optimized insulating layer which facilitates the tunneling injection [12, 16-18], the hexagonal boron nitride (hBN) layer which serves as a substrate or encapsulating layer to provide a smooth interface with reduced trapped charges [19], and the suspended sample structure to avoid impurity scattering [14, 20, 21]. Thanks to these efforts, the spin-relaxation time and spin-relaxation length of graphene have been improved up to 12.6 ns and 30.5 μm [20], respectively, approaching the theoretical prediction gradually [10, 11]. Alternatively, multilayer graphene was also proposed as a promising strategy to improve the spin transport performance since the outer layers of multilayer graphene can screen the electrical potential of impurities [22, 23]. However, this original study was limited to relatively low carrier density (close to $1.25 \times 10^{13} \text{cm}^{-2}$) because of the low efficiency of the traditional solid gate [23]. Hence, the conclusion that the spin relaxation is mainly due to the Elliott-Yafet mechanism was drawn only in the vicinity of the charge neutrality point [23]. When the Fermi level is shifted away from the charge neutrality point, the energy band structure and thus the electron properties will be quite different. Furthermore, the electron-electron interaction will be weakened due to the high-carrier density induced charge screening, which will influence the momentum relaxation time. Therefore, how spin transport evolves and which mechanism dominates the spin relaxation in the high-carrier-density range of multilayer graphene is yet to be explored.

Ionic liquid (IL) gating has been widely used in 2D materials due to its high efficiency in tuning carrier density [24, 25]. It has been central to the exploration of the electronic and optical properties [26-29], for the tuning and realization of voltage-controlled superconductivity [30],

and manipulation of the magnetic properties [31]. These studies all prove that IL gating is a mature and reliable technique.

In the present study, we explore the spin transport in multilayer graphene by tuning its carrier density using IL gating at 200 K. In a heavily doped multilayer graphene sample, the electron and hole densities reach as high as $4.3 \times 10^{13} \text{ cm}^{-2}$ and $5.7 \times 10^{13} \text{ cm}^{-2}$, respectively. The spin relaxation time and the momentum relaxation time are found to follow a directly proportional relationship, which indicates that the Elliott-Yafet mechanism, rather than the D'yakonov-Perel' mechanism, plays an important role in the high-carrier density range.

2. Results and analysis

The schematic and the optical images of the device for the spin and charge transport measurements are shown in Fig. 1(a-c), while the image with IL on the device can be found in Supplementary Fig. S1. All the electrodes are made of Co/Au (60 nm/ 20 nm). As presented in Fig. 1(a), the outer four electrodes (1, 2, 5, 6) are designed for charge transport measurement using a four-probe configuration, while the inner four electrodes (2, 3, 4, 5) are designed for spin transport measurement; the widths of the central two electrodes (3, 4) are fabricated to be 270 nm and 660 nm, respectively, forming injector and detector electrodes with different coercive fields. The thickness of the multilayer graphene flake was measured to be 4 nm by atomic force microscope. Considering the spacing of adjacent graphene layers is 0.335 nm [23], the layer number of our sample can be estimated to be 12, with an error of about 1 layer. Note that all the measurements in this report were performed at 200 K unless otherwise specified.

Fig. 1. (a) Schematic of a nonlocal spin-valve device under IL gating. (b) Optical image of a nonlocal spin-valve device (scale bar: 50 μm). A 12-layer graphene flake is used as the channel. (c) Zoom in on the image framed by the red open box in (b); the scale bar is 10 μm .

After dropping ionic liquid to cover the whole sample and setting the IL gate floating, the measurements for both spin transport and spin precession were first conducted at 300 K. Subsequently, the system was cooled down quickly to 200 K, the freezing point of ionic liquid [32], so as to avoid chemical reaction between graphene and ionic liquid. The same experiments were also performed at 200 K. As shown in Fig. 1(a), under an in-plane magnetic field (B_y), a current I is injected from electrode 3 (injector) to 2, and then spins accumulate underneath electrode 3 and diffuse along the graphene channel to both sides, inducing a voltage drop V_{NL} between electrode 4 (detector) and 5 and defining the nonlocal resistance $R_{\text{NL}} = V_{\text{NL}}/I$ (spin transport) [8]. The detector probes the chemical potential of the spins impinging from the injector. Therefore, switching the magnetization configuration of the injector and detector from parallel (P) to antiparallel (AP) by sweeping the in-plane magnetic field leads to a sudden decrease of both the nonlocal voltage (ΔV_{NL}) and the nonlocal resistance ($\Delta R_{\text{NL}} = \Delta V_{\text{NL}}/I$), as shown in Fig. 2(a) [8, 33]. The non-local spin valve signal obtained at 200 K is enhanced in comparison to that measured at 300 K (Fig. 2(a)), due to the decreased Coulomb scattering in graphene with relatively low carrier mobility, which is consistent with previous reports [34, 35]. Applying an out-of-plane magnetic field (B_z), a current I is also injected from electrode 3 to 2, and spins precess as they diffuse from electrode 3 to 4, inducing a voltage drop V_{NL} between electrode 4 and 5 and defining the nonlocal resistance $R_{\text{NL}} = V_{\text{NL}}/I$ as well (spin precession). The non-local Hanle spin precession signals in both 300 K and 200 K are plotted in Supplementary Fig. S2(a) and (b), respectively. At low magnetic fields, the signal weakens because of the reduction of the spin polarization upon increasing the field from zero. When the signal corresponding to the P (AP) configuration decreases (increases) to zero, it indicates that the average angle of the precession reaches 90° [36]. Upon increasing the field further, R_{NL} changes sign and reaches a minimum (maximum) gradually, indicating the reversal of spins (the average precession angle reaches 180°) [36]. However, even though the vertical magnetic field was applied up to 6 T, we could not observe the saturation of R_{NL} that indicates the magnetization of the electrodes has completely rotated out of the plane. This is because the large-resistance

background blurs the spin precession signal [37]. Thus, we can only estimate that R_{NL} saturates at around 1.5 T, at which the slopes of the curves decrease suddenly (Supplementary Fig. S2).

Fig. 2. (a) Non-local spin valve signal of the 12-layer graphene flake applying an in-plane magnetic field at 200 K and 300 K. The magnetization configurations of the electrodes are illustrated by the arrows. The amplitude of the nonlocal resistance, ΔR_{NL} . (b) σ (left) and ΔR_{NL} (right) as functions of the carrier density n , respectively.

To verify the validity of IL gating at 200 K, we performed electronic measurements and then obtained the standard transfer curve of graphene, with the charge neutrality point located at $V_{CNP} = 0.59 \text{ V}$ (Fig. S3). This means that IL gating works even at 200 K, suggesting that the molecules of IL are still mobile at 200 K. Moreover, from Fig. S3, the mobilities (μ) of both electron and hole are extracted to be $242 \text{ cm}^2/\text{V} \cdot \text{s}$ and $197 \text{ cm}^2/\text{V} \cdot \text{s}$, respectively, by using the equation, $\mu = \Delta\sigma/(C_{IL}\Delta V_g) = (\Delta(I_{ds}/V_d) \cdot L/W)/(C_{IL}\Delta V_g)$, where σ is the conductivity, I_{ds} is the drain-source current, V_d is the drain-source voltage, V_g is the gate voltage, C_{IL} is the capacitance per unit area of IL, L and W are the channel length and width, respectively [38]. C_{IL} is obtained to be $2.86 \text{ } \mu\text{F}/\text{cm}^2$ and $2.55 \text{ } \mu\text{F}/\text{cm}^2$ for the electron and hole, respectively (Supplementary Note 1), much lower than the values reported previously [26, 39]. There are two factors leading to the small C_{IL} : the extra TiO_x layer and the screening effect in graphene. First, the inserted TiO_x layer (about 1 nm thick) increases the effective dielectric thickness of the geometrical capacitance C_g , which is defined by $C_g = \epsilon_{IL}\epsilon_0/d$, where ϵ_{IL} is the relative permittivity of ionic liquid, ϵ_0 is the vacuum permittivity, and d is the dielectric thickness [40]. Second, the charges distributed in the first 3 or 4 layers screen the electric field and quench its penetration, and then

C_g saturates to a constant as the layer number is larger than 4 [40]. In Fig. 2(b), the IL gate voltage is transformed to the carrier concentration N by using the formula, $N = C_{IL} \frac{(V_g - V_{CNP})}{e}$, where $e < 0$ is the elementary charge [32, 41]. Then the highest electron (hole) density can be calculated to be $4.3 \times 10^{13} \text{ cm}^{-2}$ ($5.7 \times 10^{13} \text{ cm}^{-2}$). Also in Fig. 2(b), the changes of the non-local resistances were extracted by performing the non-local spin transport at a series of carrier densities, as a comparison to the carrier density dependence of the conductivity. The relationship between ΔR_{NL} and σ in Fig. 2(b) follows neither $\Delta R_{NL} \sim \sigma$ nor $\Delta R_{NL} \sim \frac{1}{\sigma}$, implying that the electrode contact could be a pinhole type [12]. As discussed in the previous study, the contact interface mainly affects the polarization of the injected current [23], and the pinhole-type contacts could also create long spin relaxation time and spin relaxation length [20], thus the contacts barely influence the spin diffusion and spin relaxation in our study.

Fig. 3. (a) Nonlocal Hanle spin precession signal of the 12-layer graphene flake at the electron density of $2.5 \times 10^{13} \text{ cm}^{-2}$, P and AP magnetizations of the central two electrodes are illustrated in the insets. σ (b), D_s (c), τ_s (d) and λ_s (e) as a function of the carrier density n , respectively.

The spin precession at different carrier densities is studied applying a perpendicular magnetic field at 200 K, as shown in Fig. 3. As an example, Fig. 3(a) shows the nonlocal Hanle

spin precession signals with both P and AP magnetization of the central electrodes at $V_g = 2$ V, corresponding to the electron density of $2.5 \times 10^{13} \text{ cm}^{-2}$. Since the magnetic field is applied perpendicular to the plane, the spins with in-plane orientation begin to precess, following the Bloch equation for the spin accumulation μ_S , $D_S \nabla^2 \mu_S - \frac{\mu_S}{\tau_S} + \omega_L \times \mu_S = 0$ [23]. The first, second and third term describes the spin diffusion, spin relaxation, and spin precession, respectively, with D_S is the spin diffusion coefficient, τ_S is the spin relaxation time, and $\omega_L = \frac{g\mu_B \mathbf{H}}{\hbar}$ is the Larmor frequency ($g = 2$ is the effective Lande factor, μ_B is the Bohr magneton, and $\mathbf{H}$ is the out-of-plane magnetic field). Moreover, D_S and τ_S can be extracted by fitting the Hanle precession signal at each carrier density with the solution of the above Bloch equation, $R_{NL}^{P(AP)} = (\pm) S_0 \int \frac{1}{\sqrt{4\pi D_S t}} \exp\left(-\frac{L^2}{4D_S t}\right) \cos(\omega_L t) \exp\left(-\frac{t}{\tau_S}\right) dt$ [42] (where $R_{NL}^{P(AP)}$ is the nonlocal magnetoresistance, $+$ ($-$) sign is for the P (AP) state, S_0 is the amplitude, and L is the channel length between the central two electrodes), while the spin diffusion length λ_S can be calculated by $\lambda_S = \sqrt{D_S \tau_S}$ [23]. Note that before fitting the result, the P curve of the Hanle effect was subtracted from the AP curve (e.g., Fig. 3(a)) to remove the background that is not related to the spin signal [23]. Therefore, the relationships between σ and n , D_S and n , τ_S and n , and λ_S and n are presented in Fig. 3(b-e), respectively. D_S , τ_S , and λ_S all diminish with decreasing the electron or the hole density, and reach the minimum values in the vicinity of the charge neutrality point, resembling the relationship between σ and n . Compared with the previous data obtained using the traditional solid gate in multilayer graphene [23], these parameters show more significant changes because of the stronger carrier density modulation induced by the IL gating. The asymmetry of D_S for electrons and holes in Fig. 3(c) may be due to the nonuniformities in the carrier density [43], while the asymmetry of τ_S in Fig. 3(d) could be traced to the different impacts of the spin scatterings to the electrons and holes.

Fig. 4. (a) Linear relationship between τ_S and D_S , extracted from Fig. 3(c) and (d). (b) Linear relationship between λ_S and D_S , extracted from Fig. 3(c) and (e).

In semiconductors, there are two main mechanisms that contribute to spin relaxation. The first one is the Elliott-Yafet mechanism, which stems from momentum transfer between spin and orbital angular momenta during momentum scattering events, resulting in τ_S directly proportional to τ_P (τ_P is the momentum relaxation time) [43, 44]. The second one is the D'yakonov-Perel' mechanism, which stems from scattering-induced destructive interference of spins coherently precessing around the spin-orbit field in the band structure. In this case, τ_S is inversely proportional to τ_P [43, 44]. Thus, to identify which spin scattering mechanism dominates the spin relaxation, we need to determine the relationship between τ_S and τ_P . Fig. 4(a) shows the linear dependence between τ_S and D_S ($\tau_S \propto D_S$), extracted from Fig. 3(c) and (d); while in Fig. 4(b), λ_S and D_S are also in direct proportion, extracted from Fig. 3(c) and (e). The larger slopes for the hole regimes indicates the averagely lower spin-flip probability for holes in each scattering [23]. These relationships are consistent with the previous studies in both monolayer and multilayer graphene [23, 43, 45]. Combining the facts that $D_S \propto D_P$ (D_P is the momentum diffusion coefficient) [23] and $D_P \propto \tau_P$ [42, 43], we obtain $\tau_S \propto \tau_P$. This indicates that the Elliott-Yafet mechanism still plays a more important role in the spin relaxation process of multilayer graphene at high carrier density [43, 44]. Moreover, a large perpendicular electric field can lead to a considerable in-plane Rashba spin-orbit field, which will affect the spin transport properties, i.e., the spin relaxation time [37]. In our experiment, the gate voltage up to 3 V is applied, and the electric field penetrates to the first 3 or 4 layers of graphene. Considering the ionic liquid molecule (diameter~1 nm) and the TiO_x layer (thickness~1 nm), the electric field reaches around

1 V/nm in graphene theoretically. However, we do not see the deviation of the spin relaxation time in Fig. 3(d), which means that the induced spin-orbit field is quite weak, because of the strong screening of the charges to the applied electric field.

3. Outlook

Recently, to realize more functions in graphene-based spin devices, the heterostructures such as graphene/topological insulators [46], graphene/magnetic insulator [47], and graphene/Transition metal dichalcogenides [48] have been constructed. These heterostructures combine both the excellent spin properties of graphene and the exotic properties of other materials via proximity effect. Moreover, the nano-graphene based spin device is also drawing more attention [49-52]. Due to the weak tuning capability of the traditional solid gate, the gate voltage that is used to switch on/off the spin current is always as high as several tens of volts. We believe that the IL gate can be used to improve the gating effect in these devices and allow them to work practically at low voltages.

4. Conclusion

In conclusion, we provide more insights into the spin dynamics in multilayer graphene and verified that IL gating is a powerful technique investigating spintronics in 2D materials. After applying IL gate, the electron and hole densities in multilayer graphene are increased up to $4.3 \times 10^{13} \text{ cm}^{-2}$ and $5.7 \times 10^{13} \text{ cm}^{-2}$, respectively. The spin relaxation time and the momentum relaxation time are found to follow a directly proportional relationship, indicating that the Elliott-Yafet mechanism still dominates the spin relaxation in multilayer graphene at high carrier density.

5. Methods

Multilayer graphene samples were exfoliated from natural graphite onto SiO_2/Si (300 nm / 500 μm) substrate. To avoid the extra etching process, the samples which were relatively long and narrow were located by optical microscope (Carl Zeiss Imager.A2 Vario with AxioCam

HRC), and then their thicknesses were confirmed by atomic force microscope (Asylum Research MFP-3D) using tapping mode. After that, 0.8 nm Ti was evaporated onto the samples using e-beam evaporator, and then they were exposed in air for 0.5 h to form an oxidation layer (the TiO_x layer not only plays a role of tunneling layer to conquer the conductance mismatch problem but also prevents the sample from reacting with ionic liquid) [53]. Subsequently, six electrodes were fabricated for each sample using e-beam lithography (EBL) and followed e-beam evaporation of Co/Au (60 nm / 20 nm). After the lift-off process, the samples were immersed in chloroform (60 °C, 4 hours) to remove the polymer residues to facilitate the usage of IL, which was (N,N-diethyl-N-methyl-N-(2-methoxyethyl)ammonium Bis(trifluoromethanesulfonyl)imide (DEME-TFSI)) (Kanto Chemical Co., Inc.). With Keithley 2636B applying IL-gate voltage, an alternating current of 20 μA at 13 HZ was applied by Keithley 6221 and the voltage was measured by lock-in amplifier SR830 in the spin transport measurements, while the alternating current was changed to 1 μA in the electronic transport measurements. The transfer curves in the supplementary material were measured using the Keithley 4200-SCS semiconductor characterization system.

Conflicts of interest

The authors declare no competing financial interests.

Acknowledgments

This publication is based on research supported by the King Abdullah University of Science and Technology (KAUST), under Award Nos. OSR-2018-CRG7-3717 and OSR-2017-CRG6-3427.

Appendix A. Supplementary data

Supplementary data to this article can be found online at XXXXXXXXXX.

251 **References**

- 252 [1] C.R. Dean, A.F. Young, I. Meric, C. Lee, L. Wang, S. Sorgenfrei, et al., Boron nitride substrates for high-
253 quality graphene electronics, *Nat. Nanotechnol.* 5 (2010) 722-726.
- 254 [2] M. Gmitra, S. Konschuh, C. Ertler, C. Ambrosch-Draxl, J. Fabian, Band-structure topologies of
255 graphene: Spin-orbit coupling effects from first principles, *Phys. Rev. B* 80 (2009) 235431.
- 256 [3] H. Min, J.E. Hill, N.A. Sinitsyn, B.R. Sahu, L. Kleinman, A.H. MacDonald, Intrinsic and Rashba spin-orbit
257 interactions in graphene sheets, *Phys. Rev. B* 74 (2006) 165310.
- 258 [4] F. Schwierz, Graphene transistors, *Nat. Nanotechnol.* 5 (2010) 487-496.
- 259 [5] M.V. Kamalakar, C. Groenveld, A. Dankert, S.P. Dash, Long distance spin communication in chemical
260 vapour deposited graphene, *Nat. Commun.* 6 (2015) 6766.
- 261 [6] S. Roche, A. Rinzler, J. Kerman, B. Beschoten, J.C. Charlier, M. Chshiev, et al., Graphene spintronics: the
262 European Flagship perspective, *2d Materials* 2 (2015) 030202.
- 263 [7] D. Khokhriakov, B. Karpiak, A.M. Hoque, S.P. Dash, Two-dimensional spintronic circuit architectures
264 on large scale graphene, *Carbon* 161 (2020) 892-899.
- 265 [8] N. Tombros, C. Jozsa, M. Popinciuc, H.T. Jonkman, B.J. van Wees, Electronic spin transport and spin
266 precession in single graphene layers at room temperature, *Nature* 448 (2007) 571-574.
- 267 [9] J. Sinova, S.O. Valenzuela, J. Wunderlich, C.H. Back, T. Jungwirth, Spin Hall effects, *Rev. Mod. Phys.* 87
268 (2015) 1213-1259.
- 269 [10] D. Huertas-Hernando, F. Guinea, A. Brataas, Spin-orbit-mediated spin relaxation in graphene, *Phys.*
270 *Rev. Lett.* 103 (2009) 146801.
- 271 [11] D. Pesin, A.H. MacDonald, Spintronics and pseudospintronics in graphene and topological insulators,
272 *Nat. Mater.* 11 (2012) 409-416.
- 273 [12] W. Han, K. Pi, K.M. McCreary, Y. Li, J.J.I. Wong, A.G. Swartz, et al., Tunneling spin injection into
274 single layer graphene, *Phys. Rev. Lett.* 105 (2010) 167202.
- 275 [13] A. Avsar, I.J. Vera-Marun, J.Y. Tan, G.K.W. Koon, K. Watanabe, T. Taniguchi, et al., Electronic spin
276 transport in dual-gated bilayer graphene, *NPG Asia Mater.* 8 (2016) e274.
- 277 [14] M.H.D. Guimaraes, A. Veligura, P.J. Zomer, T. Maassen, I.J. Vera-Marun, N. Tombros, et al., Spin
278 transport in high-quality suspended graphene devices, *Nano Lett.* 12 (2012) 3512-3517.
- 279 [15] P.J. Zomer, M.H.D. Guimaraes, N. Tombros, B.J. van Wees, Long-distance spin transport in high-
280 mobility graphene on hexagonal boron nitride, *Phys. Rev. B* 86 (2012) 161416(R).
- 281 [16] M.V. Kamalakar, A. Dankert, J. Bergsten, T. Ive, S.P. Dash, Enhanced tunnel spin injection into
282 graphene using chemical vapor deposited hexagonal boron nitride, *Sci Rep* 4 (2014) 6146.
- 283 [17] S. Singh, J. Katoch, J.S. Xu, C. Tan, T.C. Zhu, W. Amamou, et al., Nanosecond spin relaxation times in
284 single layer graphene spin valves with hexagonal boron nitride tunnel barriers, *Appl. Phys. Lett.* 109
285 (2016) 122411.
- 286 [18] M.V. Kamalakar, A. Dankert, P.J. Kelly, S.P. Dash, Inversion of Spin Signal and Spin Filtering in
287 Ferromagnet vertical bar Hexagonal Boron Nitride-Graphene van der Waals Heterostructures, *Sci Rep* 6
288 (2016) 21168.
- 289 [19] M. Gurram, S. Omar, B.J. van Wees, Electrical spin injection, transport, and detection in graphene-
290 hexagonal boron nitride van der Waals heterostructures: progress and perspectives, *2D Materials* 5
291 (2018) 032004.
- 292 [20] M. Drogeler, C. Franzen, F. Volmer, T. Pohlmann, L. Banszerus, M. Wolter, et al., Spin lifetimes
293 exceeding 12 ns in graphene nonlocal spin valve devices, *Nano Lett.* 16 (2016) 3533-3539.
- 294 [21] M. Drogeler, F. Volmer, M. Wolter, B. Terres, K. Watanabe, T. Taniguchi, et al., Nanosecond spin
295 lifetimes in single- and few-layer graphene-hBN heterostructures at room temperature, *Nano Lett.* 14
296 (2014) 6050-6055.

- [22] L. Li, I. Lee, D. Lim, S. Rath, M. Kang, T. Uemura, et al., Spin diffusion and non-local spin-valve effect in an exfoliated multilayer graphene with a Co electrode, *Nanotechnology* 27 (2016) 335201.
- [23] T. Maassen, F.K. Dejene, M.H.D. Guimaraes, C. Jozsa, B.J. van Wees, Comparison between charge and spin transport in few-layer graphene, *Phys. Rev. B* 83 (2011) 115410.
- [24] H.W. Du, X. Lin, Z.M. Xu, D.W. Chu, Electric double-layer transistors: a review of recent progress, *J. Mater. Sci.* 50 (2015) 5641-5673.
- [25] T. Fujimoto, K. Awaga, Electric-double-layer field-effect transistors with ionic liquids, *Phys. Chem. Chem. Phys.* 15 (2013) 8983-9006.
- [26] J.T. Ye, M.F. Craciun, M. Koshino, S. Russo, S. Inoue, H.T. Yuan, et al., Accessing the transport properties of graphene and its multilayers at high carrier density, *Proc. Natl. Acad. Sci. U. S. A.* 108 (2011) 13002-13006.
- [27] D. Braga, I.G. Lezama, H. Berger, A.F. Morpurgo, Quantitative determination of the band gap of WS₂ with ambipolar ionic liquid-gated transistors, *Nano Lett.* 12 (2012) 5218-5223.
- [28] X. He, N. Tang, L. Gao, J.X. Duan, Y.W. Zhang, F.C. Lu, et al., Formation of p-n-p junction with ionic liquid gate in graphene, *Appl. Phys. Lett.* 104 (2014) 143102.
- [29] X. He, Z.H. Zhang, C.H. Zhang, Y. Yang, M. Hu, W.K. Ge, et al., Exploration of exciton behavior in atomically thin WS₂ layers by ionic gating, *Appl. Phys. Lett.* 113 (2018) 013104.
- [30] Y. Saito, T. Nojima, Y. Iwasa, Gate-induced superconductivity in two-dimensional atomic crystals, *Supercond. Sci. Technol.* 29 (2016) 093001.
- [31] Y.J. Deng, Y.J. Yu, Y.C. Song, J.Z. Zhang, N.Z. Wang, Z.Y. Sun, et al., Gate-tunable room-temperature ferromagnetism in two-dimensional Fe₃GeTe₂, *Nature* 563 (2018) 94-99.
- [32] M.M. Perera, M.W. Lin, H.J. Chuang, B.P. Chamlagain, C.Y. Wang, X.B. Tan, et al., Improved carrier mobility in few-layer MoS₂ field-effect transistors with ionic-liquid gating, *ACS Nano* 7 (2013) 4449-4458.
- [33] F.J. Jedema, A.T. Filip, B.J. van Wees, Electrical spin injection and accumulation at room temperature in an all-metal mesoscopic spin valve, *Nature* 410 (2001) 345-348.
- [34] T. Maassen, J.J. van den Berg, N. Ijbema, F. Fromm, T. Seyller, R. Yakimova, et al., Long spin relaxation times in wafer scale epitaxial graphene on SiC(0001), *Nano Lett.* 12 (2012) 1498-1502.
- [35] T. Yamaguchi, R. Moriya, S. Masubuchi, K. Iguchi, T. Machida, Spin relaxation in weak localization regime in multilayer graphene spin valves, *Jpn. J. Appl. Phys.* 52 (2013) 040205.
- [36] F.J. Jedema, H.B. Heersche, A.T. Filip, J.J.A. Baselmans, B.J. van Wees, Electrical detection of spin precession in a metallic mesoscopic spin valve, *Nature* 416 (2002) 713-716.
- [37] M.H.D. Guimaraes, P.J. Zomer, J. Ingla-Aynes, J.C. Brant, N. Tombros, B.J. van Wees, Controlling spin relaxation in hexagonal BN-encapsulated graphene with a transverse electric field, *Phys. Rev. Lett.* 113 (2014) 086602.
- [38] T. Kim, H. Kirn, S.W. Kwon, Y. Kim, W.K. Park, D.H. Yoon, et al., Large-scale graphene micropatterns via self-assembly-mediated process for flexible device application, *Nano Lett.* 12 (2012) 743-748.
- [39] Y.J. Zhang, J.T. Ye, Y. Matsushashi, Y. Iwasa, Ambipolar MoS₂ thin flake transistors, *Nano Lett.* 12 (2012) 1136-1140.
- [40] E. Uesugi, H. Goto, R. Eguchi, A. Fujiwara, Y. Kubozono, Electric double-layer capacitance between an ionic liquid and few-layer graphene, *Sci Rep* 3 (2013) 1595.
- [41] R. Misra, M. McCarthy, A.F. Hebard, Electric field gating with ionic liquids, *Appl. Phys. Lett.* 90 (2007) 052905.
- [42] K. Pi, W. Han, K.M. McCreary, A.G. Swartz, Y. Li, R.K. Kawakami, Manipulation of spin transport in graphene by surface chemical doping, *Phys. Rev. Lett.* 104 (2010) 187201.
- [43] C. Jozsa, T. Maassen, M. Popinciuc, P.J. Zomer, A. Veligura, H.T. Jonkman, et al., Linear scaling between momentum and spin scattering in graphene, *Phys. Rev. B* 80 (2009) 241403(R).
- [44] A. Avsar, T.Y. Yang, S. Bae, J. Balakrishnan, F. Volmer, M. Jaiswal, et al., Toward wafer scale fabrication of graphene based spin valve devices, *Nano Lett.* 11 (2011) 2363-2368.

- [45] W. Han, R.K. Kawakami, Spin relaxation in single-layer and bilayer graphene, *Phys. Rev. Lett.* 107 (2011) 047207.
- [46] D. Khokhriakov, A.W. Cummings, K. Song, M. Vila, B. Karpiak, A. Dankert, et al., Tailoring emergent spin phenomena in Dirac material heterostructures, *Science Advances* 4 (2018) eaat9349.
- [47] B. Karpiak, A.W. Cummings, K. Zollner, M. Vila, D. Khokhriakov, A.M. Hoque, et al., Magnetic proximity in a van der Waals heterostructure of magnetic insulator and graphene, *2d Materials* 7 (2020) 015026.
- [48] A. Dankert, S.P. Dash, Electrical gate control of spin current in van der Waals heterostructures at room temperature, *Nat. Commun.* 8 (2017) 16093.
- [49] Z.Q. Fan, F. Xie, X.W. Jiang, Z.M. Wei, S.S. Li, Giant decreasing of spin current in a single molecular junction with twisted zigzag graphene nanoribbon electrodes, *Carbon* 110 (2016) 200-206.
- [50] Z.Q. Fan, W.Y. Sun, X.W. Jiang, Z.H. Zhang, X.Q. Deng, G.P. Tang, et al., Redox control of magnetic transport properties of a single anthraquinone molecule with different contacted geometries, *Carbon* 113 (2017) 18-25.
- [51] Z.Q. Fan, W.Y. Sun, Z.H. Zhang, X.Q. Deng, G.P. Tang, H.Q. Xie, Symmetry-dependent spin transport properties of a single phenalenyl or pyrene molecular device, *Carbon* 122 (2017) 687-693.
- [52] T. Enoki, M. Kiguchi, Magnetism of Nanographene-Based Microporous Carbon and Its Applications: Interplay of Edge Geometry and Chemistry Details in the Edge State, *Physical Review Applied* 9 (2018) 037001.
- [53] C. Song, J.H. Miao, F. Li, Y.N. Yan, B. Cui, F. Pan, Anomalous Hall effect in one monolayer cobalt with electrical manipulation, *J. Alloy. Compd.* 696 (2017) 315-320.