Final Report: Room-Temperature Spin-Mediated Coupling in Hybrid Magnetic, Organic, and Oxide Structures and Devices

During the full period of this project we have (1) demonstrated room-temperature effects of the remanent magnetization from electrically-isolated magnetic films on the conductivity and electroluminescence of organic devices, (2) developed and applied a percolation theory of magnetoresistance to describe magnetic field effects on spin transport in organic semiconductors, (3) discovered and theoretically explained the effects of traps and unpaired spins on room-temperature magnetoresistance, (4) developed a theory for spin diffusion in hopping transport due to hyperfine interaction and spin-orbit interaction, (5) predicted and measured spatial interference of spin transport due to hyperfine interaction and spin-orbit interaction.
Final Report: Room-Temperature Spin-Mediated Coupling in Hybrid Magnetic, Organic, and Oxide Structures and Devices

ABSTRACT

During the full period of this project we have (1) demonstrated room-temperature effects of the remanent magnetization from electrically-isolated magnetic films on the conductivity and electroluminescence of organic devices, (2) developed and applied a percolation theory of magnetoresistance to describe magnetic field effects on spin transport in organic semiconductors, (3) discovered and theoretically explained the effects of traps and unpaired spins on room-temperature magnetoresistance, (4) developed a theory for spin diffusion in hopping transport due to hyperfine interaction and spin-orbit interaction, (5) predicted and measured spatial interference patterns from multiple coherent spin torque oscillators, (6) predicted spin-wave dispersion relations in magnonic crystals, (7) predicted electric-field control of spin-wave propagation velocities and interferometers, (8) developed a theory for spin lifetimes and spin Hall conductivities for oxide two-dimensional electron gases, (9) measured spin-orbit interactions in nanoscale wires in oxide semiconductor interfaces, (10) demonstrated electrical control of ferromagnetism in the LAO/STO system, (11) demonstrated hybrid organic/inorganic spin valves, (12) developed interfacial magnetic systems based on oxide superlattices, and (13) demonstrated GHz operation of sketched oxide transistors at the LAO/STO interface.
Enter List of papers submitted or published that acknowledge ARO support from the start of the project to the date of this printing. List the papers, including journal references, in the following categories:

(a) Papers published in peer-reviewed journals (N/A for none)

<table>
<thead>
<tr>
<th>Received</th>
<th>Paper</th>
</tr>
</thead>
<tbody>
<tr>
<td>08/31/2011 16.00</td>
<td>Cheng Cen, Daniela F Bogorin, Jeremy Levy. Thermal activation and quantum field emission in a sketch-based oxide nanotransistor, Nanotechnology, (12 2010): 475201. doi:</td>
</tr>
<tr>
<td>08/31/2011 11.00</td>
<td>Ran Lin, Fujian Wang, Markus Wohlgenannt, Chunyong He, Xiaofang Zhai, Yuri Suzuki. Organic spin-valves based on fullerene C60, Synthetic Metals, (04 2011): 553. doi:</td>
</tr>
<tr>
<td>08/31/2011 8.00</td>
<td>Ferran Macià, Andrew D Kent, Frank C Hoppensteadt. Spin-wave interference patterns created by spin-torque nano-oscillators for memory and computation, Nanotechnology, (03 2011): 0. doi: 10.1088/0957-4484/22/9/095301</td>
</tr>
</tbody>
</table>

Oleg Chalaev, G. Vignale, Michael E. Flatté. Spin-orbit interaction from low-symmetry localized defects in semiconductors, EPL (Europhysics Letters), (04 2012): 0. doi: 10.1209/0295-5075/98/17013


F. Macià, P. Warnicke, D. Bedau, M.-Y. Im, P. Fischer, D.A. Arena, A.D. Kent. Perpendicular magnetic anisotropy in ultrathin Co|Ni multilayer films studied with ferromagnetic resonance and magnetic x-ray microspectroscopy, Journal of Magnetism and Magnetic Materials, (03 2012): 3629. doi:


Ferran Macià, Fujian Wang, Nicholas J. Harmon, Andrew D. Kent, Markus Wohlgenannt, Michael E. Flatté. Organic magnetoelectroluminescence for room temperature transduction between magnetic and optical information, Nature Communications, (04 2014): 3609. doi: 10.1038/ncomms4609


TOTAL: 57
Number of Papers published in peer-reviewed journals:

(b) Papers published in non-peer-reviewed journals (N/A for none)

Received Paper


TOTAL: 1
Number of Papers published in non peer-reviewed journals:

(c) Presentations

“Room-Temperature Electronically-Controlled Ferromagnetism at the LaAlO3/SrTiO3 Interface”, Feng Bi, Mengchen Huang, Chung-Wung Bark, Sangwoo Ryu, Chang-Beom Eom, Patrick Irvin, Jeremy Levy, APS March Meeting 2014, Denver, CO


“Spin Torque Oscillators with Perpendicularly Magnetized Layers,” A. D. Kent, Current-Driven Magnetization Dynamics Workshop, Leeds, UK


“Fringe-field Organic Magnetoresistance”, M. E. Flatté, Spin Chemistry Meeting (SCM-2013), Bad Hofgastein, Austria, April 25, 2013.


“Spin Relaxation Theory in Amorphous Silicon and Germanium”, N. Harmon and M. E. Flatté, American Physical Society March Meeting


Number of Presentations: 23.00

<table>
<thead>
<tr>
<th>Non Peer-Reviewed Conference Proceeding publications (other than abstracts):</th>
</tr>
</thead>
<tbody>
<tr>
<td>Received</td>
</tr>
</tbody>
</table>

TOTAL:

Number of Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

<table>
<thead>
<tr>
<th>Peer-Reviewed Conference Proceeding publications (other than abstracts):</th>
</tr>
</thead>
<tbody>
<tr>
<td>Received</td>
</tr>
</tbody>
</table>

TOTAL:
(d) Manuscripts

Received          Paper


08/31/2012 21.00 Franklin J. Wong, Miaofang Chi, Rajesh V. Chopdekar, Brittany B. Nelson-Cheeseman, Nigel D. Browning, Yuri Suzuki. Potential Barrier Lowering and Electrical Transport at the LaAlO3/SrTiO3 Interface, Submitted to physical review letters (09 2008)

08/31/2012 49.00 Nicholas Harmon, Michael E. Flatté. Magnetoresistance in Organic Semiconductors, SPIE: Optics+Photonics (07 2012)

08/31/2012 42.00 Oleg Chalaev. Matrices, bases and matrix elements for cubic double crystallographic groups, Preprint (06 2012)


09/01/2011 10.00 P. Warnicke, D. Bedau, M.-Y. Im, F. Macia, P. Fischer, D. A. Arena, A. D. Kent. Perpendicular magnetic anisotropy in ultrathin Co|Ni multilayer films studied with ferromagnetic resonance and magnetic x-ray microspectroscopy, Submitted to Journal of Magnetism and Magnetic Materials (09 2011)

09/01/2011 18.00 Cheng Cen, Daniela F. Bogorin, Vanita Srinivasa, Jeremy Levy. Quantum Transport in Oxide Nanostructures, Submitted to physical review letters 1103.036 (09 2011)

09/23/2009 1.00 E. Golovatski, M. Flatte. Spin torque and charge resistance of ferromagnetic semiconductor 2 and domain walls, ( )

12/05/2011 22.00 Glade Sietsema, Michael Flatté. Magnonic band structure of a two-dimensional magnetic superlattice, (submitted) (11 2011)

12/05/2011 23.00 Elizabeth Golovatski, Michael Flatté. Domain wall attraction and repulsion during spin-torque-induced coherent motion, (submitted) (11 2011)

TOTAL: 18

Number of Manuscripts:

Books

Received Book


TOTAL: 2
TOTAL:

**Patents Submitted**

**Patents Awarded**


**Awards**

Ted Sanders, National Science Foundation Predoctoral Fellowship

<table>
<thead>
<tr>
<th>NAME</th>
<th>PERCENT_SUPPORTED</th>
<th>Discipline</th>
</tr>
</thead>
<tbody>
<tr>
<td>Matthew Gray</td>
<td></td>
<td></td>
</tr>
<tr>
<td>FTE Equivalent:</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Total Number:</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

**Names of Under Graduate students supported**

<table>
<thead>
<tr>
<th>NAME</th>
<th>PERCENT_SUPPORTED</th>
<th>Discipline</th>
</tr>
</thead>
<tbody>
<tr>
<td>Matthew Entwistle</td>
<td></td>
<td>Physics</td>
</tr>
<tr>
<td>FTE Equivalent:</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Total Number:</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
**Student Metrics**

This section only applies to graduating undergraduates supported by this agreement in this reporting period.

The number of undergraduates funded by this agreement who graduated during this period: ...... 0.00

The number of undergraduates funded by this agreement who graduated during this period with a degree in science, mathematics, engineering, or technology fields:...... 0.00

The number of undergraduates funded by your agreement who graduated during this period and will continue to pursue a graduate or Ph.D. degree in science, mathematics, engineering, or technology fields:...... 0.00

Number of graduating undergraduates who achieved a 3.5 GPA to 4.0 (4.0 max scale):...... 0.00

Number of graduating undergraduates funded by a DoD funded Center of Excellence grant for Education, Research and Engineering: ...... 0.00

The number of undergraduates funded by your agreement who graduated during this period and intend to work for the Department of Defense ...... 0.00

The number of undergraduates funded by your agreement who graduated during this period and will receive scholarships or fellowships for further studies in science, mathematics, engineering or technology fields: ...... 0.00

---

**Names of Personnel receiving masters degrees**

<table>
<thead>
<tr>
<th>NAME</th>
<th>Total Number:</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
</tr>
</tbody>
</table>

**Names of personnel receiving PHDs**

<table>
<thead>
<tr>
<th>NAME</th>
<th>Total Number:</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tianyu Liu</td>
<td>2</td>
</tr>
<tr>
<td>Daniel Gopman</td>
<td></td>
</tr>
</tbody>
</table>

**Names of other research staff**

<table>
<thead>
<tr>
<th>NAME</th>
<th>PERCENT_SUPPORTED</th>
</tr>
</thead>
<tbody>
<tr>
<td>FTE Equivalent:</td>
<td></td>
</tr>
<tr>
<td>Total Number:</td>
<td></td>
</tr>
</tbody>
</table>

**Sub Contractors (DD882)**

**Inventions (DD882)**
During the final year, which was a no-cost extension, some final projects were brought to completion or further ahead, especially those associated with the oxide effort that had been delayed during Suzuki's relocation to Stanford University.

**Imaging spin-wave patterns with XMCD**

Using XMCD the emission of spin-wave patterns from a spin torque nanocontact has been directly observed (slide #1 attached). This was one of the main goals of this project. Magnetic soliton propagation can be seen as well, and temporally resolved. This was work done at SLAC in collaboration with Dr. Hendrik Ohldag. We used his scanning x-ray transmission microscope (STXM) at beamline 13-1 to image the excitation patterns in STNO. We focused on the Co L-edge and were able to observe localized spin-waves at the electric contact to a NiCo multilayer film. We are presently analyzing the results and preparing an article on these results for publication. This work represents the first time x-rays have been used to observe spin-waves associated with the spin-transfer interaction.

**Organic Magnetoresistance via traps and radical spins**

A theory of organic magnetoresistance via traps has been formulated and is in agreement with the experimental measurements previously reported for this MURI by the Wohlgenannt/Suzuki collaboration. The compared results are shown in slide #2, attached. Enhancing the organic magnetoresistance was one of the main goals of this project, and this has now been achieved and explained.

Controlled doping of an organic material using radical spins can suppress the organic magnetoresistance, due to the exchange coupling between the radical spins and the spin transport sites. A new regime, where the radical spin exchange couples strongly with one site of a bottleneck pair, has been demonstrated and the results published in PRB Rapid Communications.

**Spin Hall effect in oxides**

Collaborative work between Vignale and Flatté has shown that strain at the LaAlO3/SrTiO3 can suppress the spin Hall conductivity, as shown in slide #3 attached. A paper on these results is in preparation for submission, including strain along arbitrary axes.

Suzuki has made significant progress over the past year in understanding the role of disorder in the transport characteristics of LaAlO3/SrTiO3 interfaces through the study of mobility and carrier concentration in doped LaAlO3/SrTiO3 interfaces. Furthermore she has found unexpected long-range antiferromagnetic correlations and structural distortions in CaRuO3/CaMnO3 superlattices that may explain the modulation of the interfacial ferromagnetism with CaMnO3 thickness.

**Interfacial ferromagnetism in CaRuO3/CaMnO3 superlattices**

Suzuki has performed studies of the structural distortions associated with these superlattices. Detailed analysis of half order peaks in the X-ray diffraction indicates the doubling of the unit cell for superlattices with particular combinations of even and odd CaMnO3 and CaRuO3 layers. The doubling of the unit cell may be correlated with the modulation of the interfacial ferromagnetism that we had observed previously. The interfacial moment alternates between 1µB/interface ion for even CaMnO3 layers and 0.5 µB INTERFACE Mn ion for odd CaMnO3 layers. This modulation may be explained in terms of long-range antiferromagnetic correlation of the bulk of the antiferromagnetic CaMnO3 layers combined with the doubling of the unit cell observed by x-ray diffraction.

**Doped quasi-2DEG at the interface of LaAlO3/SrTiO3.**

Through her study of rare earth doped LaAlO3 on SrTiO3 interface, Suzuki has discovered that the presence of dopants on the LaAlO3 side of the interface does not significantly affect the mobility, carrier concentration as well as magnetotransport compared to undoped LaAlO3 films. (This is shown on slide #4 attached) The motivation behind the rare earth doping is to insert additional spin-orbit coupling, in the case of Lu, and spin-orbit coupling as well as magnetic moment, in the case of Tm, on the LaAlO3 side of the LaAlO3/SrTiO3 interface. We had previously found that there is (i) an inverse relationship between mobility and carrier concentration that is not consistent with impurity scattering dominated transport and (ii) a steep dependence of the mobility with carrier concentration similar to undoped LaAlO3/SrTiO3. Detailed analysis suggests that mobility and carrier concentration values derived from Hall effect measurements are extremely sensitive to the assumptions associated with the types of carriers. More specifically many groups appear to extract carrier concentration and mobility values by fitting a linear dependence of Hall voltage on magnetic field. Non-linear Hall curves that appear at lower temperatures suggest that perhaps multiband transport is at work. However since there is no unique fit of mobility and carrier values, it is extremely difficult to accurately compare the trends in mobility and carrier concentration. This uncertainty combined with the similarity in doped and undoped samples indicate that transport in this system is dominated by disorder.

**Nanoscale control of the metal-insulator transition of the LaAlO3/SrTiO3 interface with a conductive-atomic force microscope (c-**
AFM) technique by Levy has enabled a variety of electrical and photonic device concepts and provides a pathway for new types of reconfigurable, oxide-based nanoelectronics. The oxide heterostructure LaAlO3/SrTiO3 supports a two-dimensional electron liquid with a variety of competing phases, including magnetism, superconductivity, and weak antilocalization because of Rashba spin-orbit coupling. Further confinement of this two-dimensional electron liquid to the quasi-one-dimensional regime can provide insight into the underlying physics of this system and reveal new behavior.

Levy performed transport experiments with nanowire-based single-electron transistors at the interface between SrTiO3 and a thin layer of lanthanum aluminate, LaAlO3. These experiments demonstrate the existence of a robust electronic phase in which electrons pair without forming a superconducting state. Key experimental signatures are captured by a model involving an attractive Hubbard interaction that describes real-space electron pairing as a precursor to superconductivity. Examples of these recent results are shown in the attached slide #5.

**Technology Transfer**

Support has been received from the UI GAP Fund to commercialize the fringe-field magnetoresistive devices. This funding provides support for demonstration of a prototype using thermally-activated delayed fluorescence materials. A start-up company is contemplated to further develop the technology.
imaging spin-wave patterns using XMCD

PRL 115, 127205 (2015)

XMCD contrast

Δμ_{On/Off} t

+0.003

0

-0.003

σ^+

28.8 mA

29.9 mA

σ^+

31.0 mA

33.2 mA

34.3 mA

σ^−

400 nm

2(b)

(a)

FIG. 2 (color online). XMCD images of the nanocontact region showing XMCD contrast in our STXM images [see Fig. 3(a)]. The envelope of the propagating mode clearly corresponds to a Co thickness of 150 nm. It is instructive to compare the measured spin-polarized x-ray transmission with a theoretical model that describes small amplitude excitations by linear propagating and localized modes predicted by Slonczewski and Buschow. Theoretical predictions can be useful for understanding the origin of the observed effects and for the interpretation of the data.

The observed excitations can be explained by considering a propagating mode predicted by Slonczewski in a perpendicular magnetic configuration. The observed excitations are caused by a change in the magnetic anisotropy of the nanocontact, which is responsible for the observed XMCD contrast.

The spatial extension of the observed excitations is mostly localized, as shown in Fig. 3(a). This is consistent with a decrease in the XMCD contrast in our STXM images and is consistent with a reduction in the vertical component of the magnetization.

The observed contrast reverses its sign upon reversing the x-ray helicity. Each point (black squares) represents an average of the raw (unfiltered) XMCD data over a half-week ending 18 SEPTEMBER 2015.

We do not believe this represents a change in the vertical component of the magnetization and only probe changes in the element specificity and composition.

The observed contrast is consistent with a reduced magnetization component, as shown in Fig. 3(a). This suggests that the observed feature appears abruptly at a certain distance to the right of the nanocontact center for a current of 28.8 mA at negative x-ray helicity. The fact that the observed contrast reverses its sign upon reversing the x-ray helicity is consistent with a reduced magnetization component.

We continue by quantitatively analyzing the image contrast by constructing one-dimensional profiles through the nanocontact. This is shown in Fig. 3(a). The envelope of the propagating mode clearly corresponds to a Co thickness of 150 nm. It is instructive to compare the measured spin-polarized x-ray transmission with a theoretical model that describes small amplitude excitations by linear propagating and localized modes predicted by Slonczewski and Buschow.

The observed excitations can be explained by considering a propagating mode predicted by Slonczewski in a perpendicular magnetic configuration. The observed excitations are caused by a change in the magnetic anisotropy of the nanocontact, which is responsible for the observed XMCD contrast.

The spatial extension of the observed excitations is mostly localized, as shown in Fig. 3(a). This is consistent with a decrease in the XMCD contrast in our STXM images and is consistent with a reduction in the vertical component of the magnetization.

The observed contrast reverses its sign upon reversing the x-ray helicity. Each point (black squares) represents an average of the raw (unfiltered) XMCD data over a half-week ending 18 SEPTEMBER 2015.
organic magnetoresistance from traps

Let us now turn to the discussion of the experimental findings, starting with the reduction in spin-diffusion length. For the purpose of this discussion, we will compare the magnetoconductance traces shown are for the bias voltage that gives the largest magnetoconductance [see Figs. 2(a) and 2(b)]. The pristine device (scatter plots with square symbol) and an x-ray-exposed device (scatter plots with triangle symbol, 20 mins exposure time). From this data the spin-diffusion length can be extracted. (b) Voltage dependence of the observed spin-valve response at 12 K, both for a pristine (thick lines) and irradiated (thin lines) devices at 12 K [4]. Figure 3 shows measured magnetoconductance traces for the pristine device. The three panels show that the irradiated devices exhibit a spin-diffusion length of approximately 0.5%. This value decreased to 3.5% and 7.0% for the prepared devices, whereas this value increased to 7.7% and 15.5% for the pristine and x-ray-exposed devices. The pristine device was exposed to x-rays, and spin-valve devices, starting with the OMAR devices. Dediu and co-workers [25] suggested a mechanism related to bistability effects recently reported in organic spin-valves. Next, we investigate the effect of traps on the spin-diffusion length. For this bistability based on charge storage in deep traps. Our experiment also supported by MURI: JAP 116, 043707 (2014) theory: PRL 109, 076603 (2012)

We will now discuss the effect of traps on both OMAR and spin-valve devices, starting with the OMAR devices. To the best of our knowledge, two different reports [20] that demonstrated that the production of reactions that produce traps and luminescence quenchers. Comparing Fig. 2(a) with the corresponding luminescence data in Figs. 1(a) and 1(b), we arrive at the, experiment also supported by MURI previously published.
strain effect on SHC in STO 2degs

Increasing strain reduces intrinsic Spin Hall Conductivity.
rare-earth doping of LAO/STO

mobility anticorrelated with sheet density - independent of doping character
Critical pairing fields markers of ZBP at the conductance versus gate voltage at zero bias (Fig. 2f) enables the demonstration of single-electron transistor behavior in the sketch-FET, and the negative-U model describes the filling. The negative-U is sufficient to pair the electrons on the nanoscale.