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# SP 7 Magnetism and spintronics in organic materials and carbon based systems

## SP7 - Parallel Session 1

### SP7 - Parallel Session 1

#### SP7.1.02

### Theoretical modeling of spin diffusion length, spin-filter and spin Hall effects in organics

M. R. Mahani, U. Chopra, E. R. McNellis, J. Sinova

**Text** The generation, detection and efficient transfer of spin polarized electrons across a system, are some of the major concerns in spintronic technologies. Theoretical understanding of these complex processes, particularly for the relatively new field of organic spintronics, is of great importance. Here, as part of a multiscale modeling framework, we describe spin diffusion length, spin-filter and spin Hall effects in organics. The ability to use a single molecule or a molecular layer as a spin filter, chiral induced spin selectivity, provides an efficient way to create spin-polarized electrons, instead of common use of magnetic materials in traditional inorganic spin filters. On the basis of an effective model Hamiltonian and the Landauer-Buttiker formalism, we elucidate the underlying physical mechanisms for high spin selectivity observed in chiral molecules.

In order to calculate the SHE in organic materials, we numerically solve the master equations for electrical and spin hall conductivities in the hopping regime. We show the density of states at the Fermi level, the spin-orbit coupling and three-site hopping are salient features to control the SHE in these materials.

Finally, we discuss the long-distance spin transport in highly-doped conjugated polymers. We calculate the long spin diffusion length in poly(2,5-bis(3-alkylthiophen-2-yl)thieno[3,2-b]thiophene), PBTTT, in the limit of high carriers concentration, based on parameters calculated from first-principles.



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### SP7 - Parallel Session 1

#### SP7.1.03

#### Tailoring the spin structure of surface alloys by adsorption of organic molecules

B. Stadtmüller, J. Seidel, N. Haag, L. Grad, S. Mousavion, L. Lyu, D. Jungkenn, G. van Straaten, M. Franke, H.-J. Elmers, C. Kumpf, M. Cinchetti, M. Aeschlimann

**Text** The performance of spintronic devices crucially depends on our ability to design and control the spin-dependent electronic states of device relevant materials such as ferromagnets or materials with large spin-orbit coupling. Model systems for the latter class of materials are topological insulators or Rashba systems which are characterized by spin-textured surface states. The surface character of these transport relevant states opens a unique possibility for manipulation of the surface spin-structure by the adsorption of adsorbates.

Here, we will demonstrate our novel approach to control the spin-dependent electronic properties of Rashba-type surface alloys by the adsorption of organic adsorbates [1]. For the surface alloy  $\text{Pb}_1\text{Ag}_2/\text{Ag}(111)$ , we will show that the formation of  $\sigma$ -like bonds between functional groups of organic molecules and the Pb alloy atoms results in a modification of the spin-dependent surface band structure and in a vertical relaxation of the Pb atoms. In contrast, no change of the properties of the surface alloy was found for molecules without functional groups. Similar modifications of the spin-dependent band structure were observed for the ferromagnetic  $\text{Dy}_1\text{Ag}_2/\text{Ag}(111)$  surface alloy upon adsorption of organic molecules.

Along these lines, our results demonstrate the unique potential of organic molecules to modify the spin-structure of metallic surface alloys by the formation of tailor-made metal-organic bonds.



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## SP7 - Parallel Session 1

### SP7.1.04

#### Tuning spin injection by molecular design

A. Wittmann, G. Schweicher, D. Venkateshvaran, K. Broch, C. Jellett, A. Marks, M. Little, I. McCulloch, E. McNellis, J. Sinova, J. Wunderlich, H. Siringhaus

**Text** Organic semiconductors have recently been found to have a comparably large spin diffusion time and length. This makes them ideal candidates for spintronic devices. However, spin injection and transport properties in organic semiconductors have yet to be fully understood.

The efficiency of spin injection at ferromagnetic resonance (FMR) from a ferromagnetic material (FM) into an adjacent non-magnetic material (NM) is given by the spin mixing conductance  $g_{\text{eff}}$ . It can be quantified by measuring the linewidth broadening of the FMR absorption of the FM due to an increase in Gilbert damping caused by spin injection into the NM. Here, we use this technique to systematically study spin injection from a metallic ferromagnet, Ni<sub>80</sub>Fe<sub>20</sub>, into organic small molecules and conjugated polymers. In particular, we investigate the influence of the molecular structure at the interface on the spin injection efficiency. One of the best performing small molecule organic semiconductors to date is dinaphtho[2,3-b:2',3'-f]thieno[3,2-b]thiophene (DNTT). Comparing DNTT with C8-DNTT, we observe a significant suppression of the linewidth broadening and a decrease in spin diffusion length in C8-DNTT, which can be attributed to a larger separation of the core of the molecule from the interface due to the addition of alkyl side chains. This demonstrates that both spin mixing conductance as well as the spin diffusion length can be tuned through molecular design.



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#### SP7.1.05

#### **What is true and what is false: The magnitude of the inverse spin-Hall effect in PEDOT:PSS**

M. M. Qaid, G. Schmidt

**Text** Spin current injected from a ferromagnet into adjacent conductor can be detected as a charge current if the conductor exhibits the inverse spin-Hall effect (ISHE). Using a typical measurement geometry this mechanism can be prone to artifacts[1], especially when the ferromagnet has a large magnon propagation length and the conductor has a large Seebeck coefficient, long spin diffusion length (SDL) and weak spin orbit coupling (SOC) as is the case for  $\pi$ -conjugated polymers. In this work, we report systematic investigations for the ISHE detection in the conducting polymer PEDOT:PSS using a sequence of control experiments with different layer stacks and measurement geometries. With these configurations, we are able to quantify and exclude artifacts generating voltages normally indistinguishable from the ISHE that either result from thermovoltages caused by non-reciprocal magnetostatic surface spin waves (MSSW)[2] or from the spin pumping into the contacts. As a result, we show that in PEDOT:PSS a measurable ISHE exists. Nevertheless, its true value is several orders of magnitude lower than that obtained in a standard measurement geometry. In addition, a sample geometry is presented which avoids the above mentioned artifacts and gives direct access to the ISHE.

[1] P. Wang, et al. Phys. Rev. Lett., 120, 047201 (2018)

[2] T. An, et al. Nature Mater. 12, 549–553 (2013)



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# SP 7 Magnetism and spintronics in organic materials and carbon based systems

## SP7 - Parallel Session 1

### SP7.1.06

#### Spin precession and spin Hall effect in monolayer graphene/Pt nanostructures

W. Saverio Torres, J. F. Sierra, L. A. Benitez, F. Bonell, M. V. Costache, S. O. Valenzuela

**Text** Spin Hall effects (SHE) have surged as promising phenomena for spin logics operations without ferromagnets [1]. So far, the exploration of the SHE using nonlocal measurements has been performed in metals using Pt as the large SOC material. However, the magnitude of the detected electric signals at room temperature in metallic systems has been so far underwhelming. Here, we demonstrate a two-order of magnitude enhancement of the signal in monolayer graphene/Pt devices when compared to their fully metallic counterparts [2]. The enhancement stems in part from efficient spin injection and the large resistivity of graphene but we also observe 100% spin absorption in Pt and an unusually large effective spin Hall angle of up to 0.15. The large spin-to-charge conversion allows us to characterise spin precession in graphene under the presence of a magnetic field. Furthermore, by developing an analytical model based on the 1D diffusive spin-transport, we demonstrate that the effective spin-relaxation time in graphene can be accurately determined using the (inverse) spin Hall effect as a means of detection. This is a necessary step to gather full understanding of the consequences of spin absorption in spin Hall devices, which is known to suppress effective spin lifetimes in both metallic and graphene systems.

[1]J. Sinova, S.O. Valenzuela, J. Wunderlinch, C. H. Back Rev. Mod. Phys. 87, 1213 (2015)

[2]W. Saverio Torres et al. 2D. Mater. 4 041008 (2017)



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### SP 7 Magnetism and spintronics in organic materials and carbon based systems

#### SP7 - Parallel Session 1

#### SP7.1.07

#### Magnetocaloric effect in thin films of Prussian blue analogues

M. Fitta, W. Sas, T. Korzeniak, M. Bałanda

**Text** In the last few years, there has been a significant interest in the study of magnetocaloric effect (MCE) motivated by a possibility of applying this effect in magnetic refrigeration. Because the MCE material must be in contact with the system to be cooled, therefore, the 2D geometry of a thin film is the most appropriate for the heat flow because of the large surface to volume ratio.

Here we present the first example of study on MCE in thin films of Prussian blue analogues (PBAs). The thin film of nickel hexacyanochromate with thickness of 450 nm and roughness of 68 nm was obtained by "layer

-by-layer" deposition [1].

ferromagnetically ordered state at  $T_c = 66$  K. The different magnetic response measured for fields lower than 1 T for films oriented parallel and perpendicular to the direction of magnetic field has been explained by the geometry of a demagnetization factor. Besides the analysis of the  $\Delta S$  magnetic entropy temperature dependence and its variation upon the rotating the sample from in-plane to out-of-plane orientation, we have studied the scaling behavior of MCE for the sample in both positions.

MCE was also measured in the thin films of PBAs synthesized by electrochemical deposition: iron hexacyanochromate which is ferromagnetic with  $T_c = 22$  K and ferrimagnetic chromium hexacyanochromate with  $T_c = 225$  K.

[1] M. Fitta, M. Szuwarzyński, P. Czaja, G. Lewińska, M. Bałanda, Eur. J. Inorg. Chem. 2017, 4817–4822



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## SP 7 Magnetism and spintronics in organic materials and carbon based systems

### SP7 - Parallel Session 2

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#### SP7.2.02

### **Gluing lanthanides spins with unpaired electrons inside fullerenes: Towards Single Molecule Magnets (SMMs) with high blocking temperatures**

G. Velkos, F. Liu, L. Spree, D. Krylov, S. Avdoshenko, A. Popov

**Text** Coupling magnetic atoms into larger clusters is a viable strategy towards molecular magnets. Fullerenes provide an opportunity to stabilize in their interior otherwise non-existent lanthanide clusters. Of particular interest are the air-stable endohedral dimetallofullerenes (di-EMFs) featuring the single-electron covalent Ln–Ln bond. The local magnetic moments of Ln ions exhibit giant exchange coupling with the spin of the delocalized unpaired electron residing on the Ln–Ln bonding orbital, leading to the [Ln–e–Ln] system with a single giant-spin. In case of Dy<sub>2</sub>@C<sub>80</sub> and Tb<sub>2</sub>@C<sub>80</sub>, each Ln ion has an easy-axis magnetic anisotropy with the parallel alignment of the axes of two Ln centers. As a result, these molecules exhibit very high blocking temperatures of magnetization (29 K for Tb, 22 for Dy) and the effective relaxation barrier near 600-800 K assigned to the exchange excitation. Below 29 K, Tb<sub>2</sub>@C<sub>80</sub> exhibits giant hysteresis with coercivity up to 8 T. In case of Ho<sub>2</sub>@C<sub>80</sub>, Ho ions also have an easy-axis magnetic anisotropy, but the axes of individual centers are tilted from the Ho–Ho direction, leading to a much faster relaxation. Finally, Gd<sub>2</sub>@C<sub>80</sub> and azafullerene Gd<sub>2</sub>@C<sub>79</sub>N featuring a single-electron Gd–Gd bond at low temperature behave as isotropic S=15/2 spin system. The effective exchange coupling constant between the Gd spins and the spin of the unpaired electron of the Gd–Gd bond is determined to be 170±10 cm<sup>-1</sup>.



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## SP 7 Magnetism and spintronics in organic materials and carbon based systems

### SP7 - Parallel Session 2

#### SP7.2.03

#### Enhancing the magnetic hybridization of C60 on Fe(001) by two-dimensional oxides

A. Brambilla, A. Picone, D. Giannotti, A. Calloni, G. Bussetti, G. Fratesi, S. Achilli, M. I. Trioni, G. Vinai, P. Torelli, G. Panaccione, M. Finazzi, L. Duò, F. Ciccacci

**Text** The formation of hybridized interface states (HIS) when organic layers are coupled to ferromagnetic materials (spinterfaces) leads to the onset of a magnetic moment in the molecules. This opens the way to a possible exploitation of spinterfaces in spintronics systems, such as tunneling junctions and spin-transport channels, on account of the long spin lifetimes in organic layers. A detailed control over the interface characteristics is therefore of paramount importance to define and, eventually, to shape physical systems with improved capabilities [1]. A novel approach to tailor the formation of HIS consists in the insertion of ultra-thin oxides at spinterfaces. We have been working on prototypical spinterfaces between C60 fullerene and Fe(001) and thoroughly investigated the morphology and electronic properties of C60 on differently prepared Fe(001) substrates. In particular, we have compared the reference Fe(001) case to variants including monolayer FeO and Cr4O5 oxides at the interface. We have demonstrated, by means of X-ray Magnetic Circular Dichroism, that the presence of 2D oxides enhances the magnetic hybridization [2,3]. The efficacy of the tailoring approach is further confirmed by ab-initio simulations that allow us to link the observed spectra with the electronic structure of the substrate [3].

[1] M. Cinchetti et al., *Nature Mater.* 16, 507 (2017)

[2] A. Picone et al., *ACS Appl. Mater. Interfaces* 8, 26418 (2016).

[3] A. Brambilla et al., *Nano Lett.* 17, 7440 (2017).





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# SP 7 Magnetism and spintronics in organic materials and carbon based systems

## SP7 - Parallel Session 2

### SP7.2.04

#### Spin dynamics in high mobility conjugated polymers

S. Schott, R. Carey, A. Melnyk, M. Ruggiero, R. Di Pietro, E. McNellis, C. McNeill, D. Andrienko, J. Sinova, H. Siringhaus

**Text** Polymeric semiconductors and conductors are enabling flexible, large-area optoelectronic devices, such as organic light-emitting diodes, transistors, and solar cells. Due to their exceptionally long spin lifetimes, these carbon-based materials could also have an impact on spintronics<sup>1</sup>.

Weak spin orbit- and hyperfine interactions lie at the origin of their long spin lifetimes<sup>2</sup> but the coupling mechanism of a spin to its environment remains elusive. For instance spin-phonon coupling, the charges' hopping motion through a disordered local field environment<sup>3</sup>, and Elliott-Yafet type spin flips at spatial scattering events<sup>4</sup> can all potentially contribute to spin relaxation.

In this work, we present spin resonance measurements on field induced charges in a transistor architecture down to liquid helium temperatures. We compare coherence and spin lattice relaxation times for a series of state-of-the art donor-acceptor polymers with excellent charge transport properties and demonstrate that spin dynamics at low temperatures are governed by the charge's hopping motion. We identify distinct temperature regimes where competing mechanisms become dominant and show a subtle dependence of spin lifetimes on changes in thin film morphology, corroborated by X-ray scattering and THz vibrational spectroscopy .

<sup>1</sup> G. Szulczewski, et al., Nat. Mater. 8, 693 (2009).

<sup>2</sup> S. Schott, et al. Nat. Comm. 8, 15200 (2017).

<sup>3</sup> H. Matsui, et al. Phys. Rev. B 85, 035308 (2012).

<sup>4</sup> Z. G. Yu, Phys. Rev. B 85, 115201 (2012).



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## SP 7 Magnetism and spintronics in organic materials and carbon based systems

### SP7 - Parallel Session 2

#### SP7.2.05

#### Magnetic subunits within a single molecule-surface hybrid

R. Friedrich, V. Heß, V. Caciuc, F. Matthes, D. E. Bürgler, N. Atodiresei, C. M. Schneider, S. Blügel

**Text** Single molecule-surface hybrid systems are a promising platform for molecular spintronics due to their adjustable magnetic properties [1]. The adsorption of organic molecules on a ferromagnetic substrate allows to engineer the exchange interactions and hysteretic behavior of the magnetic surface [2]. In this contribution we demonstrate the creation of multiple intramolecular subunits within a single molecule-surface hybrid system employing *ab initio* density functional theory and spin-polarized scanning tunneling microscopy [3]. Upon deposition of a polycyclic aromatic molecule onto a ferromagnetic surface an asymmetric (chiral) adsorption geometry is obtained. This gives rise to the formation of different structural, electronic and magnetic properties of each aromatic ring hybridizing with the surface. More specifically, each molecular lobe shows different shape and intensity in the STM topography images. Moreover it is found that the spin polarization varies on an intramolecular length scale within the single molecule-surface hybrid system. Finally, we demonstrate the creation of magnetic subunits, which are distinguished from one another by their exchange coupling strength [3]. These results might hence be regarded as an important step towards intramolecular based spin logic devices.

[1] N. Atodiresei *et al.*, Phys. Rev. Lett. **105**, 066601 (2010).

[2] R. Friedrich *et al.*, Phys. Rev. B **91**, 115432 (2015).

[3] V. Heß *et al.*, New J. Phys. **19**, 053016 (2017).



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## SP 7 Magnetism and spintronics in organic materials and carbon based systems

### SP7 - Parallel Session 2

#### SP7.2.06

#### **Spin-flip spectroscopy along a molecular spin chain coupled to the ferromagnetic electrodes of a sub-micronic junction**

K. Katcko, E. Urbain, L. Kandpal, F. Schleicher, B. Chowrira, B. Leconte, N. Beyer, A. Boulard, J. Arabski, E. Beaurepaire, W. Weber, S. Boukari, M. Bowen

**Text** Low-temperature local-probe experiments demonstrate how to electrically manipulate molecular spin chains (i.e. spin-flip spectroscopy). To manipulate this quantum object at higher temperatures within a solid-state device effectively requires a sub-micronic device, rather than the typically studied shadow mask junctions with  $\sim 10$  micron dimensions. Thus far, this has only been achieved using a nano-indentation technique, but this one-of-a-kind approach inherently generates structural/chemical disorder for at least one interface.

To address this long-standing technological bottleneck, we have adapted a nanosphere processing technique so as to study 500nm-wide Fe/CoPc(20nm)/Co organic junctions. We observe in-plane magnetoresistance only when  $T < 100\text{K}$ , and once a conductance jump has taken place for  $V \geq 50\text{mV}$ . For  $60 < T(\text{K}) < 100$ , we observe a symmetric  $R(H)$  curve with one reversal field  $H_f > 1\text{T}$ . We conclude from minor loop analysis that one of the FM/CoPc interfaces is phenomenologically experiencing exchange blocking. Below  $T = 60\text{K}$ , we witness very asymmetric, reversible  $R(H)$  curves which suggests that the second interface is now magnetically coupled to the first through the spin chain and a sharp MR jump at  $H_f = 1.4\text{T}$ . An in-plane angular dependence attributes this reversal to a spin-flop event. Our results address the enduring controversy between an exchange bias and interfacial spin frustration description of magnetism at the FM/molecule interface.



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# SP 7 Magnetism and spintronics in organic materials and carbon based systems

## SP7 - Parallel Session 2

### SP7.2.07

#### Transport regimes of organic spintronic devices and applications in neuromorphic computing

A. Riminucci, R. Legenstein, Z.-G. Yu, R. Cecchini, P. Graziosi, M. Prezioso, I. Bergenti, V. A. Dediu

**Text** Despite the extensive research efforts devoted to the understanding of spin transport in organic semiconductors, key results, such as the Hanle effect, are still missing. Many of the results in this field were obtained with the organic semiconductor (OS) tris(8-hydroxyquinoline)aluminum (Alq3). We have identified two distinct sets of devices, one with low resistance that shows spin valve magnetoresistance, and one with high resistance, with no magnetoresistance.

To understand the low resistance devices, that show magnetoresistance and resistive multistability we have expanded on the impurity band model. In our devices we envisage oxygen to be the impurity and bistability is attributed its migration within the organic semiconductor/AIOx bilayer.

These devices can be used as a synapse for neuromorphic computing and can be used in several types of neural networks. The presence of magnetoresistance adds a unique tool to effect parallel, selective changes on the weight of synapses.

We tested the effectiveness on reward-based learning in an actor-critic framework. The model consists of two networks. The actor network learns a policy, that is, a suitable action (output)  $y(x)$  for the current input  $x$ . The critic network learns the expected  $R(x)$  reward for this input. The critic network is used to improve learning of the actor network. In both cases, the nonlinear update has a significant advantage in terms of the speed at which the performance goal of a mean reward of 0.95 is reached.