Magnetic Resonance Force Detection and Imaging of Electron Spins in a Laminate Thin Film

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Primary CNF Tools Used: SC 4500 Combination Thermal/E-gun Evaporation System, Zeiss Supra SEM

Abstract:

Magnetic resonance force microscopy (MRFM) is a nanoscale imaging technique that uses a force sensor such as a cantilever, nanowire, or membrane, along with a field gradient to detect magnetic resonance and spatially resolve spins. The Marohn group is interested in using MRFM to image soft materials such as membrane proteins and other biomolecules. Detection of a few hundred nuclear spins or one electron and resolution of less than a nanometer have been demonstrated [1-3]. The Marohn group uses equipment at the Cornell NanoScale Facility (CNF) and the Cornell Center for Materials Research (CCMR) to produce cantilevers with nanomagnet tips as well as waveguides to deliver radio- and microwave frequency radiation to manipulate sample spins. In a previous report we detailed sample preparation methods we had developed to reduce noise due to sample charge fluctuations while avoiding sample damage. In this report we provide an update on measurement experiments performed on the sample prepared using our newly developed techniques.

Summary of Research:

Using a protocol developed at the CNF, the Marohn group fabricated attonewton-sensitivity cantilevers and nanomagnets that were affixed to the leading edge using a STEM-FIB system at the CCMR. An experiment on a 40 nm thin film demonstrated 500 proton-sensitivity in a mHz bandwidth [1], a sensitivity equivalent to that required for single electron radical imaging.

In our report for the 2017-2018 CNF Research Accomplishments, we described experiments performed on a 200 nm thick polystyrene thin film doped to 40 mM with 4-amino-TEMPO, a nitroxide radical. Our detection scheme modulates the sample electron spin polarization and detects the change in magnetization as a modulated shift in the cantilever frequency. To minimize cantilever frequency noise from fluctuating sample charges, we used e-beam evaporation to apply a 10 nm layer of gold to the polystyrene sample surface (SC 4500 Combination Thermal/E-gun Evaporation System). Based on our experimental measurements and later conventional ESR of test films, however, we believe that a 20 nm thick layer of the sample is destroyed by the gold deposition. This "dead layer" increases the standoff distance between the magnet tip and sample spins being measured, making it difficult to do single electron detection in a practical measurement time. In our previous report we presented a new "laminate" sample preparation technique that protects the cantilever from sample charge fluctuations without exposing sample nitroxide radicals to direct exposure from physical vapor deposition of metals (Figure 1).

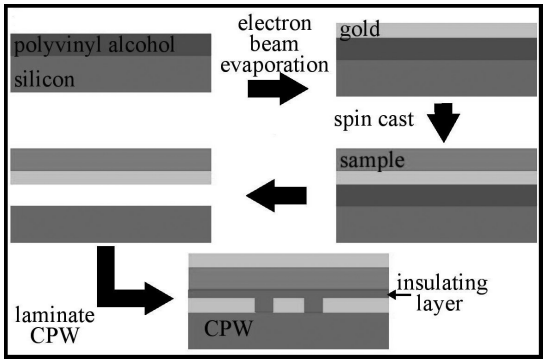

Figure 1: An outline of the sample/top contact transfer process which allows samples to be produced without exposing sensitive nitroxide electron radicals to physical deposition of the gold top contact.

In May-June 2019 we performed experiments on one of these laminate samples using a magnet-tipped cantilever prepared by epoxying a 4-micron-diameter spherical nickel magnet to one of our CNF-fabricated cantilevers. This larger tip reduces per-spin sensitivity of our system but puts more spins in resonance at a given microwave frequency, allowing us to more easily test and calibrate our system.

Our first measurement was to scan the irradiation frequency at a variety of tip-sample separations. This provides us with the tip-field at each separation, tells us about the field-gradient, and lets us fit the result to an analytical model of a spherical magnet (Figure 2).

We also used MRFM to detect the spin-lattice relaxation time T1 of sample spins using a "phase-kick" protocol described by Moore, et al. [4]. Our measured values match Moore's and provide evidence that the laminate sample preparation does not degrade the sample (Fig.3).

Another test of our setup was to perform a proof-of-concept imaging experiment. In theory our sample should not be interesting to image, being a uniform thin film with a constant dopant concentration. However, we found a protuberance in the sample that we could measure. We scan laterally over the sample in small steps to form a 12 × 12 grid. We also vary the irradiation frequency to change the shape of the resonant slice. As the resonant slice scans over the protuberance, it adds to the signal observed at that grid point. By changing the irradiation frequency, we change the radius of the resonant slice and therefore the size of the rings of increased signal (Figure 4). We expect imaging of smaller scale objects to behave in a similar manner.

Future work will involve using a smaller magnet tip to continue imaging experiments on samples with nanoscale features of interest for imaging.

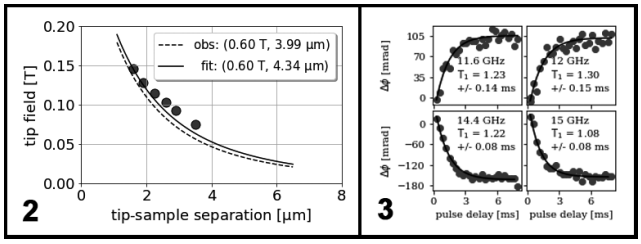

Figure 2, left: *Tip field versus tip-sample separation along with a fit to an analytical magnet field for a spherical model.* ***Figure 3, right:*** *Cantilever phase shift versus pulse delay measured at different irradiation frequencies. Inset shows best fit spin-lattice relaxation time.*

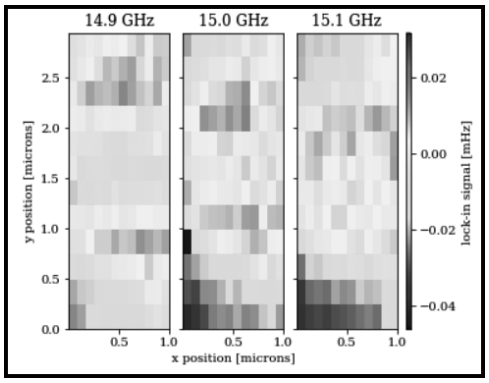

Figure 4: Map of spin-induced cantilever frequency shift at various irradiation frequencies. Step size: Δx = 91 nm, Δy = 258 nm. (See pages vi-vii for full color version of figure.)

References:

[1] Longenecker, et al. ACS Nano 2012, 6 (11), 9637-9645.

[2] Rugar, et al. Nature 2004, 430 (6997), 329-332.

[3] Grob, et al. Nano Letters 2019, 19 (11), 7935-7940.

[4] Moore, et al. PNAS 2009, 106 (52), 22251-22256.