

Why Spintronics using 2D Materials ?

How the substrate controls spin dynamics?

Giant spin transport anisotropy in graphene induced by strong SOC Proximity effect

Weak antilocalization & Spin Hall Effect in Graphene/TMDC









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Spintronics and its industrial/Societal impact



Magnetic field sensors used to read data in hard disk drives, microelectromechanical systems (MEMS), minimally invasive surgery Automotive sensors for fuel handling system, Anti-skid system, speed control & navigation Magnetoresistive random-access memory (MRAM) Spin transfer Torque MRAM









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Spin-based information processing ?



Active devices based on **Spin manipulation**?



Need for spin information transport on long distance (room T) Spin injection and detection (ferromagnets/non magnetic materials)

Metals/semiconductors... short spin diffusion length (spin lifetime 0.1-1ns), 1% (or below) of MR signal

What makes graphene attractive



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- Ambipolar/tuneable transport
- Large mobilities (> 100k cm²/V.s at RT, 1M cm²/V.s at 4K)
- Low spin-orbit interaction
- Graphene properties can be tailored by proximity effects



M. Drögeler et al Nano Lett. 16 (6), 3533 (2016)

Magnetic oxides induce spin filtering/gap



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Yang, Hallal, Terrade, Waintal, Roche, Chshiev, PRL 110, 046603 (2013) Hallal et al. 2D materials 4, 025074 (2017)

Exchange splitting (G/YIG) = 40 meV

36 ICN29

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Potential of 2d Materials for STT-MRAM technologies







GRAPHENE FLAGSHIP

Perpendicular Magnetic Anisotropy in FM/Ox and FM/Graphene interfaces : Strongly enhanced PMA of Co realized by graphene coating

Layer and orbital resolved contributions unveil the PMA mechanisms

Superlattice structures to obtain Giant PMA



Yang, Chshiev et al, Nano Letters 16, 145 (2015)

Graphene-based Spintronic logic demonstrators

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W. Yan et al. Nature Comm. 7, 13372 (2016) SPIN-FET





Hua Wen et al. Phys. Rev. Applied **5**, 044003 (2016)

Experimental demonstration of XOR operation in graphene Magnetologic Gates at room temperature







EDITORIAL

2D Materials

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2D Mater. 2 (2015) 030202



Graphene spintronics: the European Flagship perspective

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"Unprecedented spin lifetimes in ultraclean graphene"??

Homogeneous Rashba + disorder (density of impurities)

Numerical calculation of the spin relaxation time by performing Monte Carlo simulations

Along any given classical trajectory $[{\bf r}(t), {\bf k}(t)]$ the spin dynamics is described by Bloch spin equation

$$\frac{d\mathbf{S}}{dt} = \Omega_R[\mathbf{r}(t)](\mathbf{n}[\mathbf{k}(t)] \wedge \mathbf{S})$$

Spin lifetime is calculated by averaging over random trajectories with different initial momenta , assuming

$$t \gg \tau_{tr}$$

$$S_{\alpha}(t) \sim e^{-t/\tau_{\alpha}}$$

 $\tau_{\alpha} \rightarrow \mu s - ms!!!$

Maximum at Dirac point Elliot Yaffet



C. Ertler, S. Konschuh, M. Gmitra and J. Fabian, Phys. Rev. B 80, 041405(R) (2009)

Experimental spin lifetime features

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Graphene on SiO2



Suspended Graphene



Epitaxial graphene on SiC Graphene on BN



charge mobility $\mu \sim 100 - 100.000 \text{cm}^2 \text{V}^{-1} \text{s}^{-1}$

Room temperature

$$\tau_s \sim 0.1 - 10 \text{ ns}$$

Avsar et al, Nano Lett. 11, 2363 (2011) Drögeler et al. Nano Lett. 14, 6050 (2014) Guimarães et al Phys Rev Lett 113, 086602 (2014)



Which relaxation mechanism at play ?





Drögeler et al. Nano Lett. 14, 6050 (2014) Guimarães et al PRL 113, 086602 (2014)

Cleaner samples for SiO₂ substrates lead to lower spin lifetime

"opposite trend" for hBN Substrates?

Origin of spin dephasing Spin-orbit coupling in graphene



"clean limit"

Intrinsic spin-orbit coupling Substrate effects (Rashba electric field) and electron-hole puddles

"dirty limit"

Etching transfer processes : contamination with ionic impurities (from metal etchants) or **metallic residues (from incomplete etching)**, + PMMA residues (transfer)

sp³ defects (σ–π hybridization) *Hydrogen ad-atoms-*



Transition- metal adatoms (Cu, Ni, Au,...)



Deformation fields (Strain, ripples, bubbles...)



Spin relaxation in supported clean graphene

-beyond semiclassical approximations-

Disorder: Electron-holes puddles Spatial (long range) charge density fluctuations



Electrons locally screen charged impurities trapped in the substrate

J. Martin et al, Nat. Phys. 4, 144 (2008)



 $\tau_s \sim \mu s - ms ???$

Uniform Rashba SOC-field **10** µeV (substrate effect/mirror symmetry breaking)

Spin-orbit interaction :

Tight-binding Modelling

$$\mathcal{H} = -\gamma_0 \sum_{\langle ij \rangle} c_i^+ c_j + \sum_{\langle i \rangle} V_i c_i^+ c_i + i V_R \sum_{\langle ij \rangle} c_i^+ \vec{z} \cdot (\vec{s} \times \vec{d}_{ij}) c_j$$

Screened Coulomb potential Long range (Gaussian) potential Rashba SOC

 $V_R \sim 20 \mu eV$



Spin dynamics of propagating wavepacket

$$\left\langle \Psi_{\perp}(0) \right\rangle = \left(\begin{array}{c} 1\\ 0 \end{array} \right) |\varphi_{RP} \rangle \qquad |\Psi(t)\rangle = e^{-i\hat{\mathcal{H}}t/\hbar} |\Psi(0)\rangle$$

 $s_i(t) = |\Psi_i^{\uparrow}(t)|^2 - |\Psi_i^{\downarrow}(t)|^2$ (time-dependent) Local spin density in real space





Graphene on SiO₂

electron-hole puddles drive the relaxation

 $\tau_p^{\mathrm{SiO}_2}/T_\Omega \ll 1$





-300

-200

-100

E(meV)

 $\tau_s(E) \approx 4T_\Omega \approx 4\frac{\pi\hbar}{\lambda_P}$

 $\tau_s \simeq 1 - 10 \text{ ns}$

(for $\lambda_R \to 5\mu eV$)

200

300

100

entangled dynamics between spin and pseudospin

D. Van Tuan et al, Nature Physics 10, 857 (2014) ,, Sci. Reports 6, 21046 (2016) A.W. Cummings and SR, PRL 116, 086602 (2016)

"Unique properties of Clean graphene"

pseudospin $|\Downarrow\rangle = \begin{pmatrix} 0\\ 1 \end{pmatrix}$

Long range potential Intravalley scattering (short momentum transfer)

Anomalous quantum transport

- Ballistic conductivity $\sigma \sim 4e^2/\pi h$
- Klein tunneling
- Diverging zero-energy Mean free path/mobility
- Weak antilocalization (quantum interferences)
- Anomalous vs conventional QHE
- Spin transport ?

Pseudospin-driven spin relaxation mechanism in graphene

Dinh Van Tuan^{1,2}, Frank Ortmann^{1,3,4}, David Soriano¹, Sergio O. Valenzuela^{1,5} and Stephan Roche^{1,5}*

Relaxation in the ultraclean limit....

Exponentially-decaying cosine, with frequency ω_0 and decay time $1/\alpha\eta$

$$s(t) = \mathcal{L}(E) \circ \cos(\omega(E)t) = e^{-\alpha\eta t} \cdot \cos(\omega_0 t)$$

A.W. Cummings and S. Roche , Phys. Rev. Lett. 116, 086602 (2016)

Crossover between "pure dephasing" and scattering-induced Dyakonov-Perel

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Hybrid devices of graphene and other 2D materials

(0)

2D Materials

Martin Gmitra and Jaroslav Fabian Phys. Rev. B **92**, 155403 (2015)

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Realistic Model of Graphene/TMDC

with interface disorder

DFT-TB model from

M. Gmitra , D. Kochan, P. Högl, & J.Fabian PRB **93**, 155104 (2016)

$$H_0 = -t \sum_{\langle i,j \rangle} \left(a_i^{\dagger} b_j + b_i^{\dagger} a_i \right) + \frac{\Delta}{2} \sum_i (a_i^{\dagger} a_i - b_i^{\dagger} b_i)$$

$$H_{\rm so} = \frac{2i}{3} \sum_{\langle i,j \rangle,\sigma} (\hat{\boldsymbol{s}} \times \boldsymbol{d}_{i,j})_{\boldsymbol{z},\sigma,\bar{\sigma}} \ \lambda_{\rm R} \, a_{i,\sigma}^{\dagger} \, b_{j,\bar{\sigma}} + h.c \quad \Gamma$$

$$+ \frac{1}{3} \sum_{\langle\langle i,j \rangle\rangle,\sigma} (\hat{s} \times \boldsymbol{D}_{i,j})_{z,\sigma,\bar{\sigma}} \left(\lambda_{\text{PIA}}^{(A')} a_{i,\sigma}^{\dagger} a_{j,\bar{\sigma}} + \lambda_{\text{PIA}}^{\dagger} b_{i,\sigma}^{\dagger} b_{j,\bar{\sigma}} \right) \\ + \frac{i}{\sqrt{2}} \sum_{\nu_{i,j}} (\hat{s}_{z})_{\sigma,\sigma} \left(\lambda_{I}^{(A)} a_{i,\sigma}^{\dagger} a_{j,\sigma} - \lambda_{I}^{(B)} b_{i,\sigma}^{\dagger} b_{j,\sigma} \right),$$

$$+ \frac{1}{3\sqrt{3}} \sum_{\langle\langle i,j\rangle\rangle,\sigma} \nu_{i,j}(s_z)_{\sigma,\sigma} \left(\lambda_I^{(-)} a_{i,\sigma}^{\dagger} a_{j,\sigma} - \lambda_I^{(-)} b_{i,\sigma}^{\dagger} b_{j,\sigma}\right)$$

Random distribution of n_p electron-hole puddles

S. Adam et al. PRB 84, 235421 (2011)

$$U_n(\mathbf{r}) = u_n \exp\left(-\frac{(\mathbf{r} - \mathbf{R}_n)^2}{2\xi_p^2}\right) \quad \xi_p = \sqrt{3}a \quad \text{Puddle range}$$

 $u_n \in [-U_{
m p}, U_{
m p}]$ **R**_n is the position of the center of the Gaussian pot.

Spin lifetimes in Graphene/TMDC (strong valley mixing)

density matrix $V_I(t) = \frac{1}{2}\hbar\vec{\omega}(t)\cdot\vec{s}_I(t)$ and $\vec{s}_I(t) = e^{iH_0t/\hbar}\vec{s}e^{-iH_0t/\hbar}$

Spin transport anisotropy in Graphene/TMDC (intravalley scattering only)

For higher quality interfaces Spin lifetimes (**out-of-plane**) Much smaller

Anistropy around ½ as in conventional Rashba Disordered systems

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ARTICLE

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Spin-orbit proximity effect in graphene

A. Avsar^{1,2}, J.Y. Tan^{1,2}, T. Taychatanapat^{1,2}, J. Balakrishnan^{1,2}, G.K.W. Koon^{1,2,3}, Y. Yeo^{1,2}, J. Lahiri^{1,2}, A. Carval A.S. Rodin⁴, E.C.T. O'Farrell^{1,2}, G. Eda^{1,2}, A.H. Castro Neto^{1,2} & B. Özyilmaz^{1,2,3}

"Graphene acquires spin–orbit coupling up to **17 meV**, three orders of magnitude higher than its intrinsic value, without modifying the structure of the graphene. **The proximity SOC leads to the spin Hall effect even at room temperature**, and opens the door to spin field effect transistors"

Weak antilocalization by proximity effect

Wang et al, PHYSICAL REVIEW X 6, 041020 (2016)

Dyakonov-Perel (1971); Hirsh (1999)

Skew Scattering

Side-Jump Scattering

Spin Hall Effect

Manipulation of spin by electrical means without the use of ferromagnetic materials

 k_{ii}

Intrinsic mechanism

Band Structure (e.g. Rashba)

 k_x

Spin Hall Kubo conductivity *in large scale disordered graphene models*

$$\begin{split} \sigma_{\rm sH} &= \frac{e\hbar}{\Omega} \sum_{m,n} \frac{f(E_m) - f(E_n)}{E_m - E_n} \frac{\mathcal{I}m[\langle m \mid J_x^z \mid n \rangle \langle n \mid v_y \mid m \rangle]}{E_m - E_n + i\eta}, \\ J_x^z &= \frac{\hbar}{4} \{\sigma_z, v_x\} \quad \text{is the spin current operator} \\ \text{real-space formalism} \quad \sigma_{\rm sH} &= \frac{e\hbar}{\Omega} \int du dv \frac{f(u) - f(v)}{(u - v)^2 + \eta^2} j(u, v), \\ j(u, v) &= \sum_{m,n} \mathcal{I}m[\langle m \mid J_x^z \mid n \rangle \langle n \mid v_y \mid m \rangle] \delta(u - E_m) \delta(v - E_n) \\ &= \sum_{m,n}^{M} (4\mu_{mn}g_m g_n T_m(\hat{u})T_n(\hat{v}))/((1 + \delta_{m,0})(1 + \delta_{n,0})\pi^2 \sqrt{(1 - \hat{u}^2)(1 - \hat{v}^2)}), \\ \mu_{mn} &= \frac{4}{\Delta E^2} \mathcal{I}m[Tr[J_x^z T_n(\hat{H})v_y T_m(\hat{H})]] \end{split}$$

The trace in μ_{mn} is computed by the average on a small number $R \ll N$ of random phase vectors $|\varphi\rangle$

$$\sigma_{xx} = \frac{2\hbar e^2}{\pi\Omega} \sum_{m,n=0}^{M} \mathcal{I}m[g_m(\epsilon + i\eta)]\mathcal{I}m[g_n(\epsilon + i\eta)]\mu_{mn}$$

dc-Kubo conductivity

Validation: homogeneous SOCs

Homogeneous Rashba SOC λ_R (no intrinsic SOC)
 Analytical result for the spin Hall conductivity
 A. Dyrdal, V. K. Dugaev, and J. Barnas, Phys. Rev. B. 80, 155444 (2009)

$$\sigma_{sH}(E) = \begin{cases} -\frac{e}{4\pi} \frac{\operatorname{sign}(E)E^2}{\left(E^2 - \lambda_R^2\right)} & \text{for } |E| \ge 2\lambda_R \\ -\frac{e}{4\pi} \frac{E(E + 2\operatorname{sign}(E)\lambda_R)}{2\lambda_R(E + \operatorname{sign}(E)\lambda_R)} & \text{for } |E| < 2\lambda_R \end{cases}$$

SHE induced by metallic ad-atoms onto graphene

ARTICLE

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Giant spin Hall effect in graphene grown by chemical vapour deposition

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Table 1 Graphene decorated with metallic adatoms.				
Adatom	Mobility (cm ² V $^{-1}$ s $^{-1}$)	λ _s (μm)	y	Δ (meV)
Cu-CVD	11,000	1.9	0.17	14.4
Cu-EPG	9,000	1.1	0.27	17.4
Au-EPG	15,000	2.0	0.15	18.0

CVD, chemical vapour deposition; EPG, exfoliated pristine graphene. The extracted values for Δ assume predominant intrinsic SOC (see main text).

intrinsic-like spin-orbit interaction (20meV), Elliot-Yafet and skew scattering

 $R_{n_{\rm L}}(\Omega)$

Cu-CVD

B.(T)

Giant Rashba Splitting in graphene hybridized with Au-adatoms

D. Marchenko et al, Nature Comm. 3 (1232), 2012

Au intercalation at the graphene–Ni interface creates a **giant spin–orbit splitting (60 meV)**

Spin Hall angles

 $\theta_{\rm sH} = \sigma_{\rm sH} / \sigma_{xx}$

 $heta_{
m sH}^{max} \sim 0.1 - 0.3$ (Zero-temperature)

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Impact of segregation varies with energy

For same cluster density and distribution *Intrinsic SOC gives rise to larger SHA than Rashba-SOC*

Far from dilute limit!!! SHA not expected to Be larger for much lower density...

Non-local transport & topological effects

PRL 117, 176602 (2016)

week ending 21 OCTOBER 2016

Spin Hall Effect and Origins of Nonlocal Resistance in Adatom-Decorated Graphene

D. Van Tuan,^{1,2} J. M. Marmolejo-Tejada,^{3,4} X. Waintal,⁵ B. K. Nikolić,^{3,*} S. O. Valenzuela,^{1,6} and S. Roche^{1,6,†} ¹Catalan Institute of Nanoscience and Nanotechnology (ICN2), CSIC and The Barcelona Institute of Science and Technology, Campus UAB, Bellaterra, 08193 Barcelona, Spain ²Department of Electrical and Computer Engineering, University of Rochester, Rochester, New York 14627, USA ³Department of Physics and Astronomy, University of Delaware, Newark, Delaware 19716-2570, USA ⁴School of Electrical and Electronics Engineering, Universidad del Valle, Cali AA 25360, Colombia ⁵Univ. Grenoble Alpes, INAC-PHELIQS, F-38000 Grenoble, France and CEA, INAC-PHELIQS, F-38000 Grenoble, France ⁶ICREA—Institució Catalana de Recerca i Estudis Avançats, 08010 Barcelona, Spain (Received 19 February 2016; published 20 October 2016)

Multiple Quantum Phases in Graphene with Enhanced Spin-Orbit Coupling: From the Quantum Spin Hall Regime to the Spin Hall Effect and a Robust Metallic State

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THANK YOU FOR YOUR ATTENTION

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Related Publications

- D. Van Tuan, F. Ortmann, D. Soriano, S.O. Valenzuela, S. Roche, *Pseudospin-driven spin relaxation mechanism in graphene*', Nature Physics, 10, 857-863 (2014)
- A. Cresti, D. Van Tuan, D. Soriano, A. W. Cummings, S. Roche 2014, 'Multiple Quantum Phases in Graphene with Enhanced Spin-Orbit Coupling : from Quantum Spin Hall Regime to Spin Hall Effect and Robust Metallic State', Physical Review Letters, 113, 246603 (2014).
- A.W. Cummings and S. Roche, 'Effects of Dephasing on Spin Lifetime in Ballistic Spin-Orbit Materials',

Physical Review Letters 116, 086602 (2016)

• D. Van Tuan and S. Roche, 'Spin manipulation in graphene by chemically induced pseudospin polarization',

Physical Review Letters 116, 106601 (2016)

- D. Van Tuan, J. M. Marmolejo-Tejada, X. Waintal, B.K. Nikolic, S. Roche, Physical Review Letters 117176602 (2016)
- D. Van Tuan, F. Ortmann, A.W. Cummings, D. Soriano, S. Roche, *Spin dynamics and relaxation in graphene dictated by el-h puddle* <u>Scientific Reports 6, 21046 (2016)</u>
- A. W. Cummings, J.H. García and S. Roche, ' Giant Spin Lifetime Anisotropy in Graphene Induced by Proximity Effects Physical Review Letters (submitted)