Laboratory Name: SLAC
B&R Code: KC0202010

FWP and possible subtask under FWP:
FWP: PULSE: The Stanford Photon Ultrafast Science and Engineering Center
Subtask D.2.a: Experiments with variable electron bunch length

FWP Number: SCW0063

Program Scope: This program investigates the interactions of highly relativistic electron bunches and its associated electric and magnetic fields with solid materials in the form of very thin films. The goal of the program is the elucidation of the response of a magnetic material under extreme conditions, created by electro-magnetic fields of unprecedented intensity. Of particular interest are the magnetic interactions between the magnetization of the film and the ultrafast and ultrastrong electric and magnetic fields. The fields of the electron bunch closely resemble half-cycle terahertz radiation that can be produced by other means, yet our fields are stronger by one to two orders of magnitude.

Major Program Achievements (over duration of support):
We have studied the characteristic patterns written into thin magnetic films by electron beam produced in the SLAC linac with pulse lengths varying from 100 fs to several picoseconds and field strengths up to tens of GV/m (E-field) and Tesla (B-fields). These experiments have produced three major results:
(1) ***The transformation of the electronic and magnetic structure of a metal by extreme terahertz fields***. The extreme electric field of the bunch modifies the magnetic pattern through creation of a novel magneto-electronic anisotropy and avoids current induced Joule-heating by creating a potential difference between adjacent atoms that disrupts band-like conduction.
(2) ***Non-linear transport induced in a metal by ultrastrong electric fields.*** Non-linear electron transport is common with *artificial* superlattices created from *semiconductors.* The contact-free application of very high electric fields to metals achieved by us for the first time shows the existence of similar phenomena in metals and opens the opportunity to temporarily modify the electronic structure of metals.
(3) ***Magnetic Anisotropy induced in a metal by an electric field pulse.*** Using spin precession as a diagnostic tool, we observe the generation of a large new type of magneto-electronic anisotropy in a ferromagnetic thin film subjected to ultra-fast (70 fs) and ultra-strong ($>10^9$ V/m) electric field pulses. The *E*-field induced anisotropy can assist switching of the magnetization in a thin film.

Program impact:
Ultrafast magnetization dynamics is of great basic interest as it reveals our limited knowledge of the behavior of common correlated electron systems such as the 3d-transition metals under extreme conditions. Additionally, the speed of magnetic switching is of technical interest in magnetic data storage. The fact that it can be accomplishment by terahertz pulses is a novelty with possible far reaching implications for the future of magnetic recording. Moreover, the modification of electronic properties of natural solids (as opposed to artificial superlattices) by application of electric fields promises a plethora of new applications.

Interactions:
Internal: SSRL and the PULSE center.
External: Department of Applied Physics, Stanford University, IBM Almaden Research Center San Jose, IBM Research Division Rueschlikon, Switzerland,
Bogolyubov Institute for Theoretical Physics, Kiev 03680, Ukraine

Recognitions, Honors and Awards (at least partly attributable to support under this FWP or subtask):
J. Stöhr, invited Paper at the 52nd Annual Conf. on Magn. & Magn. Mat., Tampa FL, Nov. 2007 “Toward Ultrafast Manipulation of the Magnetization”

Personnel Commitments for FY2007 to Nearest +/- 10%:
H.C. Siegmann (70%), Sara J. Gamble (100%), J. Stöhr (5%), M. Burkhardt (60%)

Authorized Budget (BA) for FY07, FY08, FY09:
FY05 BA $145 **FY06 BA** $220 **FY07 BA** $424

Laboratory Name: SLAC
B&R Code: KC0202010

FWP and possible subtask under FWP:
FWP: PULSE: The Stanford Photon Ultrafast Science and Engineering Center
Subtask D.2.b: Energy and angular momentum transfer in magnetic solids

FWP Number: SCW0063

Program Scope:
The goal of this program is to investigate the transfer of angular momentum between reservoirs in solids, using laser based approaches. Recent laser experiments by other groups show that the magnetization of a ferromagnet can be manipulated on the femtosecond time scale. Unlike other ultrafast processes, the modification of the magnetization requires transfer of angular momentum out of the spin system. In practice, this is accomplished through the spin-orbit interaction which transfers the angular momentum from the spin system to the lattice. The observed speed of this process remains a mystery, because it is expected to be slower than observed by three orders of magnitude. The goal of this project is to study the microscopic origin of angular momentum transfer, and to develop novel ways of magnetization manipulation on the ultrafast time scale.

Major Program Achievements (over duration of support):
The first year of this new program was spent to carefully plan two experiments.

The first is based on using a laboratory laser based terahertz source, with fields suitably enhanced by focusing antenna optics, to induce a motion of the magnetization in a ferromagnetic sample. This approach complements the method used in the FWP entitled "Experiments with variable electron bunch length" but does not rely on the availability of the SLAC linac. THz radiation can influence the magnetization by two mechanisms: (a) the magnetic field of the THz pulse can influence the magnetization directly, (b) the electric field of the THz pulse which can indirectly influence the magnetization by modifying the magnetic anisotropy. So far only electron accelerator sources have produced strong enough electric and magnetic fields to switch the magnetization. The use of laser based THz sources, opens the opportunity to study angular momentum transfer by sources that may be employed in technological applications.

The second experiment will utilize the Linac Coherent Light Source (LCLS) when it becomes operational in 2009, and first experiments are planned in 2008, using the free electron laser facility FLASH in Hamburg, Germany. The experiment is based on a pump-probe approach. Optical laser or soft x-ray FEL radiation pulses of femtosecond duration trigger the excitation of a magnetic sample. A soft x-ray FEL probe pulse, which is delayed relative to the pump pulse but of comparable pulse length then produces photoelectrons. Due to the high energy of the probe pulse, electrons from the entire valance band can be excited and the measured spin polarization of the emitted electrons therefore reflect the magnetic state of the sample. If the magnetization changes during the pump pulse, this will be directly seen as a change of the spin polarization produced by the probe pulse. The details of angular momentum transfer out of the spin system can therefore be directly probed. We are currently building the spin detector and planning first experiments at FLASH.

Program impact:
Not yet.
Interactions:
Internal: Stanford Nanofabrication Facility, Stanford Synchrotron Radiation Laboratory
External: Prof. Danilo Pescia, ETH Zurich; Prof. Wilfried Wurth, Free Electron Laser in Hamburg (FLASH)

Personnel Commitments for FY2007 to Nearest +/- 10%:
Yves Acremann (70%), Hans Siegmann (30%), David Bernstein (50%), Diling Zhu (50%), D. Wu (50%)

Authorized Budget (BA) for FY07, FY08, FY09:
FY05 BA $332 **FY06 BA** $378 **FY07 BA** $383

Laboratory Name: SLAC
B&R Code: KC0202010

FWP and possible subtask under FWP:
FWP: PULSE: The Stanford Photon Ultrafast Science and Engineering Center
Subtask D.2.c: Ultrafast imaging of the magnetization

FWP Number: SCW0063

Program Scope:
This program is based on the development of methods of ultrafast pump-probe and single shot imaging with LCLS. One of the goals is the single shot imaging of critical magnetic fluctuations at the ferro- to paramagnetic phase transition in a thin, quasi 2-dimensional film. This ambitious goal has been motivated by our successful demonstration of lensless imaging of a magnetic domain structure with about 50 nm spatial resolution by x-ray Fourier transform spectro-holography in 2004 (Eisebitt et al., Nature **432**, 885 (2004)). We are using the time before the actual advent of LCLS in 2009 for technique development, exploring various new concepts of time-dependent imaging.

Major Program Achievements (over duration of support):
In 2007 we demonstrated a new method, called multiple-object Fourier Transform Holography (FTH), which applies spatial multiplexing to Fourier transform holography. This technique overcomes one of the central challenges in imaging, namely to simultaneously obtain high spatial resolution and a large field of view. In conventional full-field microscopy, the two objectives compete with each other. In our experiment we demonstrated that by using multiple object and reference signals the effective field of view can be extended without compromising the numerical aperture of the detected hologram.

The key to extending the effective field of view is the strategic arrangement of reference holes with respect to neighboring object regions of interest. Images of the isolated regions should then tile, not overlap, in the autocorrelation. Furthermore, for the highest spatial resolution, the reconstructed field of view should be matched to the extent of the largest local region on the sample. This was experimentally demonstrated by recording holograms of suitable nanoscale samples prepared by ion beam milling.

Since these requirements are compatible with single shot illumination from a pulsed x-ray source, isolated regions on a sample can be imaged with a single pulse. Furthermore, by cross-beam pump-probe illumination, the utrafast temporal response of different sample regions can be simultaneously recorded. The pump laser pulse is incident at a small angle from the plane of the sample, and the x-ray probe pulse is incident along the surface normal. In this geometry, different sample regions are excited at different times but are probed at the same time. By suitably timing the pump and probe pulses, and by use of the multiple object method, it is then possible to record the temporally separated response of different sample regions through their spatial separation in the autocorrelation image.

Program impact:
Publication: W. F. Schlotter et al. Opt. Lett. **32**, 3110 (2007) - Editor's choice of Science Magazine, Science 318, 1218 (2007)
A. Scherz et al., Phys. Rev. B
Ph. D. thesis of William Schlotter, Stanford University, June 2007

Interactions:
S. Eisebitt (BESSY) and J. Lüning (Paris)

Personnel Commitments for FY2007 to Nearest +/- 10%:
Andreas Scherz (50%), Bill Schlotter (70%), Ramon Rick (50%), J. Stohr (5%)

Authorized Budget (BA) for FY07, FY08, FY09:

FY05 BA $0	**FY06 BA** $0	**FY07 BA** $167

Laboratory Name: SLAC
B&R Code: KC0202010

FWP and possible subtask under FWP:
FWP: PULSE: The Stanford Photon Ultrafast Science and Engineering Center
Subtask D.2.d: Magnetic Materials Research

FWP Number: SCW0063

Program Scope:

This program is aimed at addressing a key question about the interaction of ultrashort, high intensity X-FEL pulses with magnetic materials. Recently, ultrafast laser experiments employing fluences similar to those expected for X-FELs have raised questions regarding the utility of X-FELs for studies of the electronic structure of materials. Of central importance is the question under which conditions an XFEL pulse can be expected to act as a weak perturbation as for present x-ray sources, and when the x-ray perturbation will lead to radically altered electronic states. In the latter case, the information of the as-prepared state of the sample would be lost. This would indicate serious problems with single shot experiments aimed at recording snapshots of materials in well defined states.

Major Program Achievements (over duration of support):

We have carried out first experiments at the FLASH FEL facility in Hamburg, Germany, to address the problem whether the FEL beam modifies the electronic structure of a material in the process of interacting with it. Our first experiments employed the non-magnetic material Al, because its L absorption edge is conveniently located in the spectral range presently available at FLASH.

The Aluminum L-edge (~ 82 eV) exhibits a well defined narrow resonance feature, due to multi-electron excitations in the vicinity of the Fermi energy, the famous Mahan-Nozieres-de Dominici (MND) line-shape anomaly. When FEL radiation is absorbed by the sample, the intense beam will create a large number of photoelectrons which upon equilibration will result in a strongly modified Fermi-Dirac distribution around the Fermi energy. The shape and intensity of the L-edge peak is particularly sensitive to this modification of the electronic structure near the Fermi energy.

We have developed a technique to record x-ray absorption spectra with an FEL beam. This is accomplished by placing the thin film sample in the energetically dispersed beam after a monochromator grating and recording a series of single shot absorption spectra (in transmission geometry). The thin film sample consisted of two areas, one area is covered with the sample the other is left blank to provide the reference spectrum. The transmitted beam intensity is imaged with a 2D detector (YAG crystal + CCD camera). The intensity variation across the detector in the vertical direction then directly corresponds to the transmission spectrum of the sample and the reference intensity is horizontally offset, as determined by the sample geometry, and recorded in parallel. The final spectrum is the average of the single shot sample transmission spectra normalized by the corresponding single shot reference spectra. The samples were fabricated using conventional optical lithography. We are presently analyzing the results obtained in December 2007.

Program impact:
Not yet, just started.

Interactions:
W. Wurth, DESY, Hamburg

Personnel Commitments for FY2007 to Nearest +/- 10%:
Andreas Scherz (50%), David Bernstein (50%), Yves Acremann (30%), J. Stohr (5%)

Authorized Budget (BA) for FY07, FY08, FY09:

FY05 BA $0	**FY06 BA** $0	**FY07 BA** $195

Laboratory Name: SLAC
B&R Code: KC0202010

FWP and possible subtask under FWP:
FWP: PULSE: The Stanford Photon Ultrafast Science and Engineering Center
Subtask D.3.a: THz and strong field nonlinear spectroscopy and control of material structure

FWP Number: SCW0063

Program Scope:
Program explores the nonlinear properties of materials induced by strong fields. Special concentration is the nonlinear transient changes in materials exposed to strong and ultrafast electromagnetic fields, such as the THz fields of relativistic electron beams, as well as the strong fields of focused laser beams. Research advances are directed towards materials and x-ray science of coherent phonons in materials exposed to THz pulses; nonlinear excitation of coherent phenomenal in materials; field-induced effects in materials; transient phase changes in materials; transient domain creation and motion in materials.

Major Program Achievements (over duration of support):
Program is responsible for a non-invasive technique to record the arrival time of femtosecond electron bunches at the LCLS undulator with respect to a timing laser pulse with femtosecond resolution. The optical properties of an electro-optic crystal placed adjacent to the electron beam are strongly modified as a result of the electric-field of the electron bunch as it passes. A femtosecond laser pulse propagating through the crystal at the same time as the transient birefringence is induced has its polarization state altered and can thus be used as a probe of the relative arrival time of the electron bunch. The program also completed publication of mapping of the carrier density-dependent interatomic potential of highly excited Bi. No previous experiment has measured with both atomic spatial resolution and femtosecond temporal resolution the binding potential of a solid as it approaches a phase transition. Laser-pump x-ray probe measurement revealed electronic softening and large amplitude coherent optical phonons in Bi.

Program Impact:
This experiment makes possible a wide range of repetitive measurements with time-resolution limited by the pump and probe durations instead of the intrinsic jitter between pump and probe. Relative timing information from spatially-resolved electro-optic measurements could be extended to a resolution of order 5 fs, matching the projected performance of future XFELs into the foreseeable future. This diagnostic was duplicated at the FLASH FEL in Hamburg and will be implemented at the LCLS.

Interactions:
Internal—SPPS collaboration, LUSI project, XLAM, SSRL
External—FLASH, Hamburg; MPQ, Garching

Recognitions, Honors and Awards (at least in some part attributable to support under this program):
D.M. Fritz: 2007 SSRL William E. Spicer Young Investigator Award (co-winner); University of Michigan Rackham School of Graduate Studies Distinguished Dissertation Award

Personnel Commitments for FY2007 to Nearest +/- 10%:
David Fritz (1 month).

Authorized Budget (BA) for FY07, FY08, FY2009:
FY05 BA $0k　　**FY06 BA $**0k　　**FY07 BA $**331k

Laboratory Name: SLAC
B&R Code: KC0202010

FWP and possible subtask under FWP:
FWP: PULSE: The Stanford Photon Ultrafast Science and Engineering Center
Subtask D.3.b: Laboratory-based THz pump experiments

FWP Number: SCW0063

Program Scope:
A laboratory-based source for intense, ultrashort THz radiation is applied to investigate and control the electronic and structural properties of materials under intense electric and magnetic fields. We also seek to develop the field of ultrafast nonlinear THz spectroscopy as a new tool in materials science.

Major Program Achievements (over duration of support):
Since the turn-on of the lab in early FY07, commissioned and developed among the strongest field laboratory-based THz sources in the world. Demonstrated THz control of carrier dynamics in semiconductors. Observed, for the first time, nonlinear THz absorption effects associated with ultrafast, high intensity THz fields in materials. Developed THz pump/THz probe method and observed impact ionization effects associated with the strong field – carrier interaction. Investigated strong-field THz interaction with nanoscale materials including nanowires and nanocrystals., with application to elucidating means for increasing the quantum efficiency of solar cells and other energy-related materials.

Program Impact: Developing nonlinear THz spectroscopy as a tool for probing and controlling material properties. Developing methodology for all-optical control of material structural and electronic dynamics, including ferroelectric and ferromagnetic materials, semiconductors, nanoscale materials. This program also lays a foundation for future THz-based experiments using ultrafast electron bunches produced by the linear accelerator at SLAC.

Interactions:
Internal—Stanford Materials Science and Engineering Dept., Stanford Synchrotron Radiation Laboratory
External—Advanced Light Source at Lawrence Berkeley National Laboratory, Advanced Photon Source at Argonne National Laboratory, Materials Science Division at Argonne National Laboratory (developing ultrafast THz pump/X-ray probe experiments).

Recognitions, Honors and Awards (at least in some part attributable to support under this program):
A.M. Lindenberg – Named Terman Fellow, 2007-2010

Personnel Commitments for FY2007 to Nearest +/- 10%:
Haidan Wen (100%), Aaron Lindenberg (31%)

Authorized Budget (BA) for FY05, FY06, FY2007:
FY05 BA $200k **FY06 BA** $130k **FY07 BA $**175k

Laboratory Name: XLAM/SSRL, SLAC
B&R Code: KC0202010

FWP and possible subtask under FWP:
FWP: BES Materials Sciences (XLAM); Subtask 1: Spectroscopy & Scattering Study of Correlated Materials
FWP Number: SCW0035

Program Scope:
Application of advanced photoelectron spectroscopy, x-ray scattering and neutron scattering methods to elucidate the electronic, magnetic, orbital and lattice properties of novel materials: (1) Conduct cutting edge research by correlating information gained from single particle (photoemission) and two particle (neutron and x-ray scattering) response functions. (2) Provide scientific impetus for developing state-of-the art capabilities at DOE's user facilities. (3) Develop crystal growth and characterization capabilities. These materials have enabled internal research at SLAC as well as extensive collaborations, nationally and internationally. (4) Develop human resources by training and educating students and post-docs.

Major Program Achievements (over duration of support):
Neutron scattering: results for the spin excitations in the electron-doped superconductor NCCO reveal two characteristic energy scales, and suggest that prior suggestions of a need to reinterpret prior claims of the existence of a magnetic resonance; in this system have to be reinterpreted; polarized neutron diffraction for the model superconductor Hg1201 reveals that the pseudogap phase is associated with `hidden' magnetic order; initial inelastic neutron data for Hg1201 reveal that the magnetic resonance has a relatively high energy in this system (three manuscripts in preparation)
X-ray scattering: continuation of detailed Cu K-edge resonant inelastic X-ray scattering (RIXS) study of model Mott insulators as a function of photon energy and polarization; discovery of oxygen-order tendencies in Hg 1202 (two manuscripts in preparation).
ARPES: the photoemission program remains active in its study of novel materials, in particular the strongly correlated complex oxides, with selected highlights here: Anomalous Fermi-Surface dependent pairing in multilayer high-Tc superconductors [Phys. Rev. Lett., 97, 26401 (2006); Phys. Rev. Lett., 98, 407001 (2007)]; Evidence for the presence of two energy gaps [Science 314, 1910 (2006); Nature, 450, 81 (2007)]; Unusual electron emission from self-assembled monolayers of functionalized diamondoids [Science, 316, 1460 (2007)]; Fermi surface and quasiparticle renormalization in $Sr_{2-y}La_yRuO_4$ [Phys. Rev. Lett. 99, 187001 (2007)]; Hierarchy of many-body and electron-phonon interactions in high-Tc superconductor [Phys. Rev. B 75, 174506 (2007); 195116 (2007)]

Program Impact:
Provided fresh insights into electronic, magnetic and lattice properties for a number of important material families, and especially into the nature of the enigmatic pseudogap phase. Grow large single crystals of Hg-based compounds, the cuprates with the highest T_c, enabled collaborations.

Interactions:
Alloul, H (Paris, France);Basov, DN (UCSD); Bianconi, A (Rome, Italy); Bobroff, J (Paris, France);Bontemps, N (ESPCI, France); Bourges, P (LLB, France); Bridges, F (UCSC); Brown, SE (UCLA); Casa, D (APS); Dessau, D (Colorado);Gog, T (APS); Haase, J (Leipzig,Germany); Hasan, Z (APS);Homes, CC (Brookhaven); Ishii, K (Spring8, Japan); Kapitulnik, A (Stanford); Klein, MV (UIUC); Manoharan, HC (Stanford);van der Marel (Geneva, Switzerland); Meingast, C (Karlsruhe, Germany);Mizuki, J (Spring8, Japan); Ong, NP (Princeton); Oyanagi,H (AIST, Japan);Petitgrand, D (LLB, France); Schlichter, C (UIUC);Rubenhausen, M (Hamburg, Germany); Uemura, Y (Columbia); Vajk, OP (Missouri); Hussain (ALS), Zaanen (Leiden); Fujimori, Uchida, Nagaosa (Tokyo); Eisaki (AIST); Baumberger, Mackenzie (St. Andrews); Kim (Yongsei); Ando (Criepi), Fisher, Melosh, Kelly (Stanford), Yang, Hussain (ALS), Dahl, Carlson (Chevron)

Recognitions, Honors and Awards (at least partly attributable to support under this FWP or subtask):
2007 APS Fellowship: Greven. Large number of invited talks: Greven, Shen, Hancock, Mannella, Cuk; McMillan Award: Armitage; Student Poster award at SSRL users' meeting, Lee; Chaired Professorship at Stanford: Shen.

Personnel Commitments for FY2007 to Nearest +/-10%:
Z.-X. Shen 20%, M. Greven 20%, J. Hancock 100%, Yu, 50%, G. Chabot-Couture 50%, E. Motoyama 50%, Y. Li 50%, Y. Chen 25%, M., W.-S. Lee 50%, W. Meevasana 50%, F. Schmitt 50%, K. Tanaka 40%, S. Fujimori 100%, S. Qiao 30%.

Authorized Budget (BA) for FY05, FY06, FY07
FY05 BA $1,023K **FY06 BA** $ 1,009K **FY07 BA** $1,060K

Laboratory Name: XLAM / SSRL,SLAC
B&R Code: KC0202010

FWP and/or subtask Title under FWP:
FWP: Basic Energy Sciences Materials Sciences (XLAM); Subtask 2: Novel materials and model systems for the study of correlated phenomena
FWP Number: SCW0035

Program Scope:
(1) Obtain fundamental understanding of correlated electron behavior in complex materials by creating and studying simpler systems; (2) Design and synthesis of materials for desired properties and functions; (3) Integration of high end physical characterization in the process of materials development; (4) Act as a national resource for materials development and characterization, and in training students in these fields.

Major Program Achievements (over duration of support):

Role of valence skipping elements in correlated electron behavior: Superconductivity observed in Tl-doped PbTe and more recently In-doped SnTe. Thermodynamic, transport and spectroscopic measurements indicate mixed valence directly associated with onset of superconductivity, and exotic charge Kondo effect.

Chemical pressure and CDW formation: Investigation of RTe_2, RTe_3 & R_2Te_5 (R = rare earth) as weak-coupling analogs of charge-ordered states in more strongly correlated systems, reveal effect of band filling, "hidden 1-dimensionality" and chemical pressure on CDW formation. Observation of novel "rectangular" CDW state in RTe_3 for heaviest rare-earths.

5d magnetism: Ba_2NaOsO_6 investigated as model $5d^1$ compound. Unusual appearance of ferromagnetism attributed to orbital ordering effects.

Correlation in $SrRuO_3$: Investigation of the photoemission spectra of $SrRuO_3$ thin films with varying cation stoichiometry shows that the degree of correlation in this material grows as the Ru vacancy concentration grows. This novel behavior also explains the difference in properties between MBE and PLD made films of $SrRuO_3$, which has been a long-standing mystery.

Charge ordering in CuO: We have successfully synthesized tetragonal thin films of the normally monoclinic compound CuO using epitaxial growth. The structure and valence of the CuO were confirmed by in situ RHEED, UPS and XPD (x-ray photo diffraction).

Conducting $LaAlO_3/SrTiO_3$ interfaces: The remarkable discovery of a conducting layer at the interface between these two insulators has been found to be a result of oxygen vacancies in the $SrTiO_3$ introduced by the PLD deposition process. A high mobility results due to the vary large dielectric constant of $SrTiO_3$, that permits the carriers to move well away from the dopant centers, as in modulation doping.

Program impact:
Materials physics: substantial advances in understanding aspects of correlated electron behavior.
New materials for DOE BES: extensive collaboration based on the materials developed in this program.

Interactions:
L. Balicas (NHMFL, Tallahassee), D. Basov (UCSD), S. Billinge (Michigan), D. Blank and G. Koster (Twente University, Netherlands), F. Bridges (UCSC), V. Brouet (U. Paris Sud, France), S. Brown (UCLA), D. Casa (APS, Argonne National Laboratory), K. Char (Seoul University, Korea), A. Cottet (Orsay), L. DiGiorgi (ETH Zürich, Switzerland), S. Dugdale (Bristol University, UK), E. M. Forgan (Birmingham, UK), T. Gog (APS), C. Gough (Birmingham, UK), S. Grenier (Rutgers State University and Brookhaven), W.A. Harrison (Stanford), N. Hussey (Bristol, UK), Z. Islam (APS, ANL), A. Kaminski (Ameslab), Y.J. Kim (Toronto),L. Klein (Bar Elan University, Israel), M.V. Klein (Urbana), J. Krystek (NHMFL, Tallahassee), G. Lukovsky, North Carolina State), A. Mackenzie (St Andrews, UK), J. Reiner (Yale), S. Reymond (Lausanne), Z. Schlesinger (UCSC), J. Schmalian (Ames Lab), M. Toney (SSRL).

Recognitions, Honors and Awards (at least partly attributable to support under this FWP or subtask):
MRB: 2002 Elected to the Board of Trustees of the Associated Universities Inc.
IRF: 2003 Sloan Research Fellowship; 2004 Terman Fellowship; 2006 Brown faculty fellow

Personnel Commitments for FY2007 to Nearest +/- 10%:
I. R. Fisher (10%), M. R. Beasley (10%), T. H. Geballe (0%),
J-H. Chu (50%), K. Munakato(10%), P. SanGiorgio (50%) G. Koster (25%), A. Erickson (50%), N. Ru (50%), K. Y. Shin (50%),

Authorized Budget (BA) for FY05, FY06, FY07:
FY05 BA \$550k **FY06 BA** \$650k **FY07 BA** \$650 k

Laboratory Name: XLAM / SSRL, SLAC
B&R Code: KC0202010; KC0202020; KC0202030

FWP and possible subtask under FWP:
FWP: Basic Energy Sciences Materials Sciences (XLAM); Subtask 3: Theory of Condensed Matter Physics, including 1) nano-scale magnetism, 2) spin transport in semiconductors and 3) correlated-electron phase diagrams and 4) interaction of matter with high-intensity X-rays.

FWP Number: SCW0035

Program Scope:
This work explores fundamental physical processes governing the spin transport in semiconductors. It focuses on the physics of spin-orbit coupling and the generation of dissipationless spin current by an electric field. It explores the application of quantum spintronics in semiconductor devices with low power dissipation.

Major Program Achievements (over duration of support):
Achieving long spin life time is one of the most important objectives of spintronics. Spin-orbit coupling could lead to rapid decay of the electron spin. We propose a mechanism by which the spin decay due to spin-orbit coupling could be eliminated, in models with equal Rashba and Dresselhauss coupling constant. We predict that the spin lifetime is infinite at a "magic" wave-vector, giving rise to a Persistent Spin Helix (PSH). Recent experiments by Joe Orenstein's group, performed on materials grown in David Awschalom's group, have quantitatively verified our theoretical prediction. The optical spin grating experiments have indeed revealed enhanced spin life time for a particular grating wave length of the order of one micron. We have performed quantitative theoretical calculations for the line shape of the spin life time, and the results compare well with the experiments.

My group also worked on the quantum spin Hall effect, and its realization in semiconductors. We identified that CdTe/HgTe quantum wells are excellent candidates for the QSH state. This work was published in Science 314, 1757, (2006). The experimental confirmation of our theoretical prediction was published in Science 318, 766, (2007). Working with our colleagues at the Wuerzburg, we identified the quantum phase transition from a conventional insulator state to the quantum spin Hall state as one varies the thickness of the HgTe quantum well. We also observed the residual conductance of 2e2/h, in the nominally insulating regime, which confirms the key prediction of our theory. Furthermore, the experiments have shown that an applied magnetic field drastically reduces the residual conductance, in accordance to our theoretical prediction.

Program Impact: Two "Perspective" Articles in Science Magazine and one "Search and Discovery" Article in Physics Today reporting on our work. Presentation at the annual press conference of the Semiconductor Industry Association.

Interactions:
External—Lawrence Berkeley National Laboratory (working on spin dynamics with Joe Orenstein); University of Wuerzburg (working on the quantum spin Hall effect with Laurens Molenkamp).

Recognitions, Honors and Awards (at least in some part attributable to support under this program):
Shoucheng Zhang – 2007 Guggenheim fellowship award.

Personnel Commitments for FY2007 to Nearest +/- 10%:
Shoucheng Zhang (10%), Xiaoliang Qi (postdoc 100%), R. Laughlin 20%, A. Black-Schaffer (50%), S. Kivelson (5%)

Authorized Budget (BA) for FY05, FY06, FY2007:
FY05 BA $218K **FY06 BA** $354K **FY07 BA** $354K

Laboratory Name: XLAM/SSRL, SLAC
B&R Code: KC0202010

FWP and possible subtask under FWP:
FWP: Basic Energy Sciences Materials Sciences (XLAM); Subtask 4: Using local probes for the study of Nano-scale Phenomena in Complex Materials.

FWP Number: SCW0035

Program Scope:
The objective of this collaborative research program emphasizes the development of new tools and their use for the study of nanoscale ordering phenomena in materials exhibiting novel electronic and magnetic properties, usually correlated electronic systems. The program also emphasizes the creation of a national resource for the study of the development and characterization of novel materials using novel tools, and in training students and other technical personnel in these fields.

Major Program Achievements (in FY2007):
STM Studies: Monolayers formed from diamondoids, diamond molecules derived from individual cages of a diamond crystal, were created and imaged for the first time with STM; electronic structure of single- and double-layer epitaxial graphene investigated along with inelastic tunneling to C-C phonon modes; Observation of the coupling and focusing of molecular vibration modes through a surface two-dimensional electron gas, using molecular manipulation of CO on Cu and inelastic tunneling spectroscopy; observation of bosonic replicas in spectral mapping of Y-BSCCO, and spectral inversion to obtain coupling constants; First atomic resolution imaging of heavy-fermion compound USb_2, investigation of surface ordering and 5f electron screening.
Local Magnetic Measurements: Continued Kerr effect measurements of Sr_2RuO_4 showing no optical power dependence to the time-reversal symmetry breaking signal and linear temperature dependence of that signal near Tc. Studies of Kerr effect in high quality $YBa_2Cu_3O_{6+x}$ crystals showing a finite signal appearing at the pseudogap temperature. The sharpness of the onset of the signal suggests true broken symmetry at the pseudogap. Anomalous magnetic hysteresis pointing to TRSB above room temperature was discovered. Improved understanding of expected magnetic signals from time-reversal-symmetry breaking superconductors such as Sr_2RuO_4. Systematic measurements of the presumed time-reversal-symmetry breaking superconductors Sr_2RuO_4 and $PrOs_4Sb_{12}$ showing strong disagreement with theoretical predictions for the existence of edge currents. Manipulation of individual vortices in high quality $YBa_2Cu_3O_{6+x}$ showing pronounced anisotropy due to the vortex core properties and to nanoscale ordering of oxygen defects on the chains.

Program Impact:
Power of local measurements has been demonstrated to investigate properties of strongly correlated materials. High-resolution STM enabled the study of electronic coupling to excitations not previously possible. Leading role in elucidating the nature of time-reversal-symmetry breaking and chiral superconductors.

Interactions:
At Stanford: I.R. Fisher, M. Greven, M.R. Beasley and T.H. Geballe, S. Kivelson, S. Doniach, S.C. Zhang, N. Melosh, Z.X. Shen, M.M. Fejer and Eun-Ah Kim. Outside Stanford: A. V. Balatsky, M. Hawley, Filip Ronning (Los Alamos NL), R. Liang, D. Bonn, W. Hardy (UBC, Canada), B. A. Jones, C. P. Lutz, A. S. Heinrich, D. M. Eigler (IBM Almaden, USA), E. J. Heller (Harvard, USA), E. Fradkin (UIUC, USA), John Tranquada (BNL, USA), Yoshiteru Maeno, (Kyoto, Japan), John Berlinsky and Catherine Kallin (McMaster, Canada), Kamran Behnia (ESPCI, Paris).

Recognitions, Honors and Awards (at least in some part attributable to support under this program):

Personnel Commitments for FY2007 to Nearest +/- 10%:
A. Kapitulnik (10%), H. Manoharan (10%), K. Moler, (10%), A. Fang (50%), Elizabeth Schemm (50%), G. Zeltzer (15%), L. S. Mattos (20%,), B. K. Foster (10%), C. R. Moon (10%), C. Hicks (50%), O Auslaender, L Luan

Authorized Budget (BA) for FY05, FY06, FY2007:
FY05 BA $431k **FY06 BA $**412k **FY07 BA $**750k

Laboratory Name: XLAM/SSRL, SLAC
B&R Code: KC0202010

FWP and possible subtask under FWP:
FWP: Basic Energy Sciences Materials Sciences (XLAM); Subtask 5: Nano Magnetism

FWP Number: SCW0035

Program Scope:
The goal of this project is the detailed understanding of the processes involved in manipulating the magnetization of a thin nanoscale magnetic element by a spin polarized current. This is accomplished by use of unique time-dependent x-ray imaging techniques with tens of nanometers spatial and tens of picoseconds temporal resolution.

Major Program Achievements (over duration of support):
We have developed and used x-ray imaging techniques with 30 nm spatial and a temporal resolution down to 100 ps for studying the motion of buried magnetic sensor layers of nanoscale (~100-200 nm) dimensions under the influence of spin currents. Ultrafast x-ray motion pictures reveal a fast, sub-nanosecond switching process based on the lateral displacement of a magnetic vortex. The existence of magnetic vortices had previously been observed in large micrometer-sized magnetic structures but their existence in nanometer-sized elements and their role in magnetization switching in such elements had remained unrecognized. The switching mechanism resembles a hurricane that sweeps through the magnetic island, with the vortex core representing the eye of the storm with a circular magnetic "force field" around it. As the vortex enters the magnetic island it bends the magnetization into a curved C state around the vortex core. The "force" on the back side of the vortex core, however, has the opposite direction and as the vortex core leaves the magnetic cell it leaves the magnetization rotated into this new direction. Our first results were reported in Physical Review Letters in 2006. In this paper we described the observation that the vortex switching process may result in metastable final states which are undesirable in technological applications.

More extensive work was performed in 2007. We studied a variety of samples with different sizes to understand whether the vortex switching mechanism remains even as the sample diameter is reduced. The measurements were compared to detailed micomagnetics calculations. From this analysis a surprisingly simple story emerged. As the magnetic island becomes smaller the switching gradually becomes more uniform. At the smallest (< 100nm) diameters the magnetization remains uniform and rotates as a whole. In this regime all spins can be treated as pointing into the same direction, so that they form a giant "macro-spin". This case can also be treated in a vortex model, where the core of the vortex moves not through the island but around it at large distance. At sizes in-between the vortex core moves around the island at closer distances. The work is described in the Ph.D. thesis of John Paul Strachan, available at http://www-ssrl.slac.stanford.edu/stohr/. One publication has been submitted and others are in preparation.

Program impact:
In 2007, the work has resulted in 8 invited talks which were given by Y. Acremann, J. Stohr and John Paul Strachan at international conferences. Publications:
1. Y. Acremann, J. P. Strachan, V. Chembrolu, S. D. Andrews, T. Tyliszczak, J.A. Katine, M. J. Carey, B. M. Clemens, H. C. Siegmann, J. Stöhr, Phys. Rev. Lett. **96**, 217202 (2006)
2. John Paul Strachan, Ph.D. thesis, Department of Applied Physics, Stanford University, June 2007
3.Y. Acremann, V. Chembrolu, J.P. Strachan, T. Tyliszczak, and J. Stöhr, Rev. Sci. Instrum. **78**, 014702 (2007)

Interactions:
J. Katine, M. Carey (Hitachi Global Storage Systems); T. Tyliszczak (Advanced Light Source).
Personnel Commitments for FY2007 to Nearest +/- 10%:
Y. Acremann 40%, A. Tulapurkar 50%, J. P. Strachan 100%, V. Chembrolu 100%, X. Yu 100%
Authorized Budget (BA) for FY05, FY06, FY07:
FY05 BA $350K **FY06 BA** $350K **FY07 BA** $350K

Laboratory Name: XLAM/SSRL, SLAC
B&R Code: KC0202010

FWP and possible subtask under FWP:
FWP: Basic Energy Sciences Materials Sciences (XLAM); Subtask 7: Catalysis

FWP Number: SCW0040

Program Scope:
The main focus of this research program is to provide an understanding of the mechanistic pathway of the oxygen reduction reaction (ORR) and to design new low Pt containing catalyst by optimizing the geometric and electronic structure of catalyst materials. An essential aspect of this project is to develop synchrotron radiation based x-ray diffraction (XRD) and spectroscopy methods that allow in-situ probing of the intermediates in the catalytic cathode process where both species identification, geometric and electronic structure properties is fully characterized. In parallel to the fundamental synchrotron work, theory-guided combinatorial synthesis and high throughput electrochemical screening methodologies for fuel cell cathode catalysts will be developed and applied in order to link mechanistic hypotheses and catalyst testing under realistic conditions in high dimensional compositional and process parameter spaces.

Major Program Achievements (over duration of support)
-Demonstration of a new class of catalysts for the ORR reaction with a 4-6 times higher specific activity compared to Pt at 900mV using both rotating disk electrode and membrane assembly measurements. The specific activity is over the 2010 DOE performance goal. The catalyst consists of dealloyed Pt-Cu nanoparticles. De-alloying of very Cu rich (>= 75 atomic % Cu) Pt-Cu alloy particle precursors results in nanoparticles consisting of a Pt alloy core surrounded by a multi-layer Pt rich particle shell.
-Water will be present during the ORR reaction and experimentally we have investigated its important role in the reaction. We have found that the activation of oxygen to the adsorbed atomic phase occurs readily in the dry region, but not in the humid region process. In this case, water is acting to poison the active sites on the catalyst. On the contrary, water is found to promote the reduction of adsorbed atomic oxygen in the formation of a water-hydroxyl surface complex.
- We have investigated a potential pathway of peroxide formation on the catalyst surface and found that adsorbed hydrogen plays an important role. The presence of hydrogen on the catalyst surface promotes a transition from molecular oxygen laying flat to tilted-up molecules with respect to the surface plane. We propose that the peroxide formation reaction channel is stimulated by this transition of the oxygen orientation on the surface. Additionally, our preliminary studies further indicate that the tilted-up geometry sterically hinders the activation of oxygen that leads to dissociation on the catalyst surface.

Program Impact
This program has the potential to have a large impact on the future Hydrogen Fuel Initiative. Fuel cells are currently considered as the most promising power generation technology for a sustainable energy infrastructure.

Interactions:
A. Nilsson, M. Toney, H. Ogasawara, L. Leisch, L. Å, Näslund, T. Anniyev, J. McNoughton, D. Friebel (SSRL), P. Strasser, S. Koh, P. Mani (University of Houston), Lars Pettersson, Michael Odelius, (Stockholm University, Sweden), J. Greeley (ANL) and J. Norskov (Danish Technical University, Denmark).

Personnel Commitments for FY2007 to Nearest +/- 10%:
A. Nilsson 10%, H. Ogasawara 50 %, M. Toney 10%, J. Leisch 100%, J. McNaughton, 50%, D. Friebel 20% (SSRL), P. Strasser 10%, S. Koh 100%, P. Mani 100% (UH)

Authorized Budget (BA) for FY06, FY07, FY08:
FY05 BA $600K **FY06 BA** $600K **FY07 BA** $600K

Laboratory Name: XLAM/SSRL, SLAC
B&R Code: KC0203010

FWP and possible subtask under FWP:
FWP: Basic Energy Sciences Materials Sciences (XLAM); Subtask 6: Behavior of Charges, Excitons and Plasmons at Organic/Inorganic Interfaces

FWP Number: SCW0034

Program Scope:
A collective approach using X-ray diffraction, surface plasmon spectroscopy, and plasmon engineering is applied to the problem of exciton and charge transport at organic-inorganic interfaces. Electromagnetic finite difference time domain codes were developed to simulate time-dependent field behavior near metallic structures and plasmonic resonators. New fabrication techniques enable soft deposition of electrodes onto organic monolayers.

Major Program Achievements (over duration of support):
Discovered that the key to obtaining high charge carrier mobility in polymer field effect transistors is nucleating crystals from the dielectric surface so that the insulating side chains all point in the same direction and do not impede charge hopping from one crystal to another. Improved exciton harvesting in organic solar cells by a factor of four using long range resonance energy transfer. The Brongersma group demonstrated enhanced control over the radiative decay processes of small molecules in patterned, metallic nanocavities. Electromagnetic simulation tools were developed to analyze changes in both the directionality of the emission and the radiative decay rate of the emitters. Simulations and experiments have demonstrated an ability to realize cavities exhibiting omni-directional and highly directional emission. The Brongersma and Melosh groups also had an intensive collaboration on active plasmonic switches and the development of plasmon-based optical spectroscopies. These spectroscopies have revealed that only ~1 in 1000 rotaxane molecules is active within molecular electronic junctions, suggesting each molecule has a high on/off ratio, but poor switching efficiency.

Program impact:
Enabling more efficient solar cells and solid state lighting and characterization of [2]-rotaxane molecular switches.

Interactions:
Internal- SSRL, Shen Group
External- UC Berkeley and the Molecular Foundry, UCLA, Fraser Stoddart group

Recognitions, Honors and Awards (at least partly attributable to support under this FWP or subtask):
McGehee: Mohr Davidow Venture Innovators Award (2007)
McGehee: Materials Research Society Outstanding Young Investigator Award (2007)
Brongersma: Gores award for excellence in teaching; Stanford's highest teaching honor.

Personnel Commitments for FY2007 to Nearest +/- 10%:
Brongersma (8 %), McGehee (8 %), Melosh (8 %), J Fabbri 50%, C Peters 50%, J Liu 50%,
M Topinka 35%

Authorized Budget (BA) for FY05, FY06, FY07:
FY05 BA $275K **FY06 BA** $275K **FY07 BA** $275K