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Spin Coherence at the Nanoscale: Polymer Surfaces and Interfaces

Final Report
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DESCRIPTION OF SELECTED RECENT ACCOMPLISHMENTS:

Breakthrough results were achieved during the reporting period in the areas of organic spintronics. **(A)** For the first time the giant magnetic resistance (GMR) was observed in spin valve with an organic spacer. Thus we demonstrated the ability of organic semiconductors to transport spin in GMR devices using rubrene as a prototype for organic semiconductors. **(B)** We discovered the electrical bistability and spin valve effect in a ferromagnet /organic semiconductor/ ferromagnet heterojunction. The mechanism of switching between conducting phases and its potential applications were suggested. **(C)** The ability of $V(TCNE)_x$ to inject spin into organic semiconductors such as rubrene was demonstrated for the first time. The mechanisms of spin injection and transport from and into organic magnets as well through organic semiconductors were elucidated. **(D)** In collaboration with the group of OSU Prof. Johnston-Halperin we reported the successful extraction of spin polarized current from a thin film of the organic-based room temperature ferrimagnetic semiconductor $V[TCNE]_x$ and its subsequent injection into a GaAs/AlGaAs light-emitting diode (LED).

Thus all basic steps for fabrication of room temperature, light weight, flexible all organic spintronic devices were successfully performed. **(E)** A new synthesis/processing route for preparation of $V(TCNE)_x$ enabling control of interface and film thicknesses at the nanoscale was developed at OSU. Preliminary results show these films are higher quality and what is extremely important they are substantially more air stable than earlier prepared $V(TCNE)_x$.

In sum the breakthrough results we achieved in the past two years form the basis of a promising new technology, Multifunctional Flexible Organic-based Spintronics (MFOBS). MFOBS technology enables us fabrication of full function flexible spintronic devices that operate at room temperature.

A. Giant magnetoresistance in ferromagnet/organic semiconductor/ferromagnet heterojunctions (see Publications 7, 12, 17, 21 in Section 6 - note publication 7 is an “editor’s select pick in Physical Review B)

We reported the spin injection and transport in ferromagnet/organic semiconductor/ferromagnet (FM/OSC/FM) heterojunctions using rubrene ($C_{42}H_{28}$) as an organic semiconductor spacer. For completeness of our study, both tunneling magnetoresistance (TMR) and giant magnetoresistance (GMR) were studied by varying the thickness of the rubrene layer (5–30 nm). A thorough study of the device characteristics reveals spin-polarized carrier injection into and subsequent transport through the OSC layer. When the thickness of the rubrene layer is beyond the tunneling limit (> 5 nm), the device current is limited by carrier injection and bulk transport. We describe the carrier injection with phonon-assisted field emission mechanism. The behavior of GMR in response to bias field and temperature shows significant differences from that of TMR. Fig. 1 is a typical curve showing operation of a FM/OSC/FM heterojunction using rubrene ($C_{42}H_{28}$) as an organic semiconductor spacer.

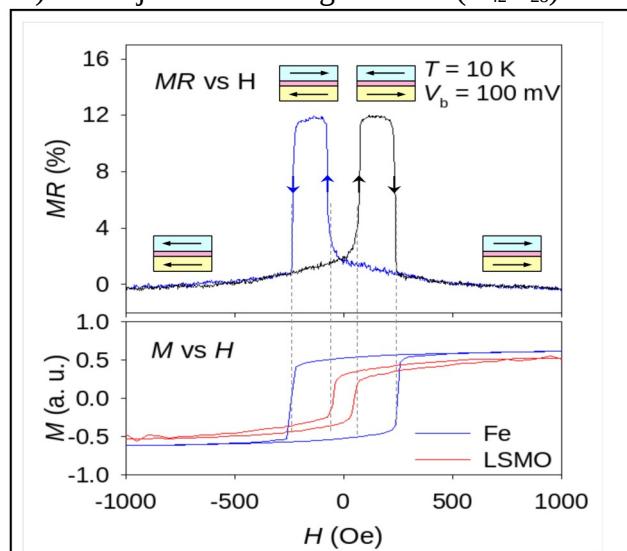


Fig. 1. (bottom) M vs H at 10 K for Fe (30 nm) and LSMO (50 nm) on LSAT(001) substrate. (top) MR curves of 5 nm rubrene device at $T=10$, data at 50, 100, and 150 K nearly the same though reduced in absolute MR and LSMO (50 nm) magnetic layers recorded by SQUID. See Section IV of Publication 39, for more details.

Fig. 3. I - V curves for 2, 20, 30 and 50 nm rubrene devices at $T = 10$ K.

To elucidate of mechanism, of spin transport through the organic spacer we comprehensively studied the IV characteristics. As the thickness of rubrene layer is increased, the device current strongly relies on carrier injection resulting in strong temperature and voltage dependent device resistance [4].

Fig. 2 displays T dependent device resistivity for the 5, 20, 50 nm rubrene devices and resistivity of the 50 nm LSMO film at bias 0.1 V. Strong nonlinear I - V curves are shown in Fig. 3 for 5, 20, 30, and 50 nm rubrene devices.

The strong T and V_b dependent current for devices with space thickness less than 100 nm originates from mainly activation of charge carriers at the metal/OSC interface

Rubrene 50 nm

T -dependence of resistance of 5, 20, 50 nm rubrene devices and 50 nm LSMO film.

Fig. 2.

barriers rather than bulk resistance. To explain the voltage and temperature dependence of current in our organic-based spin valve devices, we used the theoretical model developed for conventional semiconductors. This model describes carrier injection at the metal/semiconductor interface by combining multiphonon activation and field emission of electrons from defect states at the interfaces to the conduction band of semiconductors. Agreement between the experimental dependencies and the model predictions is excellent and the fitting parameters such as the activation energy, phonon frequency and electron-phonon coupling have reasonable values.

B. Electrical bistability and spin valve effect in a ferromagnet/organic semiconductor/ ferromagnet heterojunction (for further information see publication 19 in Section VI)

Fig. 4. I–V curves of the hybrid organic bistable device of structure LSMO(50 nm)/LAO/rubrene (50 nm)/Fe (30 nm) at 10 K. Initially, the device is in the OFF state. At about 4.5 V, the current increases sharply, indicating the transition of the device from the OFF to the ON state.

We reported discovery and study of the electrical bistability and bias-controlled spin valve effect in an organic spintronics device using rubrene ($C_{42}H_{28}$) as an organic semiconductor channel. The half-metallic $La_{0.7}Sr_{0.3}MnO_3$ (LSMO) and Fe are used as the two ferromagnetic electrodes. The device displays reproducible switching between a low-impedance (ON) state and an high impedance (OFF) state by applying different polarities of high biases. In the ON state, the device shows a spin valve effect with magnetoresistance values up to 3.75%.

The observed spin valve effect disappears when the device recovers to the initial OFF state, Fig. 6. This phenomenon opens the opportunity for multifunctional organic-based spintronic devices using both magnetic and electric fields to manipulate information.

C. Spin injection/detection using an organic-based magnetic semiconductor (Organic based Spin Valve $V(TCNE)_x(500$ nm)/Rubrene(5 nm)/LAO(1.2 nm)/LSMO(80 nm) for further information see publication 18 and 21 in Section VI)

The new paradigm of electronics, ‘spintronics’, promises to extend the functionality of information storage and processing in conventional electronics. The principal spintronics device, the ‘spin valve’, consists of two magnetic layers decoupled by a spin-transporting spacer, which allows parallel (on) and antiparallel (off) alignment of the magnetizations (spins) of the two magnetic layers. The device resistance then depends on the spin alignment controlled by the external magnetic field.

Fig. 5. The magnetoresistance curves of a $V(TCNE)_x(500$ nm)/Rubrene(5 nm)/LAO(1:2 nm)/LSMO(80 nm) junction and magnetization loops for LSMO (black) and $V(TCNE)_x$ (green).

In pursuit of semiconductor spintronics there have been intensive efforts devoted to develop room temperature magnetic semiconductors and also to incorporate both inorganic semiconductors and carbon-based materials as the spin-transporting channels. Molecule/organic-based magnets, which allow chemical tuning of electronic and magnetic properties, are a promising new class of magnetic materials for future spintronic applications. We reported the first realization of an organic-based magnet as an electron spin polarizer in the standard spintronics device geometry (Publication 44 in Section VI, *Nature Materials*).

Fig. 6. MR of organic based spin valve as a function of temperature.

A thin non-magnetic organic semiconductor layer and an epitaxial ferromagnetic oxide film were employed to form a hybrid magnetic tunnel junction. The results demonstrate the spin-polarizing nature of the organic-based magnetic semiconductor, $V(TCNE)_x$ ($x \sim 2; T_c \sim 400$ K), and its function as a spin injector/detector in hybrid magnetic multilayer devices, Fig. 5.

The MR as a function of temperature (Fig. 6) shows a maximum at ~ 100 K. The decrease of MR, as temperature increases above 100 K, is attributed to the decrease of surface spin polarization of the LSMO film. This suggests that MR at higher temperature can be achieved in magnetic tunnel junctions with organic-based magnet $V(TCNE)_x$ in tandem with other ferromagnetic metals.

D. Electrical spin injection from an organic-based magnet in a hybrid organic/inorganic heterostructure. (Publication 24 in Section VI)

Fig. 7. EL polarization (red line) for GaAs/AlGaAs light-emitting diode (LED) incorporating injection of spin polarized electrons using $V(TCNE)_x$ as the spin injector at $T = 60$ K, $I = 0.5$ mA and $V = +18.5$ V. HH polarization follows out of plane magnetization curve (green line, measured using SQUID magnetometry) of $V[TCNE]_{x \sim 2}$ and reaches saturation at ± 200 Oe.

In collaboration with the group of OSU Prof. Johnston- Halperin we reported the successful extraction of spin polarized current from a thin film of the organic-based room temperature ferrimagnetic semiconductor $V[TCNE]_x$ and its subsequent injection into a GaAs/AlGaAs light-emitting diode (LED). The orientation of this spin current is determined by polarization analysis of the electroluminescence from the LED and is found to be parallel to the magnetization of the $V[TCNE]_{x \sim 2}$ layer, in agreement with theoretical predictions. Circular polarization from EL persists up to 140 K, the highest temperature at which EL from the quantum well can be clearly resolved. This successful demonstration of spin injection in a hybrid organic/inorganic structure opens the door to a new class of active, hybrid spintronic devices with multifunctional behavior defined by the optical, electronic and chemical sensitivity of the organic layer.

Our success demonstrates the possibilities for “all-organic spintronics”, as well as “hybrid spintronics”, and “air stable, light-weight, flexible spintronics”. We have

initiated studies of these extensions to organic spintronics, with promising preliminary results.

E. New deposition technique of organic based magnets $V(TCNE)_x$ with nanoscale control (US patent is pending)

A finely-controlled film growth and smooth surface morphology of organic based magnet is crucial for future development of organic spintronic. Here we can preliminarily report the fabrication of the polymeric magnetic complex $V(TCNE)_x$ by molecular layer deposition. The developed films exhibit significant improvement on surface morphology, magnetic and electrical properties, as well as air-stability (new films air exposed maintain their properties for days instead of hours). This promising technique to grow thin films of organic-based materials enables to control growth with nanoscale accuracy and therefore allows us to deposit on flexible substrate.

Thus all basic steps for fabrication of room temperature, light-weight, flexible all organic spintronic devices were successfully performed and a promising new room temperature technology utilizing nanoscience- based fabrication FOBS is advanced.

STATISTICS FOR DOE GRANT DE-FG02-01ER45931 FOR THIS PERIOD

PUBLICATIONS (March 15, 2008 to present)

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4. J.D. Bergeson, S.J. Etzkorn, M.B. Murphey, L. Qu, J. Yang, L. Dai, and A.J. Epstein, *Iron Nanoparticle Driven Spin-valve Behavior in Aligned Carbon Nanotube Arrays*, **Applied Physics Letters** **93**, 172505/1-3 (2008).
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9. J.-W. Yoo, H.W. Jang, V.N. Prigodin, C. Kao, C.B. Eom, and A.J. Epstein, , submitted. *Giant Magnetoresistance in Ferromagnet/Organic Semiconductor/Ferromagnet Heterojunctions*, **Physical Review B** **80**, 205207/1-9 (2009).
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11. K.D. Bozdag, N.-R. Chiou, V.N. Prigodin, and A.J. Epstein, *Magnetic Field, Temperature and Electric Field Dependence of Magneto-Transport for Polyaniline Nanofiber Networks*, **Synthetic Metals** **160**, 271-274 (2010).
12. J.-W. Yoo, H.W. Jang, V.N. Prigodin, C. Kao, C.B. Eom and A.J. Epstein, *Tunneling vs. giant magnetoresistance in organic spin valve*, **Synthetic Metals** **160**, 216-222 (2010).
13. V.N. Prigodin and A.J. Epstein, *Spin Dynamics Control of Recombination Current in Organic Semiconductors*, **Synthetic Metals** **160**, 244-250 (2010).
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15. M.B. Murphey, J.D. Bergeson, S.J. Etzkorn, L. Qu, L. Li, L. Dai, and A.J. Epstein, *Spin-valve Behavior in Porous Alumina-Embedded Carbon Nanotube Array with Cobalt Nanoparticle Spin Injectors*, **Synthetic Metals** **160**, 235-237 (2010).
16. J.L. Martin, J.D. Bergeson, V.N. Prigodin, and A.J. Epstein, *Magnetoresistance for Organic Semiconductors: Small Molecule, Oligomer, Conjugated Polymer, and Non-Conjugated Polymer*, **Synthetic Metals** **160**, 291-296 (2010).

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27. K.D. Bozdag, N.-R. Chiou, V.N. Prigodin, and A.J. Epstein, *Anomalous Charge Transport in Multiple Interconnected Polyaniline Nanofibers*, **Organic Electronics** **14**, 1419-1423 (2013).

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A.J. Epstein, J.-W. Yoo, V.N. Prigodin, W.W. Shum, K.I. Pokhodnya, J.S. Miller, *Irreversible Magnetic Bistability and Nucleation of Magnetic Bubbles in Layered 2D Organic-based Magnet [Fe(TCNE)(NCMe)₂][FeCl₄]*, **11th International Conference on Molecule-based Magnets (ICMM 2008)** Florence, Italy, September 21-24, 2008.

A.J. Epstein, *Organic Based Magnets and Magnetic Phenomena in Organics*, **7th KCIST (Korea Conference on Innovative Science and Technology) Electronic Properties of Carbon-based Materials**, Phoenix Park Korea, October 19, 2008.

A.J. Epstein, *Organic-based Magnets and Organic-based Spintronics, Colloquium, Korea University*, Seoul, Korea, October 21, 2008.

A.J. Epstein, *Magnetotransport in Organic Semiconductors, 2nd Topical Meeting on Spins in Organic Semiconductors (Spinos 2009)* Salt Lake City, Utah, February 4-7, 2009.

V.N. Prigodin and A.J. Epstein, *Spin Dynamics Control of Recombination Current in Organic Semiconductors, 2nd Topical Meeting on Spins in Organic Semiconductors (Spinos 2009)* Salt Lake City, Utah, February 4-7, 2009.

A.J. Epstein, *Organic Magnetoresistance, Meeting of the American Physical Society*, Pittsburgh, Pennsylvania, March 16-20, 2009.

Y.W. Yoo, V.N. Prigodin, A.J. Epstein, *Photo-induced Magnetism and Spintronics in Organic Semiconductors, Meeting of the American Physical Society*, Pittsburgh, Pennsylvania, March 16-20, 2009.

A.J. Epstein, Plenary Lecture, *Spins in Organic Semiconductors: New Physics and New Technologies*, **55th Midwest Solid State Conference, University of Iowa**, Iowa City, IA, April 18-19, 2009.

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V.N. Prigodin, C.-Y. Chen, J.W. Yoo, C.B. Eom, H.W. Jang, C.W. Bark, and A.J. Epstein, *Spin Injection/Detection with Molecule-based Magnets: New Science and Opportunities, Trends in Spintronics and Nanomagnetism (TSN-2010), Lecce, Italy, May 23-27, 2010.*

A.J. Epstein, V.N. Prigodin, J.-W. Yoo, D. Bozdag, C.-Y. Chen, L. Fang, and E. Johnston-Halperin, *Advances in Organic-based Magnets and Organic-based Spintronics, ISSP-MDF Joint International Workshop: Spin-related Phenomena in Organic Materials*, Kashiwa, Chiba, Japan, July 1-3, 2010.

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A.J. Epstein, Keynote Lecture, *Organic-based Magnets and Organic-based Spintronics, 12th International Conference on Molecule-based Magnets (ICMM 2010)*, Beijing, China, October 8-12, 2010.

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J.-W. Yoo, C.-Y. Chen, V.N. Prigodin, H.W. Jang, C.W. Bark, C.B. Eom, and A.J. Epstein, *An Organic-based Magnetic/non-magnetic Semiconductor as a Spin Polarized Carrier Source/channel*, **Electronic Material Conference 2010**, Notre Dame, Indiana, June 24, 2010.

A. Siregar, B. Li, and A.J. Epstein, *Pentacene Morphology, Ohio State University Center for Emergent Materials Research Experience in Undergraduate Education Research Symposium*, Columbus, Ohio, August 18, 2010

K.D. Bozdag, *Optical Detection of Electrical Spin Polarized Charge Injection in a $V[TCNE]_{x \sim 2}$ -based AlGaAs/GaAs Hybrid LED Structure*, **SPINOS III, 3rd Topical**

Meeting on Spins in Organic Semiconductors Amsterdam, The Netherlands, August 30 –September 3, 2010.

SEMINARS AND COLLOQUIA (September 15, 2008 to December 14, 2010)

A.J. Epstein, *Organic-based Magnets and Organic-based Spintronics, Colloquium, Korea University*, Seoul, Korea, October 21, 2008.

A.J. Epstein, *Functional Organic and Polymer Materials from Conducting Polymers and Spintronics, Department of Chemistry, Ohio State University*, Columbus, Ohio, February 1, 2010.

J.-W. Yoo, C.-Y. Chen, V.N. Prigodin, H.W. Jang, C.W. Bark, C.B. Eom, and A.J. Epstein *Molecule-based Magnet and Organic-based Spintronics: New Science and Opportunities, Seminar at Korea Institute of Science and Technology*, Seoul, Korea, April 21, 2010.

J.-W. Yoo, C.-Y. Chen, V.N. Prigodin, H.W. Jang, C.W. Bark, C.B. Eom, and A.J. Epstein, *Molecule-based Magnet and Organic-based Spintronics: New Science and Opportunities, Seminar at Sungkyunkwan University*, Suwon, Korea, April 25, 2010.

J.-W. Yoo, C.-Y. Chen, V.N. Prigodin, H.W. Jang, C.W. Bark, C.B. Eom, and A.J. Epstein *Molecule-based Magnet and Organic-based Spintronics: New Science and Opportunities, Seminar at Korea University*, Seoul, Korea, April 27, 2010.

V.N. Prigodin and A.J. Epstein, *Organic Spintronics, Colloquium, Ioffe Institute*, St. Petersburg, Russia, May 21, 2010.

V.N. Prigodin and A.J. Epstein, *Organic Spintronics, Seminar, Naval Research Lab*, Baltimore, Maryland, August 30, 2010.

A.J. Epstein, *Towards Organic Spintronics, Colloquium, Dept. of Physics, New York University*, New York, New York, September 16, 2010.

A.J. Epstein, *Organic Spintronics, Colloquium, Institute of Chemistry, Chinese Academy of Sciences*, Beijing, China, October 8, 2010.

A.J. Epstein, *Organic Spintronics, Colloquium, Tsinghua University*, Beijing, China, October 8, 2010.

DOE SUPPORTED PERSONNEL

The Ohio State University (3/15/08 to 12/14/10)

Arthur J. Epstein, PI

K. Deniz Duman Bozdag, Graduate Research Associate

Chia-Yi Chen, Graduate Research Associate

Chi-Yueh Kao, Graduate Research Associate
Bin Li, Graduate Research Associate
Jesse Martin, Graduate Research Associate
Mark Murphey, Graduate Research Associate
Pitchi Raju Nandyala, Research Associate 2
Jung Woo Yoo, Postdoctoral Researcher

University of Utah (3/15/08 to 12/14/10)

Joel S. Miller, PI
Jordan Arthur, Graduate Research Associate
Jack Dasilva, Graduate Research Associate
Bretni Skye Cartisano Kennon, Postdoctoral Researcher
Christopher Kareis, Graduate Research Associate
Preston Erickson, Undergrad Lab Assistant

PLANNED ACTIVITIES FOR FUTURE

As before our efforts are directed on study of the interfaces between the different organic materials such as organic semiconductors and magnetic and non-magnetic metals such as Fe, Co, Ca, Al, and so on. In organic LEDs and spin valves injection at interface controls the IV characteristics and device efficiency. The study of magnetic field and electric field dependencies as well as temperature dependencies of the device current should shed light on the mechanism of injection. Additionally varying materials with different work function allows us to establish the energy levels and nature of interface states.

We are planning intensive study of $V(TCNE)_x$ in a number of aspects. First, since we have developed a new route to synthesize a “new” $V(TCNE)_x$ which is more dense, more highly electrically conductive and more air stable, we are eager to study its physical characteristics and the structure to understand what is “new” and which property is responsible for those superior behavior.

Second, study of spin valve with regular $V(TCNE)_x$. We plan to replace LSMO electrode with Fe to demonstrate the spin valve effect with $V(TCNE)_x$ at room temperature. Then we are going to use $V(TCNE)_x$ for both electrodes with different thickness or compositions. We anticipate such $V(TCNE)_x$ films have different coercive fields and therefore such a spin valve should work. Eventually in this way we plan to fabricate spin filters with $V(TCNE)_x$.

Third, we also plan to study in more detail the energy diagrams or positions of conductive level of regular $V(TCNE)_x$. It is important to carry out more sophisticated studies of the mechanism of charge transport in $V(TCNE)_x$, in particular, the role of 3d levels of Vs in charge transport because of its strong hybridization with p^* levels of TCNEs. To this purpose we plan to study a number of spin valves with varying strength and polarity of bias, magnetic field and thickness of spacer.

CURRENT AND PENDING SUPPORT FOR THIS PERIOD (March 15, 2008 to December 14, 2010)

Current

Agency: National Science Foundation

Title: Tuning the Current of Organic Semiconductors by Magnetic Fields

Total Award Period Covered: 6/1/08 to 5/31/11

Total Award Amount: \$422,811

Notes: This program complements and does not duplicate DE-FG02-01ER45931.

Agency: National Science Foundation

Title: Materials Research Science and Education Center (MRSEC)

Total Award Period Covered: 9/1/2008 to 8/30/2014

Total Award Amount: ~\$240,000 (Epstein share of total budget of \$24 million)

Notes: This program complements and does not duplicate DE-FG02-01ER45931.

Agency: National Science Foundation Division of Engineering, Education and Centers (NSEC)

Title: CANPBD II: PANI Nanofibers for Biosensing

Total Award Period Covered: 9/1/09-8/31/10

Total Award Amount: \$50,739

Notes: This program complements and does not duplicate DE-FG02-01ER45931.

Agency: UES Inc. (in contract with Air Force Research Lab)

Title: Electric and Magnetic Measurements of Photomagnet-Fullerosome Conjugates

Total Award Period Covered: 9/23/10-2/23/11

Total Award Amount: \$30,000

Notes: This program complements and does not duplicate DE-FG02-01ER45931.

Pending

None