1. DOE award #DE-SC0001480, NIST (Institution)

2. Project Title and name of the PI: SISGR: Ultrafast Magnetism Dynamics Measure Using Tabletop Ultrafast EUV Sources" Thomas Silva

3. Period covered by report: September 15, 2009 – May 10, 2011

4. Brief description of accomplishments

Magnetism at nanometer scale lengths on ultrafast timescales is not well understood either theoretically or experimentally, since no complete microscopic model of how spins, electrons, photons and phonons interact currently exists. Moroever, this topic is also directly relevant to industry, since individual bits stored on a hard disk are already of sub-20nm dimension, and advances in storage capacity and energy efficiency depend on further increases in magnetic switching speed and storage density. Until recently, measuring magnetic material dynamics use either ultrafast lasers and visible-wavelength light, or x-rays from large-scale synchrotrons. The former gives short pulses (~ 50 fs), enabling studies in the ultrafast regime but with low spatial resolution. X-rays give high spatial resolution and high contrast at the elemental absorption edges of the ferromagnetic materials; however, the time resolution is too low to see the fastest dynamics involved in domain switching. New light sources including EUV light from high-harmonic generation (HHG) or x-ray free electron lasers (XFEL) facilities have the potential to combine nm spatial resolution with ~< 10 fs time resolution. Synchrotron and XFEL x-ray studies typically make use of magnetic contrast near the L-edge of the elements at ~ 700 eV photon energy. However, recent work has shown that one can also obtain magnetic contrast suitable for imaging at the M-edges of magnetic materials, at photon energies of 55-65 eV that *are* accessible using Ti:sapphire laser-driven HHG light.

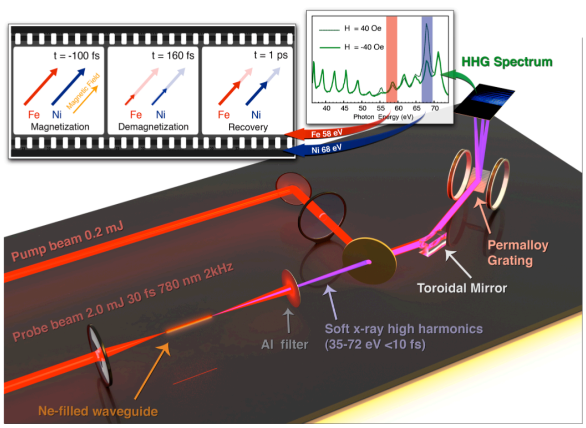

Fig. 1. Schematic of the experiment. Ultrafast soft XUV pulses (purple) are reflected from a Permalloy grating sample, which spatially separates the harmonics to form a spectrum on a CCD camera. In the T-MOKE geometry, the reflected HHG intensity at the Fe and Ni M-shell absorption edges (red and blue) depends on the magnetization transverse to the optical plane of incidence that is periodically reversed by transverse-mounted Helmholtz coils. Exciting the sample with an infrared laser pulse (red) causes the material to demagnetize on femtosecond timescales.

In our work to date, we made two significant advances. First we demonstrated element-selective demagnetization dynamics for the first time,[1] with a record time resolution for x-ray probing of 55 fs (see Figs. 1 - 3). Second, in new work, we were able to probe the timescale of the exchange interaction in magnetic materials, also for the first time. Our measurements were made using the transverse magneto-optic Kerr effect (T-MOKE) geometry, since the reflectivity of a magnetic material changes with the direction of the magnetization vector of a surface. In our experiment, we periodically reversed the magnetization direction of a grating structure made of Permalloy (Ni80Fe20) using an external magnetic field (Fig. 1). To achieve maximum contrast, we used HHG light spanning the M-shell (3p) absorption edges of Fe and Ni. Our characterization of the static magnetization of a Permalloy sample shows high magnetic asymmetry at photon energies just above and below the absorption edges at 55 eV and 65 eV, respectively. This result (Fig. 2) is in excellent agreement with measurements done on the same using a synchrotron source.

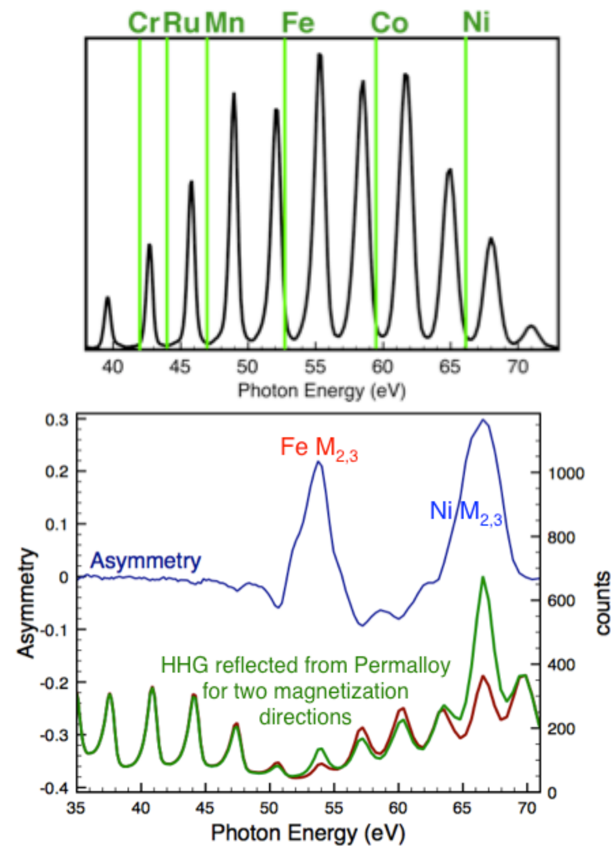

Fig. 2. (top) Experimental high harmonic emission (HHG) (black line) in the 40 – 70 eV photon energy region spanning the M absorption edges of several magnetic materials (green lines). (bottom) HHG spectrum reflected from the sample at an angle of 45 deg, for two different magnetization directions. The strong magnetic asymmetry signal for the Fe and Ni M-edges is also plotted. The asymmetry signal spans a few eV.

Figure 3 plots our data for ultrafast demagnetization. The Permalloy sample is demagnetized using an ultrafast infrared pump-beam, and the magnitude of the magnetization is monitored as a function of time-delay between the laser pump pulse and the EUV probe pulse. Our initial experiments (Fig. 3(top)) measured the demagnetization dynamics the alloy Permalloy (Ni80Fe20), in which the two transition metal species are strongly exchange coupled and, therefore, the demagnetization timescales were the same for Ni and Fe (within our initial measurement accuracy).

More recently, by significantly improving our time resolution to 10 fs, we experimentally answered the fundamental question of whether the magnetization dynamics of the individual elements Fe and Ni in Permalloy can differ on ultrafast time scales.[2] This is a very important fundamental question that has not been addressed either theoretically or experimentally to date, the answer to which reveals how the exchange interaction can control ultrafast magnetic dynamics. To answer this question, we rapidly excited Permalloy with an ultrafast laser pulse and probed the element-specific demagnetization dynamics using 10 fs high harmonic pulses. The extreme time resolution of our experiment allows us to observe for the first time that the magnetization dynamics of the Fe and Ni are transiently delayed with respect to each other by 12 ± 10 fs in Permalloy and 66 ± 10 fs in Cu-diluted Permalloy ((Ni0.8Fe0.2)1-xCux). We ascribe this transient decoupling in the magnetic behavior to the finite strength of the exchange interaction between Fe and Ni atoms in the material. Exchange energies of transition metal alloys are in the 10-100 meV range, yielding characteristic exchange times in the femtosecond range which corresponds to finite spin-flip scattering times of 10 – 100 fs. Our findings provide crucial new answers for open questions in femtosecond magnetization dynamics in the case of metallic, multi-species, exchange-coupled systems.

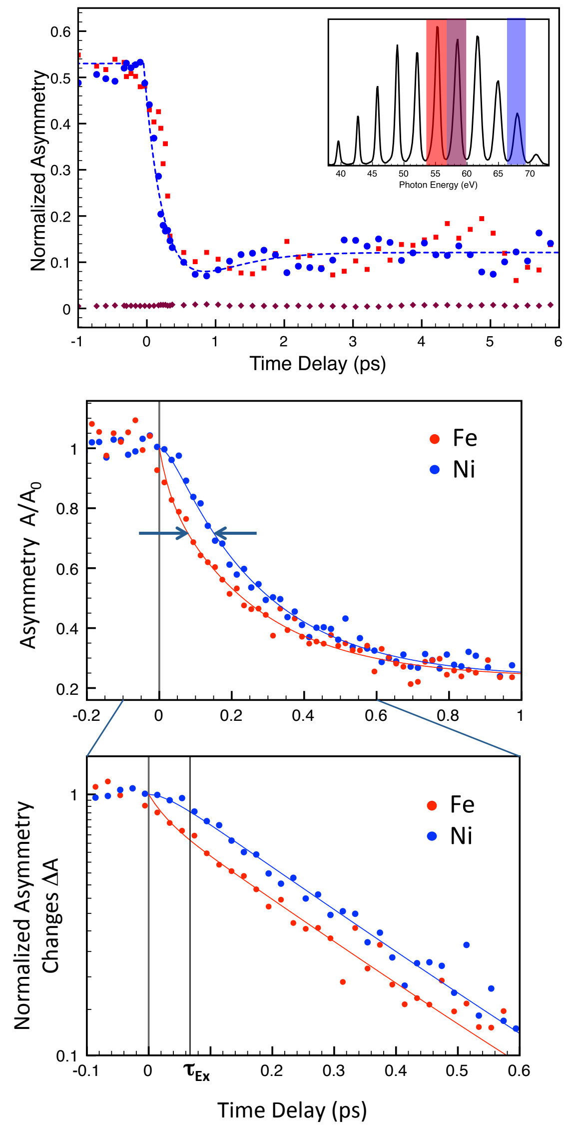

Fig. 3. (top) Initial time-resolved measurement of the femtosecond laser-pulse induced demagnetization of Permalloy with 55 fs time resolution. (center and bottom). Higher (10fs) time resolution data for ultrafast demagnetization of Fe (red dots) and Ni (blue dots) in Cu-doped Permalloy. The data is shown as a function of magnetic asymmetry on linear and log scales. A simple exponential decay fit yields the effective demagnetization constant, which results in different values for Fe and Ni. However, the log scale plot reveals that there is actually a lag between when the demagnetization of Ni and Fe, of ≈ 66 fs. After this characteristic delay time - corresponding to the exchange interaction energy - both elements demagnetize with the same decay constant.

Future Plans

In the short term, we will investigate magnetization dynamics in ferro- and antiferromagnetic coupled multilayer devices and Heusler-alloys. We are also making nanoscale heat flow measurements and eventually imaging of magnetic nanostructures in collaboration with industry. Looking to the future, it will be possible to extend these measurements to the L absorption edges of magnetic media, due to recent advances in generating bright, coherent, x-rays from femtosecond lasers in the keV region of the spectrum. In recent exciting work in collaboration with Andrius Baltuska in Vienna, we demonstrated that bright coherent high harmonic x-rays from femtosecond lasers can extend to photon energies >1.6 keV (wavelengths <7.8 Å) using mid-infrared 4 µm driving lasers at 10 Hz. Image contrast, resolution, and transparency of many materials are all greatly improved at soft x-ray L-edge energies. Thus, once we have developed a kHz version of the same driving laser at JILA, imaging of buried magnetic structures, domain interactions, strongly coupled nanolayers, and spin dynamics at the L edges will all be accessible with femtosecond and possibly even sub-femtosecond time resolution.

5. List of papers

1. C. La-O-Vorakiat, M. Siemens, M. Murnane, H. Kapteyn, S. Mathias, M. Aeschlimann, P. Grychtol, R. Adam, J. Shaw, H. Nembach, T. Silva, *Ultrafast Demagnetization Dynamics at the M Edges of Magnetic Elements Observed Using a Tabletop High-Harmonic Soft X-Ray Source*, Phys. Rev. Lett. **103**, 257402 (2009).

2. S. Mathias, C. LaOVorakiat, P. Grychtol, J. Shaw, R. Adam, H.T. Nembach, M. Siemens, S. Eich, C.M. Schneider, T.J. Silva, M.A. Aeschlimann, H.C. Kapteyn and M.M. Murnane, "Probing the timescale of the exchange interaction in a ferromagnetic alloy", submitted (2011).

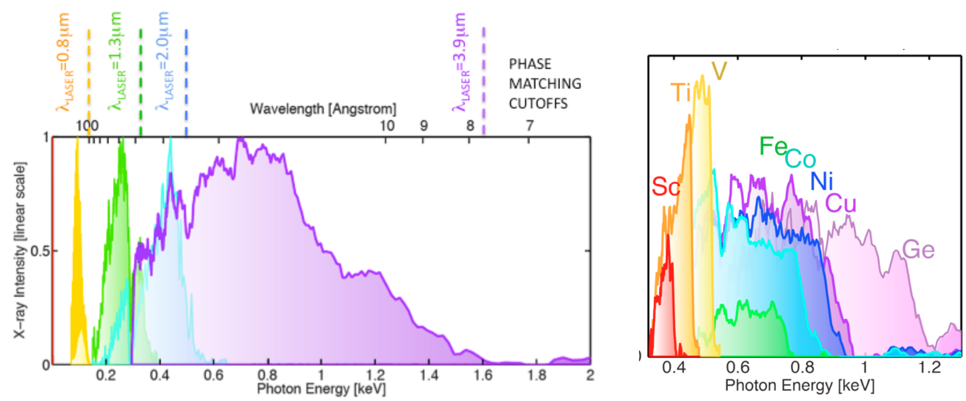

Fig. 4. (left) HHG spectra emitted under full phase matching conditions as a function of driving laser wavelength. Yellow – 0.8μm, green – 1.3μm, blue – 2μm and purple – 3.9μm. Using a 3.9μm driving laser, a broad coherent x-ray continuum spans from the EUV to the keV spectral regions. In the case of the 3.9μm (purple) spectra, the low energy EUV region is not plotted because the x-ray spectrometer grating has a finite range in efficiencies. However, the bright emission also covers the lower photon energy region. (right) Measured L-absorption edges of metals, including magnetic materials, throughout the soft X-ray region, demonstrating a useful probe for ultrafast element-selective spectroscopies.

6. A list of people working on the project

Name			Affiliation	Support
Chan La-o-vorakiat	Graduate Student	Physics	CU	This DOE project
Stefan Mathias	Postdoctoral Fellow	Materials	CU	This DOE project and postdoctoral fellowship
Henry Kapteyn	Faculty	Physics/ECE	CU	NA
Margaret Murnane	Faculty	Physics/ECE	CU	NA
Thomas Silva	Senior Scientist Faculty	EM Division	NIST	This DOE support and NIST base funding
Justin Shaw	Staff Scientist	Physics/Materials Science	NIST	NIST base funding
Hans Nembach	Postdoctoral Fellow	Physics	NIST	This DOE support
New graduate student Emrah Turgut	Graduate Student	Physics	CU	This DOE project starting May 2010
Martin Aeschlimann	Senior Scientist Faculty	Nanoscience	Germany	NA
Patrik Grychtol	Graduate Student Visitor	Physics	Germany	NA

7. Planned activities for next year

- Ultrafast demagnetization dynamics in other materials

In the short term, we will investigate magnetization dynamics in ferro- and antiferromagnetic coupled multilayer devices and Heusler-alloys. We are also making nanoscale heat flow measurements and eventually imaging of magnetic nanostructures in collaboration with industry. Looking to the future, it will be possible to extend these measurements to the L absorption edges of magnetic media, due to recent advances in generating bright, coherent, x-rays from femtosecond lasers in the keV region of the spectrum.

- Attosecond magnetization dynamics

We will extend the time-resolution to the attosecond regime. This will enable us to investigate coherent magnetization dynamics and the direct interaction between the laser electric field and the spin system in the material. In this project, we are collaborating with Guo-Ping Zhang from Indiana State University, who will support us with theoretical calculations.

8. An updated list of other support

No new effort has been funded since this grant was awarded. Thus, the previous effort table is accurate and can be sent again if needed.

9. Cost status

At CU Boulder, a new chamber was designed that used the budgeted equipment funds before the end of the grant period. At NIST, salaries are pre-paid and thus the salary funds (Dr. Hans Nembach, postdoc) were fully used before the end of the grant period.

	Budget for Current Year	**Expended or incumbered funds to end of period**	**Funds to be spent before end of period**	**Anticipated carry over**
CU	$269,980	$173,253	$76,727	$20,000
NIST	$134,961	$775	$134,186	$0