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Towards bimagnetic nanoparticle thermometry

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Abstract

Magnetic nanoparticle (MNP) thermometry is a promising approach for non-invasive and remote temperature sensing for applications in both fundamental and applied sciences. However, conventional MNPs like magnetite exhibit low thermosensitivity (temperature-dependent slope of magnetization) near room temperature. Bimagnetic nanoparticles with tunable thermosensitivity and operating temperature ranges represent a potential route to overcome these limitations. We have begun developing cobalt-doped ferrite MNPs to be used as the core in the planned core/shell bimagnetic nanoparticles. Here we present results taken with a custom-built AC relaxometer and demonstrate that the temperature-dependence of magnetization can be tuned with particle size.

I Introduction

Magnetic nanoparticle thermometry for non-invasive and remote temperature interrogation could find applications in material, chemical, and biological sciences. The key role of temperature in medical diagnoses and procedural intervention highlights the importance of accurate, real-time temperature sensing in medicine and biology. [1] Recent work in MPI has shown promise for extending temperature measurements to a 3D volume, [2,3] but quantitative thermal imaging with well-defined uncertainties has not yet been achieved. Thermometry in 3D for practical applications can only be realized if requisite temperature sensitivity can be achieved at high spatial and fast temporal resolution.

Here, we discuss our progress towards MPI-based 3D thermometry over a broad range of 200 to 400 K, traceable to the SI Kelvin standard. For commercial single-core MNPs (e.g. Fe_3O_4), the thermosensitivity (temperature-dependent slope of magnetization) is typically weak (Fig. 1a, dotted line) near room temperature, which sets a limit on the signal-to-noise ratio for temperature measurement in practical settings where concentrations are low. Our goal is to develop bimagnetic nanoparticles, composed of core and shell of different magnetic materials, which have been predicted to display significantly larger and tunable thermosensitvity. [4] Cobalt-doped ferrite can be used as the basis for a class of MNPs with engineered core/shell composition and structure [5] for exploiting interfacial antiferromagnetic exchange coupling effects for 1) increased thermosensitivity and 2) operating temperature range (Fig. 1c).

In this work, we show our progress towards a bimagnetic nanoparticle system, by first characterizing the magnetic properties of the synthesized cobalt-doped ferrite "core" of the core/shell system. We measure the temperature-dependent DC and AC magnetization for our synthesized cobalt-doped ferrite MNPs in comparison with a commercial MNP sample and demonstrate the capability to tune the former's magnetization curves. In the near future, we will add a ferromagnetic or antiferromagnetic shell layer to complete the core/shell system.

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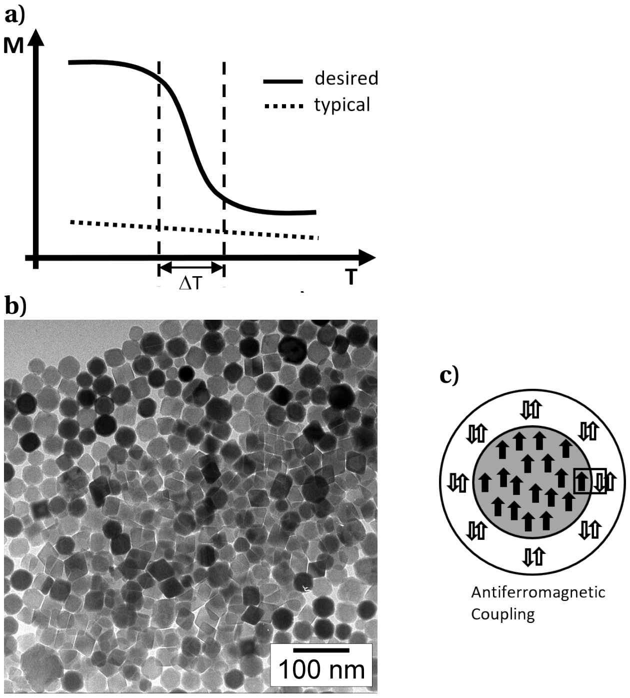

Figure 1: a) Magnetization versus temperature for the typical MNP and the desired MNP (exaggerated for viewing). The ΔT region is centered around room temperature b) Synthesized sample of 35 nm cobalt ferrite MNP. c) Antiferromagnetic coupling of the magnetic bilayer MNP.

II Material and methods

We measured the temperature-dependent magnetization with an AC magnetometer. The Helmholtz coil driving field frequency is 770 Hz with an amplitude up to 9 $\mathrm{mT_{rms}}$. MNP thermometry was performed using a thermally conductive sample holder (Fig. 2a, solid arrow) machined from Shapal [6] ceramic that is temperature-controlled by liquid flow from a recirculating chiller. We measured the sample holder temperature with four platinum resistance thermometers immersed in the body of the holder. In all experiments, the sample holder temperature was stabilized (+/- 25 mK) to the chiller setpoint, and temperatures were sampled in random order during the magnetization measurements. For the detection of AC magnetization, we performed differential measurements using a dual fluxgate (Fig. 2a, dashed arrows) arrangement. The differential signal has a noise floor of 7 dB lower (common-mode noise rejection) and MNP signal amplitude of 10 dB higher compared to the single sensor measurement, thus producing a net gain of approximately 17 dB in the signal-to-noise ratio. An additional observed advantage of the differential measurement is that the drive field and its associated harmonic distortions are suppressed up to 40 dB without the need for additional filtering circuitry. The iron concentration of the commercial Vivotrax [6] sample and our synthesized cobalt-doped ferrite MNPs is ~ 5 mg/ml in a 0.3 mL sample volume.

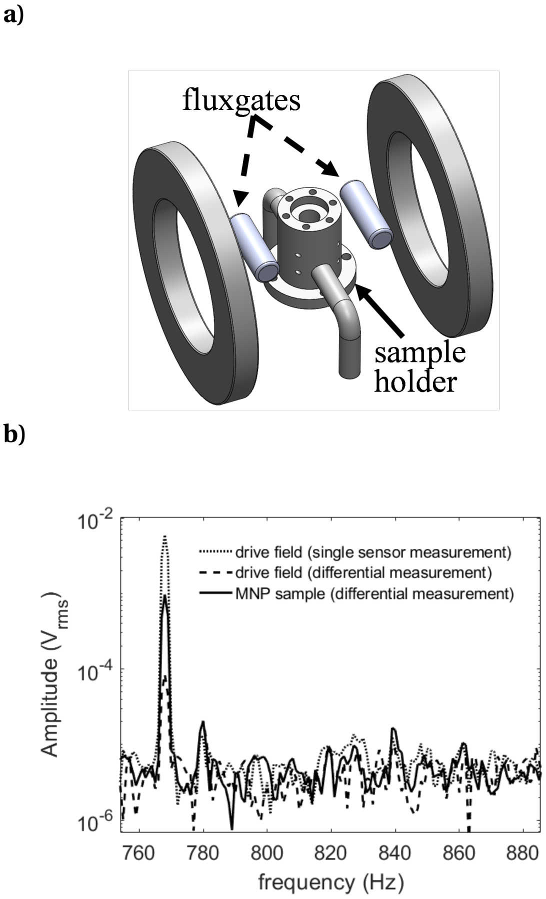

Figure 2: a) AC magnetometer with temperature-controlled sample holder and fluxgate differential measurement. b) Magnetization spectrum of commercial MNPs at a driving field of 3 mT at 770 Hz.

III Results and discussion

We measured the temperature dependence of three MNP samples: one was commercial and two were synthesized for this study. The volatile solvents (hexanes, toluene, and water) used for MNP samples limit the tunable temperature range to 275 K to 320 K. For the commercial sample, the M vs T curve displays the approximately linear behavior with negative slope described by the Langevin function (Fig. 3a). On the other hand, cobalt-doped ferrite MNPs display nonlinear curvature, and a remarkable change in the sign of the temperature-dependent magnetization slope upon increasing the particle size from 7.5 nm to 9.5 nm (Fig. 3b). The sign of the curvature was further verified by DC magnetization measurements (insets in Fig. 3b) over the temperature range of 275 to 305 K. Our synthesized 9.5 nm MNPs display a 50 % change in M vs T compared to 20 % for the commercial sample. We

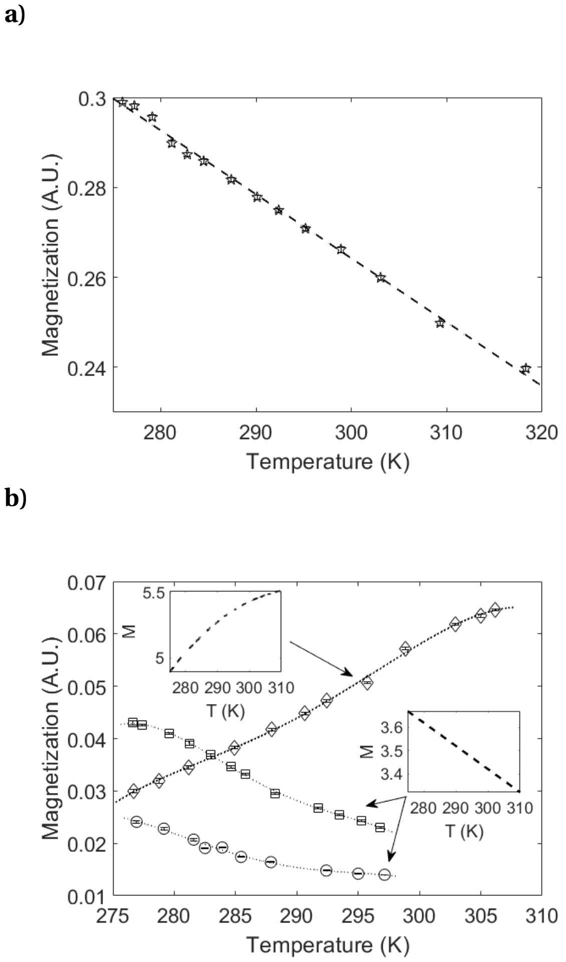

Figure 3: a) M vs T of commercial MNPs (stars). b) M vs T of 9.5 nm cobalt-doped ferrite MNPs (diamonds) and 7.5 nm cobalt-doped ferrite MNPs at two different driving field amplitudes (3 mT – circles; 4 mT – squares). Insets show DC magnetization measurements.

are still calibrating our instrument and do not yet have an absolute scale for magnetization. Even though cobalt-doped ferrite nanoparticles are not likely biocompatible, these results show their temperature-tunability with size, and bimagnetic tunability can eventually be pursued in other families of materials.

IV Conclusions

Our preliminary results reveal that cobalt-doped ferrite nanoparticles warrant further development and engineering as the basis for bimagnetic MNPs, with the anticipation that the thermosensitivity of the completed core/shell topology would be significantly better over a wide temperature range. Future temperature-dependent magnetization measurements over a range of frequencies and with solid samples will isolate contributions from Néel and Brownian relaxation mechanisms. In the future, we seek to exploit this class of MNPs for thermal 3D imaging and temperature control applications.

Acknowledgments

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Author's Statement

The authors declare no conflict of interest.

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[6] Reference is made to commercial products to adequately specify the experimental procedures involved; it does not imply recommendation or endorsement by NIST.