DOE AWARD Number: DE-FG02-07ER46264

University of Nebraska -- Lincoln

Title of Award: “Magnetic Cluster States in Nanostructured Materials”

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This report covers: 1/1/06-5/31/08

## EXECUTIVE SUMMARY

This project sought to understand how structural disorder could be manipulated to produce desired magnetic properties. Disorder would be parameterized using atomic-level measurements and correlated to the magnetic behavior of model systems. Unfortunately, sufficiently simple model systems could not be produced and their two-phase nature made a direct correlation between magnetic properties and structural properties impossible.

## GOALS/ACCOMPLISHMENTS

The goal of this proposal was: **use model ferromagnetic systems in which we have control over the type and extent of disorder to understand the effect of local disorder on determining macroscopic magnetic behavior.**

The three specific objectives are different aspects of the same question: How do different types of disorder affect macroscopic magnetic properties?

1. Determine how different types of disorder affect the magnetic phase transition temperature and breadth.
2. Determine the correlation between magnetic and structural ordering lengths as a function of different types of disorder.
3. Determine how the fundamental nature of the ferromagnetic transition changes as a result of different types of disorder and how this modifies extrinsic parameters such as coercivity.

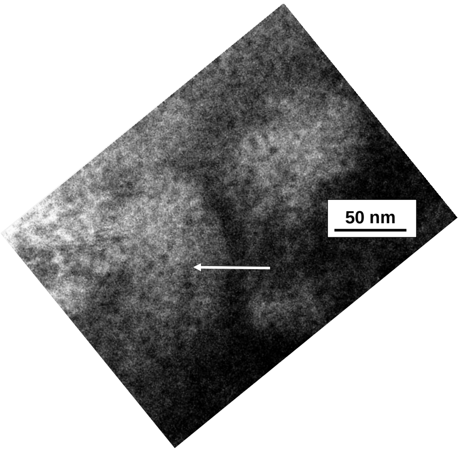
Objective 1 was partially met, as we fabricated samples using mechanical milling, inert-gas condensation and melt spinning and studied how different types of disorder (coordination number, average distance between nearest neighbors and rms deviation in that distance) were produced.

The main goal and the other two objectives were not met due to the principal investigator leaving the University of Nebraska- Lincoln in May 2008. A limited series of samples were studied ; however, the two-phase nature of the samples we were able to produce made it impossible to achieve the quantitative parameterization of disorder we sought to accomplish.

## PROJECT ACTIVITIES

**Sample Fabrication.** We have completed work on the $Gd_{100-x}Fe_x$ series (x = 0-40), with samples made by inert gas condensation (IGC) and melt spinning and are working with mechanical milling. We did not receive run time at the Advanced Photon Source at Argonne National Laboratory in summer or Fall 07 and did not submit a request due to the difficulty analyzing the XAFS data from the two-phase samples..

Human Resources Development: Graduate student David Schmitter completed his thesis this year and two papers from the thesis are being submitted to Physical Review B. Student Geoff Shelburne was funded from this grant through May 2008. The postdoc that was working on this project had to leave without notice, first in November 2006 and again for good in February 2007 because of his father's illness. A postdoctoral researcher was hired in October 2007, but left due to being offered a job in his home country in February 2008. This series of personnel instability slowed progress significantly.



## Results

Gd Fe is unique system in that the elements phase segregate, which produces samples with hcp-Gd crystallites surrounded by a grain boundary region of GdFe. We performed nano-STEM at Ames Laboratory and were able to show that most of the Fe in the samples is indeed in the grain boundary region. The Fe concentration does not go to zero because the grains are roughly spherical and the thickness of the sample means that the probe is measuring the grain boundary region that lies underneath the grain.

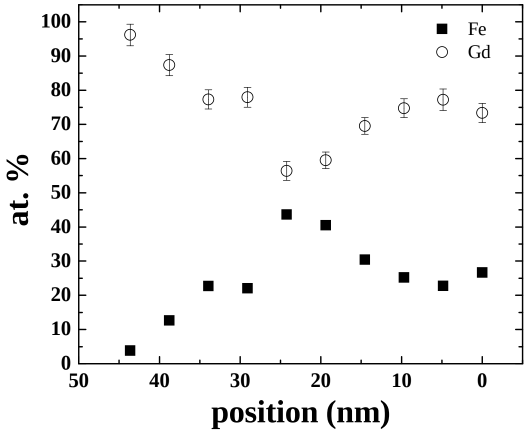


Figure 1: a) STEM image of $Gd_{80}Fe_{20}$ shows the path of the EDX analysis b) Gd and Fe concentrations along the arrow.

EXAFS data from our previous run at the APS have been analyzed and the disorder parameters extracted for the melt-spun samples. This analysis has been complicated because the samples are two-phase, but we now have results in which we are confident. Figure 1 shows STEM analysis, completed at Oak Ridge National Laboratory, of the two-phase nature of the sample. Figure 2 shows the raw XAFS data.

These systems show interesting magnetic behavior that appears to be well-described by the random anisotropy model, surprisingly, even in samples where the Gd grains are more than 100 nm in size. We have succeeded in making a series of samples of varying Fe concentration in which the Gd crystallite size is constant and only the size of the grain boundary regions changes as the volume fraction of crystalline material changes.

We have shown that the behavior of the amorphous phase in the grain boundary region is very different from bulk amorphous GdFe of comparable composition and believe the difference is in pinning at surfaces and interfaces.

The coercivity exhibits an unusual behavior in that it decreases as the Gd grains order, shows a hump, and then increases again, as is shown in Figure 4. This behavior can be explained in terms of increasing anisotropy that decouples the grains as the temperature decreases. This behavior is cluster-glass-like because some of the important signatures of cluster glasses, like a sharp increase in the coercivity below the peak in the ZFC magnetization, are not present.

We have taken the Gd-Fe system as far as we can; at this point, we need to focus on systems that are single phase to make the atomic-scale disorder parameterization possible.

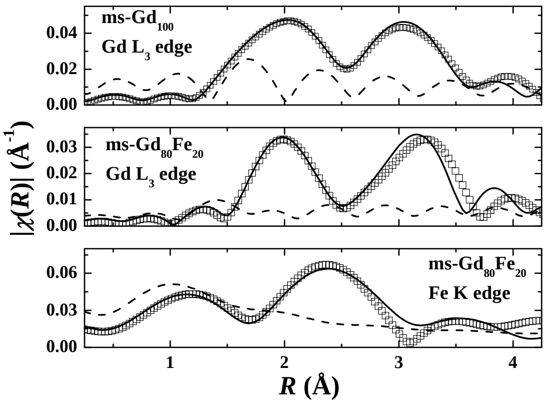


Figure 2: EXAFS fitting results for Gd-Fe samples. Points represent the data, solid lines are the fits and the dashed lines are the background term to the fits.

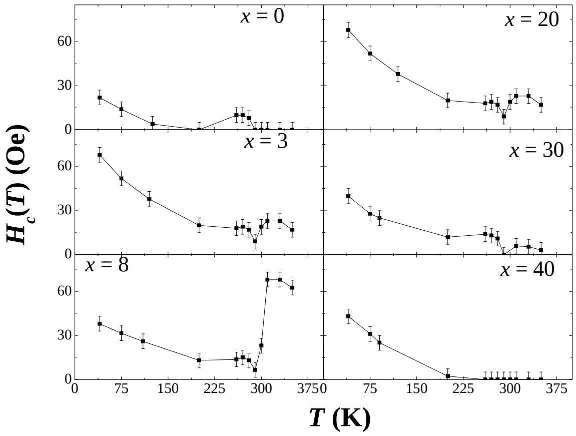


Figure 3: The temperature dependence of the coercivity

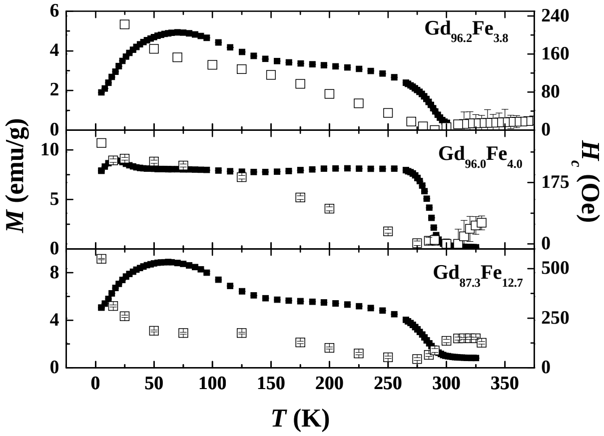


Figure 4: The temperature dependence of the magnetization, superposed with the temperature dependence of the coercivity, shows how the two are related.

## PRODUCTS

Publication: "Correlating Structure with Magnetism in Two-Phase Gd:$Gd_{100-x}Fe_x$ Structures" has been submitted to Physical Review B. This paper is co-authored with Daniel Haskel of the Advanced Photon Source.

## PERSONNEL

Kishore Sreenivasan (until 2/15/07) – postdoctoral researcher on partial support

Geoff Shelburne (all year) – third year graduate student on full support under this grant.

David Schmitter (until 8/31/07) – graduated with Ph.D. 8/07, was 25% supported.

## PLANNED ACTIVITIES FOR NEXT YEAR

A related series of samples being studied as a result of the Gd-Fe results is GdN. GdN has very interesting properties: it can be semiconducting or metallic, and has been reported as a ferromagnet and a spin glass. The properties are determined by the nanostructure. In addition, GdN has a lower $T_C$ and allows us to measure at temperatures far above $T_C$, without changing the nanostructure. GdN normally is difficult to make, requiring nitriding at very high temperatures for long times; however, we have fabricated Gd/GdN phases using both melt-spinning and inert-gas condensation. Figure 2 shows the temperature dependence of the zero-field-cooled magnetization (open symbols, at 100 Oe) and the coercivity (closed symbols) for an IGC-GdN sample with a grain size of 18 nm. (This sample was annealed at 600°C for 10 hours to complete the nitriding). Coercivity is observed in the nitrided sample at temperatures up to 400 K, which is significantly higher than the $T_C$ of 60 K. (No iron has been observed in these materials.) We have attempted to obtain single-phase GdN samples by inert-gas condensation using reactive sputtering, post-deposition exposure to N with and without annealing, and by nitriding melt-spun samples in flowing N. Reactive sputtering appears to be most promising for producing single-phase GdN.

Since the magnetic and transport properties are highly dependent on the atomic-level structure, XAFS and XMCD measurements at APS will be made. We have just started working with Tb-based materials, which will allow us to do neutron scattering measurements at Los Alamos in collaboration with Jim Rhyne. We anticipate similar magnetic behavior for the Tb-based systems, although at lower transition temperature than for the Gd-Fe system. (Gd, being a strong neutron absorber, is a poor candidate for neutron scattering measurements.) We also plan to investigate the low-temperature peak in GdFe and GdN these materials and use the structural results from XAFS to understand the origin of this glassy behavior.