



AFRL-AFOSR-VA-TR-2023-0091

Spatiotemporal imaging of magnetization dynamics at the nanoscale

**Fuchs, Gregory
CORNELL UNIVERSITY
341 PINE TREE RD
ITHACA, NY, 14850
USA**

**10/19/2022
Final Technical Report**

DISTRIBUTION A: Distribution approved for public release.

Air Force Research Laboratory
Air Force Office of Scientific Research
Arlington, Virginia 22203
Air Force Materiel Command

DISTRIBUTION A: Distribution approved for public release.

REPORT DOCUMENTATION PAGE

PLEASE DO NOT RETURN YOUR FORM TO THE ABOVE ORGANIZATION.

| | | | |
|--|---|--|---|
| 1. REPORT DATE 20221019 | 2. REPORT TYPE Final | 3. DATES COVERED | |
| | | START DATE 20140901 | END DATE 20200831 |
| 4. TITLE AND SUBTITLE Spatiotemporal imaging of magnetization dynamics at the nanoscale | | | |
| 5a. CONTRACT NUMBER | 5b. GRANT NUMBER FA9550-14-1-0243 | 5c. PROGRAM ELEMENT NUMBER 61102F | |
| 5d. PROJECT NUMBER | 5e. TASK NUMBER | 5f. WORK UNIT NUMBER | |
| 6. AUTHOR(S) Gregory Fuchs | | | |
| 7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) CORNELL UNIVERSITY 341 PINE TREE RD ITHACA, NY 14850 USA | | | 8. PERFORMING ORGANIZATION REPORT NUMBER |
| 9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES) Air Force Office of Scientific Research 875 N. Randolph St. Room 3112 Arlington, VA 22203 | | 10. SPONSOR/MONITOR'S ACRONYM(S) AFRL/AFOSR RTB1 | 11. SPONSOR/MONITOR'S REPORT NUMBER(S) AFRL-AFOSR-VA-TR-2023-0091 |
| 12. DISTRIBUTION/AVAILABILITY STATEMENT A Distribution Unlimited: PB Public Release | | | |
| 13. SUPPLEMENTARY NOTES | | | |
| 14. ABSTRACT In this AFOSR project, we develop time-resolved magneto-thermal microscopy and apply it to materials and devices relevant to emerging magnetic memory and logic technologies. This technique is a spatiotemporal probe with sub 100 ps temporal resolution and high sensitivity to magnetic orientation. Because it uses heat rather than light as excitation source, it not limited by optical diffraction, and can obtain spatial resolution below 100 nm using a scanning probe without sacrificing temporal resolution. | | | |
| 15. SUBJECT TERMS | | | |
| 16. SECURITY CLASSIFICATION OF: | | 17. LIMITATION OF ABSTRACT | 18. NUMBER OF PAGES |
| a. REPORT U | b. ABSTRACT U | c. THIS PAGE U | 6 |
| 19a. NAME OF RESPONSIBLE PERSON ALI SAYIR | | 19b. PHONE NUMBER (Include area code) 426-7236 | |

Final Report, AFOSR grant #FA9550-14-1-0243, "Spatiotemporal imaging of magnetization dynamics at the nanoscale: Breaking the Diffraction Limit."

In our first work, we demonstrated a new method of spatiotemporal magnetic microscopy based on the time resolved anomalous Nernst effect (TRANE). Our concept is that by using the interaction with the local magnetization with heat rather than light, we can potentially image magnetization with spatial resolution below the optical diffraction limit because heating is not fundamentally limited by optical diffraction. To see if this concept is valid, there are essentially two crucial questions. First, does thermal diffusion limit the spatial resolution such that we could never obtain 50 nm spatial resolution? Second, are thermal relaxation time scales too long to image ferromagnetic resonance phenomena (picoseconds)? In this work, using a combination of experimental measurements and theoretical modeling, we show that the answer to both of these questions is 'no'. We find that when we use picosecond duration light pulses to heat our sample, we are able to obtain spatial resolution comparable with the size of the thermal source. Furthermore, using numerical modeling, we show that if we have a smaller thermal source, the spatial resolution will improve at least down to the 50 nm level. Secondly, we show through direct measurements of thermal lifetimes and measurements of ferromagnetic resonance with heat pulses, that the thermal relaxation times are in the 10 to 30 ps range. The full manuscript is J. M. Bartell, D. H. Ngai, Z. Leng, and G. D. Fuchs, "Table-top Measurement of Local Magnetization Dynamics Using Picosecond Thermal Gradients: Toward Nanoscale Magnetic Imaging." *Nature Communications* **6**, [8460 \(2015\)](#).

In our second work, we demonstrate the application of TRANE microscopy and its extensions to studying ferromagnetic resonance in magnetic bilayer samples where we excite directly with an applied microwave current. We show that we can simultaneously image both the microwave current and the ferromagnetic resonance in a phase sensitive measurement, enabling us to establish their phase relationship from experimental measurement. Additionally, we find that when we image a sample with a simple, uniform current channel, the microwave current amplitude and phase are uniform. If, on the other hand, we image a sample with a slightly more complex shape – a "Hall" cross – we find substantial shifts in the amplitude and phase of the microwave current. Correspondingly, we find shifts in the magnetic precession amplitude and phase as a function of position within the sample. This kind of local stimulus – response imaging can establish directly the relationship between a driving current and the magnetic response, which is a unique capability to understanding spin-orbit torques including the spin Hall effect and torques from topological insulators. The full manuscript is Feng Guo, J. M. Bartell, D. H. Ngai, and G. D. Fuchs, "Phase-sensitive imaging of ferromagnetic resonance using ultrafast heat pulses." *Phys. Rev. Applied* **4**, 044004 (2015)

Building on that work, we applied phase-sensitive TRANE microscopy to understanding the ferromagnetic resonance response in spin Hall effect multilayers, where spin-transfer torques along with magnetic fields drive the magnetic resonance. By performing phase-sensitive imaging, we developed a method to quantitatively measure the spin-torque vector with phase sensitive FMR imaging. When applied to Pt/Hf/FeCoB bilayers, we observed a significant phase (total torque angle) variation across the sample. This is important because an electrical version of FMR that assumes uniform phase precession (but is blind to its variations) is widely used to quantify these torque vectors as a way to measure the spin Hall efficiency. We show that in our devices, there is a roughly 30% correction to the spin Hall efficiency that one obtains from these measurements. This work was published this year: Feng

Guo, Jason M. Bartell, and Gregory D. Fuchs "Ferromagnetic resonance phase imaging in spin Hall multilayers." *Phys. Rev. B* **93**, 144415 (2016).

We are also using these materials to study spin Hall torques in magnetic insulator systems such as Pt/Yttrium Iron Garnet (YIG) bilayers. Because current doesn't flow in the YIG, this is an even more efficient system for manipulating magnetization with current-induced torques. TRANE microscopy works in these systems because the thermal gradient in the YIG produces a spin current in the Pt via the longitudinal spin Seebeck effect, which is transduced into a measurable voltage by the inverse spin Hall effect. I note that TRANE microscopy is one of the very few techniques that can measure the magnetic moment of micro-fabricated YIG devices. In Pt/YIG devices we have recently demonstrated

- In ultra-thin YIG devices, the LSSE is ultra-fast, operating on picosecond time-scales
- Imaging of YIG magnetization with $0.3^\circ/\text{Hz}^{1/2}$ sensitivity
- Spatial resolution of $0.6\ \mu\text{m}$.
- Temporal resolution of $< 100\ \text{ps}$.
- Ferromagnetic resonance imaging of YIG with phase sensitivity.

This work is published in Bartell *et al.*, *Phys. Rev. Appl.* **7**, 044004 (2017).

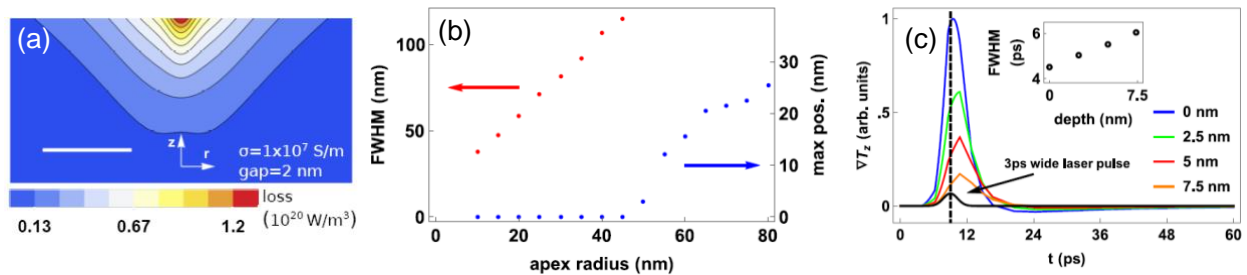


Figure 1. Characteristics of plasmonic thermal excitations from a sharp gold tip. (a) Point-spread function for a sample conductivity of $10^7\ \text{S/m}$ and a 2 nm gap between the tip apex and sample surface. Horizontal scale bar indicates 50 nm, sample thickness is 10 nm. (b) The FWHM of the thermal excitation as a function of tip radius. For radii above 45 nm, the point-spread function becomes toroidal, and thus we also plot the peak radius of the thermal excitation. (c) Spatiotemporal response of the thermal excitation for typical sample parameters. Figures modified from Ref. {Formatting Citation}.

We have also been pushing toward the ultimate goal of our project – sub-diffraction spatiotemporal magnetic imaging based on near-field heating. The challenge is to create a scanned, picosecond thermal excitation of the sample with nanoscale area. The concept of localized, near-field heating was previously studied in the context of heat-assisted magnetic recording, but not yet adapted to applications in nanoscale magnetic microscopy. First we studied the situation theoretically using finite element modeling. Figure 2 summarizes some of the basic results in which we theoretically clarified experimental questions and refined our understanding of how to reach our targets. This work validated our goal - we expect a thermal gradient with a 50 nm full-width at half maximum from a 15 nm gold tip. We also see that plasmonic nano-focusing retains the full temporal resolution of ordinary magneto-thermal microscopy: for a 3 ps wide exciting laser pulse, the thermal gradient temporal FWHM is ~ 10 ps for a typical sample. Additionally, we have carefully understood how these values interact with the conductivity of the target sample material, and suggested that samples could be made uniform by

coating them in < 10 nm of Pt or Ru. This theoretical analysis was recently accepted in APL photonics [Karsch *et al.*, APL Photonics **2**, 086103 (2017)].

Simultaneously achieving picosecond temporal resolution and nanometer scale spatial resolution is a key goal in magnetic microscopy because it enables study of active and driven magnetic textures at length scales that are interesting for fundamental science and technology. A key example is the imaging of individual skyrmion dynamics, one aim of this project. This period we made key advances in establishing scanning near-field magneto-thermal microscopy as an important tool in that arena. We characterized the nanoscale spatial resolution of the instrument by current imaging, and demonstrated its picosecond temporal resolution enabling phase-sensitive dynamics measurements. The operating principle of our microscope for magnetic metals is based on the Anomalous Nernst effect.

There are two modalities of the instrument. The laser-induced temperature gradient ∇T enables magnetization imaging (the first term), through either the anomalous Nernst effect (ANE) or longitudinal spin Seebeck effect (LSSE) for ferromagnetic metals, ferrimagnetic insulators and antiferromagnets. The laser-induced temperature increase ΔT (the second term) additionally enables imaging of an applied current density, from DC to microwave frequencies, which allows us to directly probe both a stimulus and its response.

Spatial resolution by current imaging:

We previously characterized the spatial resolution by imaging a magnetic domain wall in a CoFeB sample. However, we found that the characteristic length over which we could detect contrast was limited by the domain wall width rather than the instrument resolution, given to the low magnetic anisotropy of the material. Since extensive materials engineering are required to tune a domain wall to the right width, we instead turn to current imaging mode as a test of the instrument resolution, and lithography pattern a sample with narrow constrictions that confine the current density in a nanoscale region.

The sample is a $\text{Ni}_{80}\text{Fe}_{20}$ (5 nm)/Ru (2 nm) film, fabricated using e-beam lithography into a 2 μm -diameter disk with two 150 nm wide necks (Fig. 3(a) inset). Fig. 3(a) and (b) show topography and near-field current density images taken with the near-field scanning probe, at an applied current of -0.03 mA. We see that the current density is indeed concentrated at the neck. By measuring line scans through two necks (Fig. 3(a) inset), we compare signals collected using focused light (Fig. 3(c)) and scanning probe near-field microscopy (Fig. 3(d)). In Fig. 3(c), we measure this sample in our highest-resolution focused light setup³ with numerical aperture of 0.9. We extract a FWHM resolution of 740 nm which is consistent with the setup resolution³. In Fig. 3(d), the scanning near-field data has higher resolution than the far-field data. We quantitatively extract spatial resolution from Fig. 2(d), based on a sharp feature in the line scan (the right-side peak). We simulate the current density distribution around the neck using COMSOL and model the instrument response by convolving it with a Gaussian point spread function. We fit the simulated result to our data and extract a Gaussian width of $\delta = 74$ nm (corresponding to a FWHM of 165 nm). This resolution is 1/4

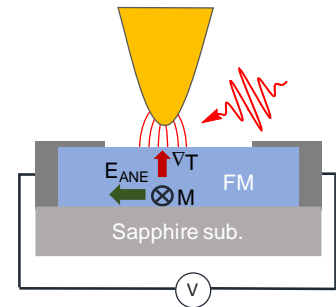


Figure 2. Schematic showing the operating principle of scanning near-field magneto-thermal microscopy.

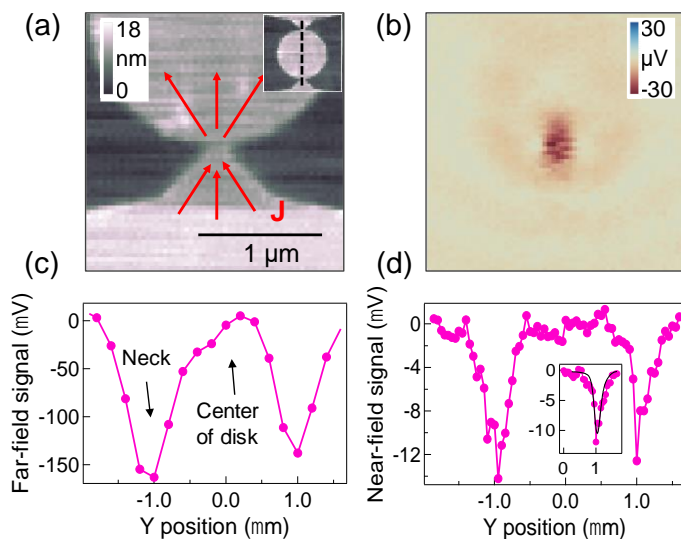


Figure 3. Current imaging and spatial resolution. (a) Topography and (b) current density images. Line cuts of (c) far-field and (d) near-field signals for resolution comparison. (d) inset shows the simulated fit to the data.

of our focused light magneto-thermal microscopy resolution with highest NA at the same wavelength, which is consistent with the sub-diffraction resolution of near-field microscopy. We note that this 100 nm level spatial resolution is only an upper bound for magnetization imaging, because (1) magnetic resolution (determined by thermal gradient ∇T) is higher than current resolution (determined by temperature increase ΔT); (2) the spatial resolution depends on the tip radius, which we estimate at 65 nm when this data was acquired. We expect to have improved spatial resolution by optimizing the tips for better endurance. This work is published in: C. Zhang, J. M. Bartell, J. C. Karsch, I. Gray, and G. D. Fuchs,

“Nanoscale Magnetization and Current Imaging Using Time-Resolved Scanning-Probe Magnetothermal Microscopy,” *Nano Letters* **21**, 4966 (2021).

List of Publications:

1. J. M. Bartell, D. H. Ngai, Z. Leng, and G. D. Fuchs, “Towards a table-top microscope for nanoscale magnetic imaging using picosecond thermal gradients,” *Nature Communications* **6**, 8460 (2015).
2. F. Guo, J. M. Bartell, D. H. Ngai, and G. D. Fuchs, “Phase-sensitive imaging of ferromagnetic resonance using ultrafast heat pulses,” *Physical Review Applied* **4**, 044004 (2015).
3. F. Guo, J. M. Bartell, and G. D. Fuchs, “Ferromagnetic resonance phase imaging in spin Hall multilayers,” *Physical Review B* **93**, 144415 (2016).
4. J. M. Bartell, C. L. Jermain, S. V. Aradhya, J. T. Brangham, F. Yang, D. C. Ralph, and G. D. Fuchs, “Imaging Magnetization Structure and Dynamics in Ultrathin $Y_3Fe_5O_{12}$ / Pt Bilayers with High Sensitivity Using the Time-Resolved Longitudinal Spin Seebeck Effect,” *Physical Review Applied* **7**, 044004 (2017).
5. J. Bartell, PhD Thesis, TIME-RESOLVED MAGNETIC MICROSCOPY USING NEAR- AND FAR-FIELD PICOSECOND HEATING, Cornell University (2018).
6. J. C. Karsch, J. M. Bartell, and G. D. Fuchs, “Near-field coupling of gold plasmonic antennas for sub-100 nm magneto-thermal microscopy,” *Apl Photonics* **2** (2017).
7. A. B. Mei, I. Gray, Y. Tang, J. Schubert, D. Werder, J. Bartell, D. C. Ralph, G. D. Fuchs, and D. G. Schlom, “Local Photothermal Control of Phase Transitions for On-Demand Room-Temperature Rewritable Magnetic Patterning,” *Advanced Materials* **32**, 2001080 (2020).
8. C. Zhang, J. M. Bartell, J. C. Karsch, I. Gray, and G. D. Fuchs, “Nanoscale Magnetization and Current Imaging Using Time-Resolved Scanning-Probe Magnetothermal Microscopy,” *Nano Letters* **21**, 4966 (2021)

Invited presentations related to supported research:

1. "Magneto-thermal Microscopy and Single-spin Microscopy of Complex Magnetic Materials," Spin Caloritronics, UIUC, 2022
2. "Imaging complex magnetic materials using single spins and heat," Petaspin seminar, IEEE Magnetics society Italy chapter (virtual) 2022
3. "Magneto-thermal and Single-spin Microscopy of Magnetic Quantum Materials," NSF Workshop on Emerging Opportunities at the Intersection of Quantum and Thermal Sciences, 2021
4. "Magneto-thermal microscopy of spin-torque switching and uncompensated moments in antiferromagnetic materials," Spintronics XIII, San Diego, CA 2020 [Cancelled due to COVID-19]
5. "Magneto-thermal microscopy of spin-torque switching and uncompensated moments in antiferromagnetic materials" Physics and Applications of Spin Phenomena in Solids (PASPS) X1, State College, PA 2020 [Cancelled due to COVID-19]
6. "Magneto-thermal microscopy of spin-torque switching and uncompensated moments in antiferromagnetic materials," Spin Caloritronics XI, University of Illinois, Champaign-Urbana, 2020 [Postponed until 2021 due to COVID-19]
7. "Magneto-thermal microscopy of spin-torque switching and uncompensated moments in antiferromagnetic materials," Online Spintronics Seminar Series, 2020
8. "Magneto-thermal microscopy of spin-torque switching and uncompensated moments in antiferromagnetic materials," Workshop on Quantum Magnonics, International Institute of Physics, Natal, Brazil 2020 [Postponed due to COVID-19]
9. "Magneto-thermal microscopy of spin-torque switching and uncompensated moments in antiferromagnetic materials," Intermag 2020, Montreal, CA, 2020 [Cancelled due to COVID-19]
10. "Time-resolved magneto-thermal microscopy: high-resolution dynamic imaging of magnetic materials using picosecond heat pulses", Spring Meeting of the MRS, Phoenix, AZ 2019
11. "A New Kind of Magnetic Microscope: Using Ultrafast Heat Pulses to Image Spin-Orbit Torques and Dynamics in Ferromagnetic and Antiferromagnetic Devices," Condensed Matter Physics Seminar, Texas A&M University, College Station, TX 2019
12. "Time-resolved magneto-thermal microscopy: high-resolution dynamic imaging of magnetic materials ranging from ferromagnetic metals to antiferromagnetic insulators," Spin Caloritronics IX, Columbus, OH 2018
13. "Probing the magnetic dynamics of B20 chiral materials and spin-orbit heterostructures using time-resolved magneto-thermal microscopy," Seminar, Ernst Ruska-Centre for Microscopy and Spectroscopy with Electrons, Forschungszentrum, Jülich, Germany 2018
14. "Time-resolved magneto-thermal microscopy: dynamic imaging of magnetic metals and magnetic insulators in devices," Laboratory for Surface Modification/Institute for Advanced Materials, Devices, and Nanotechnology Seminar, Rutgers, New Brunswick, NJ 2018
15. "Time-resolved magneto-thermal microscopy: dynamic imaging of magnetic metals and magnetic insulators in devices," Annual Conference on Magnetism and Magnetic Materials, Pittsburg, PA 2017
16. "Nanoscale magnetic imaging using picosecond thermal gradients," Intel Metrology Seminar, Intel Corporation, Hillsboro, OR 2017

17. "Nanoscale magnetic imaging using picosecond thermal gradients," March Meeting of the APS, New Orleans, LA 2017