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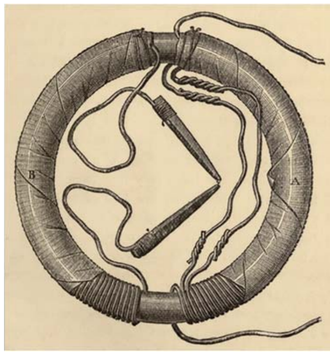

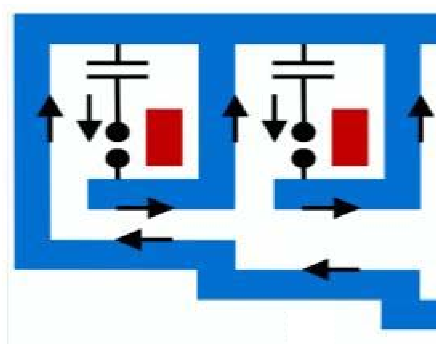
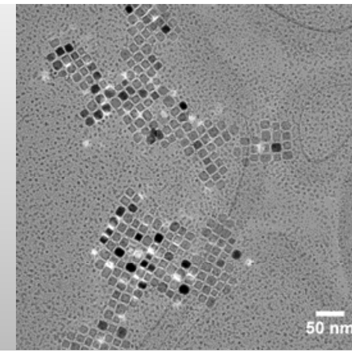

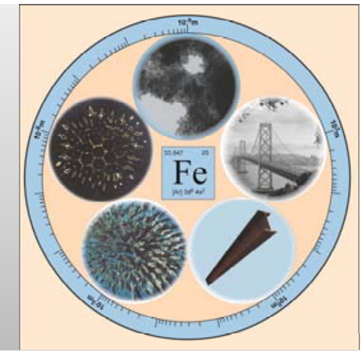

Nanocomposites for LTD Magnetic Cores

Dale L. Huber

Center for Integrated Nanotechnologies

Sandia National Laboratories

U.S. DEPARTMENT OF ENERGY

NNSA National Nuclear Security Administration

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Dale.Huber@sandia.gov

Current State of the Art

- Wound composite with Metglas layers alternating with Mylar
- Parasitic current within an LTD cavity reduces the current delivered to the load. Presently, the magnitude of the parasitic current is ~10% of the load current.
- Present core materials have hysteresis, which means each core throughout the entire accelerator would need to be re-set after every shot using a low-amplitude long current pulse.
- At current manufacturing capacity it would take decades to fabricate the cores required for a 50-MA accelerator.

Dale.Huber@sandia.gov

Materials for Magnetic Core Inductors

Saturation Magnetizations (Tesla)	
Fe	**2.15**
SiFe alloy	**1.97**
Metglas 2605	**1.56**
SNL Iron nanoparticles	**2.04**

- Desired Properties of Magnetic Cores
  - High susceptibility
  - High saturation magnetization
  - Low/no hysteresis
- Current material of choice is wound amorphous iron alloy tape
  - Iron for high magnetization
  - Amorphous to decrease hysteresis
  - But, amorphous materials have depressed magnetization

- Iron nanoparticles provide an opportunity to improve all of the critical properties
  - Iron has the highest saturation magnetization of any element
  - Nanoparticles have enhanced susceptibility
  - Even crystalline iron nanoparticles can be hysteresis free
  - *Superparamagnetic nanoparticles have properties that are not possible in bulk materials*

Enabling Science

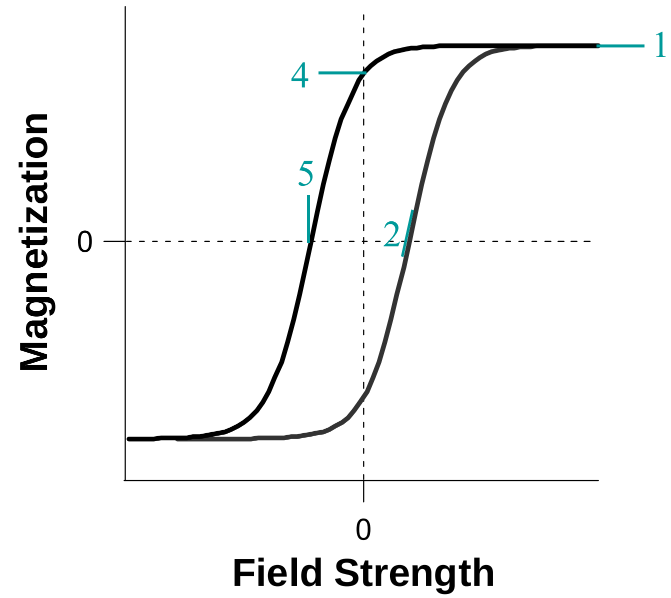
- Chemistry
  - Current approaches are not scalable
  - Developing new methods, one has been patented, but others can be once they are reduced to practice (eg continuous microwave synthesis)
  - Minimize cost through use on inexpensive reagents, processes.
  - Develop appropriate matrix with maximum loading and good mechanical properties—we have done this for high refractive index nanocomposites.
- Physics
  - Only the physics of ideal magnetic nanoparticles are well understood. Ideal implies isolated particles in non-interacting matrix
  - Real systems are non-ideal so we must understand:
    - Particle-matrix interactions (matrix can enhance or decrease magnetism)
    - Multi-particle dipolar interactions
    - Formation of structured, anisotropic materials

Project Plan

- Determine optimum size for the iron nanoparticles
  - We expect ~20 nm will be ideal
- Optimize particle loading
  - Loading too high may yield hysteresis
  - Low loading decreases magnetization
  - Develop a material with good flow properties before curing to allow molds to be poured. (Monson and Huber patent application relating to TiO_2)
- Create small scale test structures
  - We can make and test small parts in realistic configurations.
- Develop a detailed plan for scale up and tech transfer
  - A number of methods can be used for large scale synthesis of nanoparticles
    - Microwave synthesis
    - Continuous reaction (Huber patent recently issued)

Terminology in Magnetic Measurments

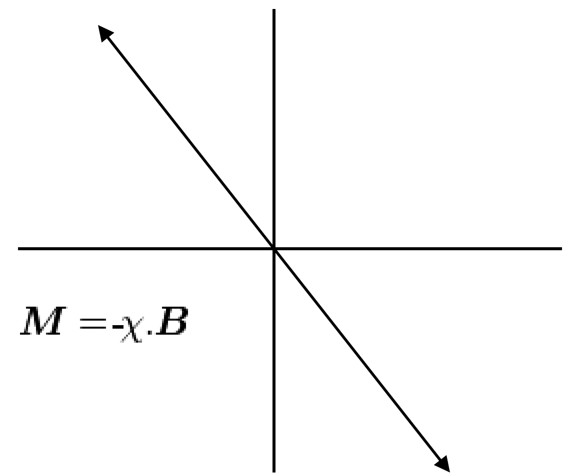
1. Saturation magnetization, maximum magnetization for a material (M_{sat}, M_s, σ_s)
2. Susceptibility, ease of magnetization, slope of magnetization curve (χ, χ_i)
3. Coercivity, amount of hysteresis.
4. Remanence, remaining magnetization when the external field is removed.
5. Coercive Field, the reverse field required to get to 0 magnetization. (H_c)



Diamagnetism

- Weakly repulsive force
- Caused by the repulsion between a magnetic field and moving electrons.
- Everything has diamagnetism, not everything is diamagnetic.
- Susceptibility of about -10^{-6}
- No saturation
- No coercivity
- No temperature dependance
- Examples: water, gold, element & compounds with full valence shells.
- Superconductors are “superdiamagnetic” with a susceptibility of -1

M v B
For all fields



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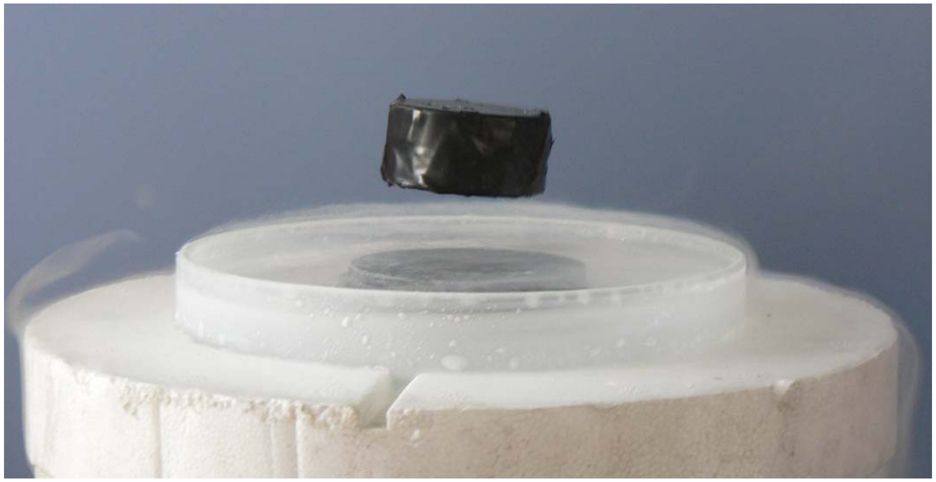
Diamagnetism Demonstrations



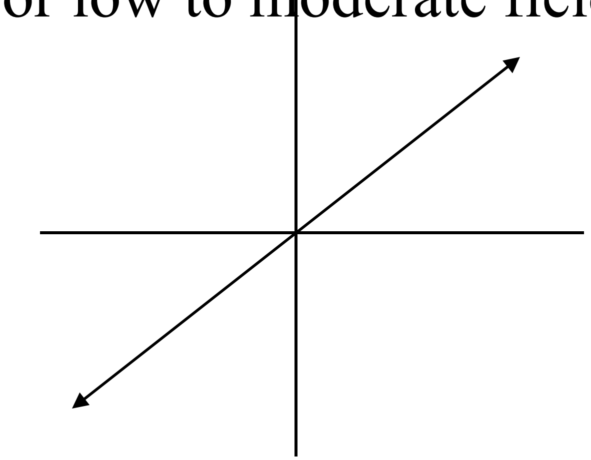
Dale.Huber@sandia.gov

Paramagnetism

- Weakly attractive, susceptibility of $<10^{-3}$
- Caused by attraction between magnetic field and unpaired electrons.
- Materials are "paramagnetic" when their paramagnetism is greater than their diamagnetism.
- Temperature dependant, higher T, lower χ
- "Strongly" paramagnetic materials include most of the d- and f-block elements and ions

M v B

For low to moderate fields

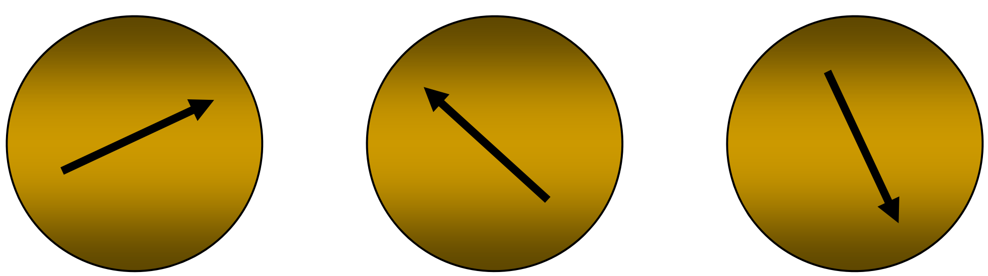


Curie's law

$$\boldsymbol{M} = \chi.\boldsymbol{B} = C.\frac{\boldsymbol{B}}{T}$$

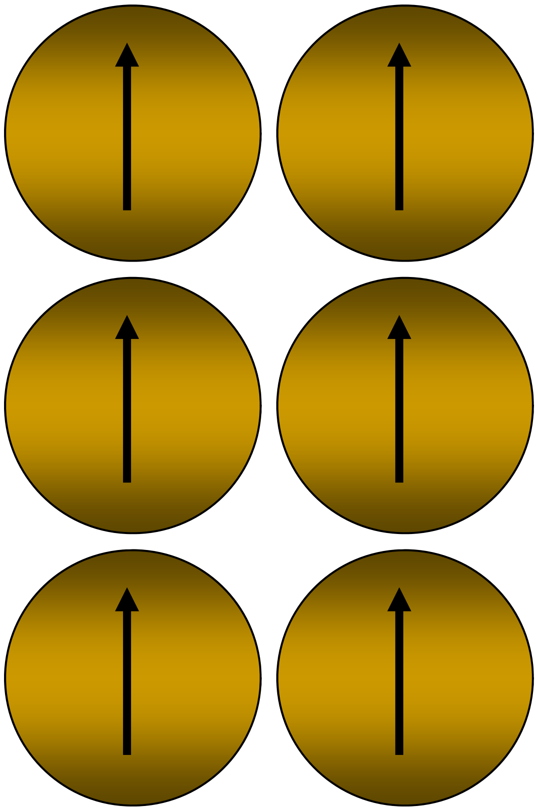
T in K

Material specific Curie constant. This relation holds only when far from saturation



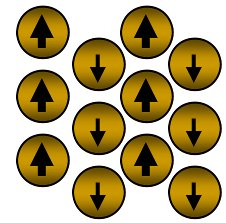
Ferromagnetism

- Paramagnetic atoms acting collectively with spins reinforcing
- Strongly attractive, susceptibility can be as high 10^6
- These are what we typically think of a magnetic: iron, cobalt, nickel, etc

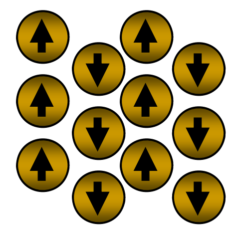


Other forms of collective magnetism

- Ferrimagnetic: magnetite and other ferrites, most magnetic metal oxides.
  - Spins opposed, but do not cancel out completely



- Antiferromagnetic: chromium
  - Spins opposed, cancel out completely



- Spin vortices and other unusual structures are present in rare earth elements.

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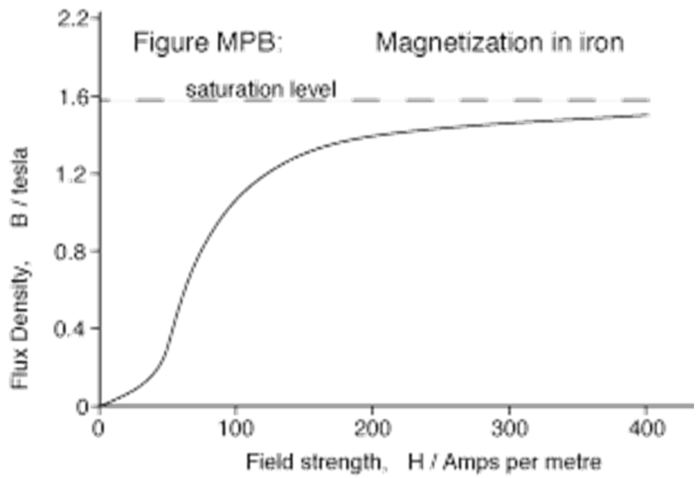
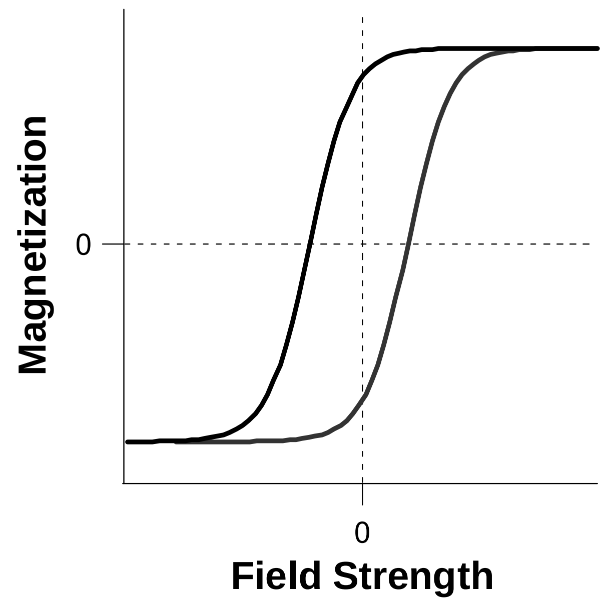
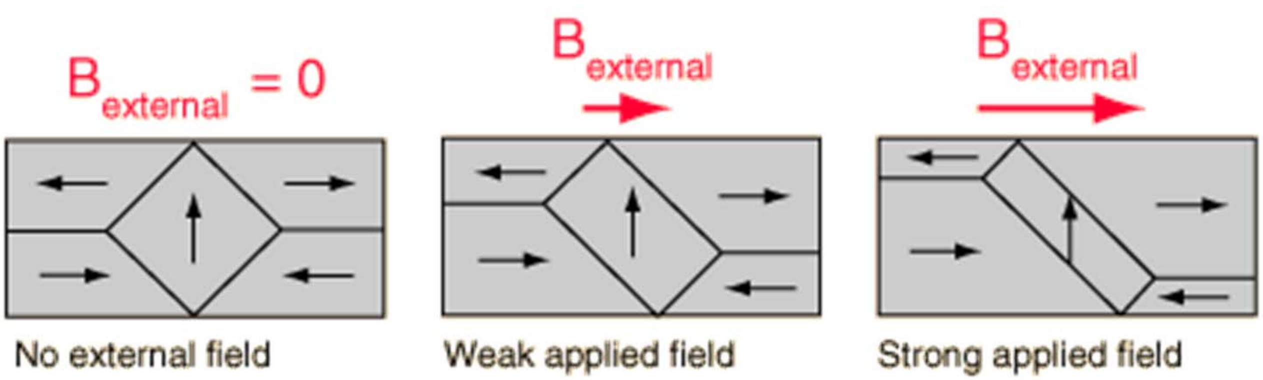
Magnetizing a Ferromagnet





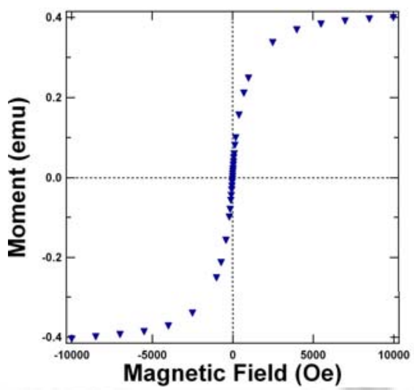


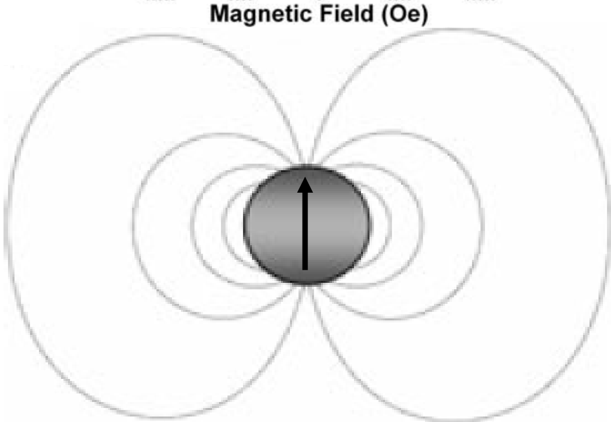
Dale.Huber@sandia.gov

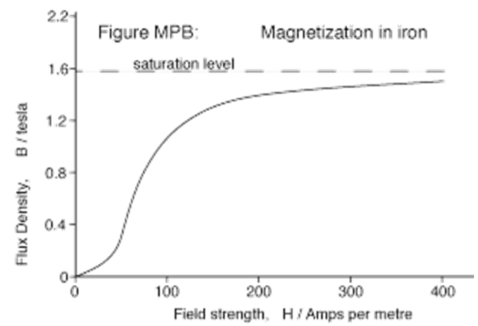
The Other Form of Magnetism: Superparamagnetism

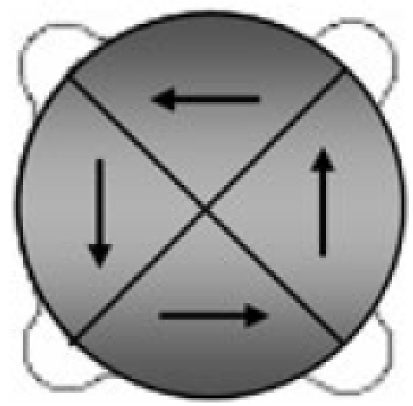
Superparamagnetic particles are single domain magnetic particles whose electron spins are aligned into a single giant moment.

Below a certain size (~40 nm diameter) the energy required to create a domain wall becomes larger than the decrease in energy the particle would experience through minimizing flux leakage.









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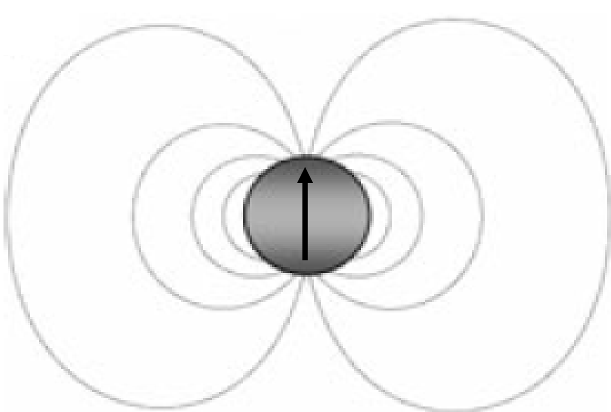
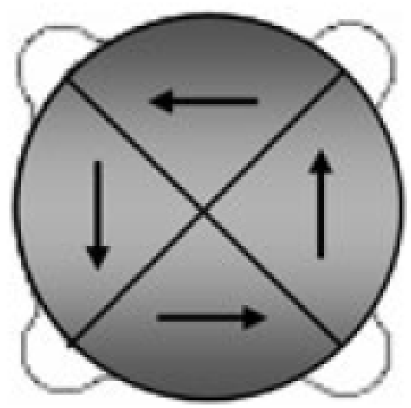
Single Domain:

- More flux leakage
- No energy spent creating domain walls



Multidomain:

- Less flux leakage
- Energy required to create domain walls

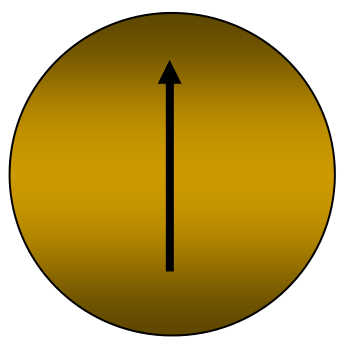


Dale.Huber@sandia.gov

Blocking Temperature

Blocking Temperature (T_B): Point where a particle's dipole can reorient in the timescale of an experiment (100 s).

- T_B is directly dependent upon the product of the magnetocrystalline anisotropy and particle volume.
- Above the T_B particles are superparamagnetic (no hysteresis).
- Below T_B they are ferromagnetic (hysteresis, coercivity).
- Particle susceptibility (χ) has a maximum at T_B
- χ values much larger than multiple-domain particles ($\chi \leq 3$ for multiple-domain particles)



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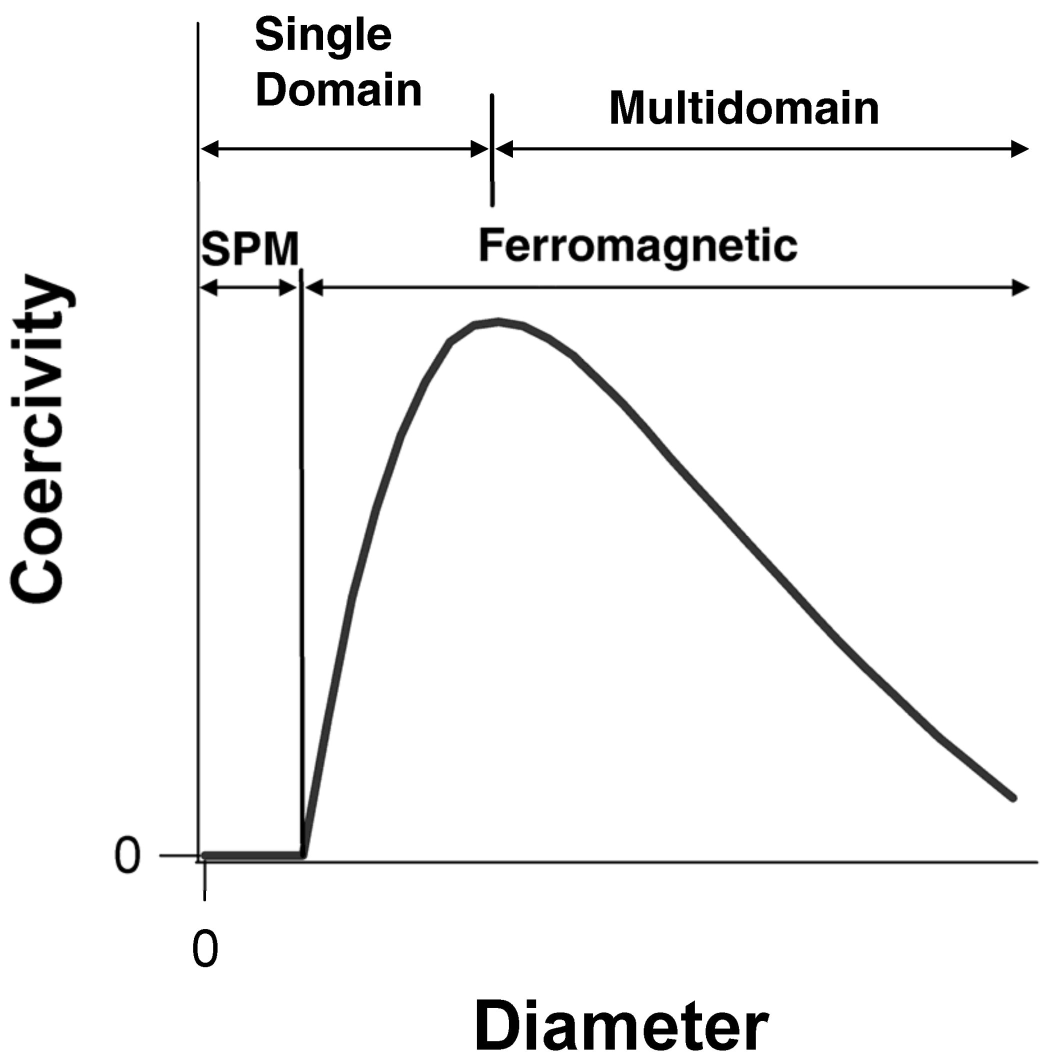
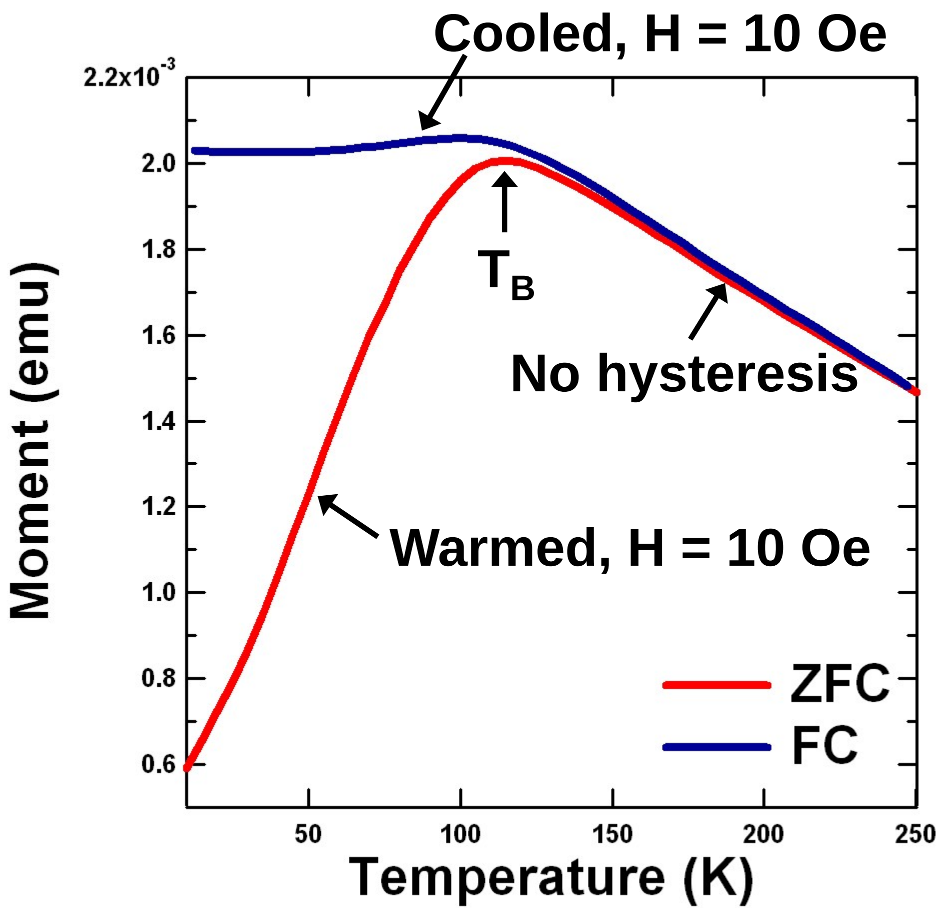
Size Dependent Behavior



- Plot is for a fixed Temperature
- Behavior is for an ensemble of magnetic nanoparticles
- Particles cease to be superparamagnetic above ~ 40 nm in diameter
- Gradual approach to bulk behavior

SQUID Magnetometry – Thermal Response



- ZFC: Zero field cooled
  - Cooled to low T while H = 0
  - Small field (H = 10 Oe) applied
  - Data collected as sample warms
- FC: Field cooled
  - Data collected while cooled to starting point
- Particles dispersed in frozen solvent, measuring Néel relaxation only (internal rotation of moment)
- T_B is maximum in ZFC data
- Double diameter, increase T_B by 8

$$T_B = \frac{KV}{30k_B}$$

$T_B \propto r^3$

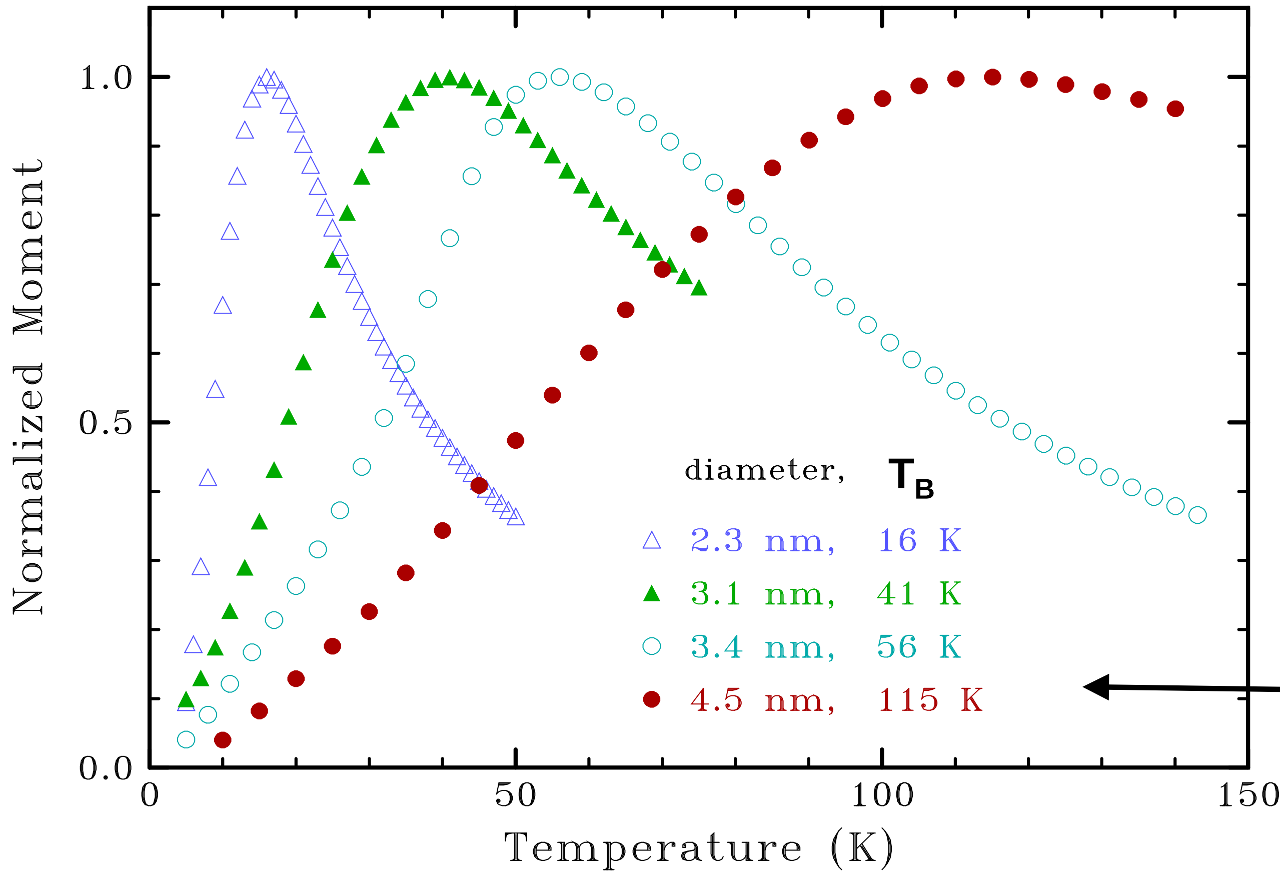
K = anisotropy

V = particle volume

$k_B = 1.3807\times10^{-16}$ ergs/K

Dale.Huber@sandia.gov

T_B vs. Fe Nanoparticle Diameter

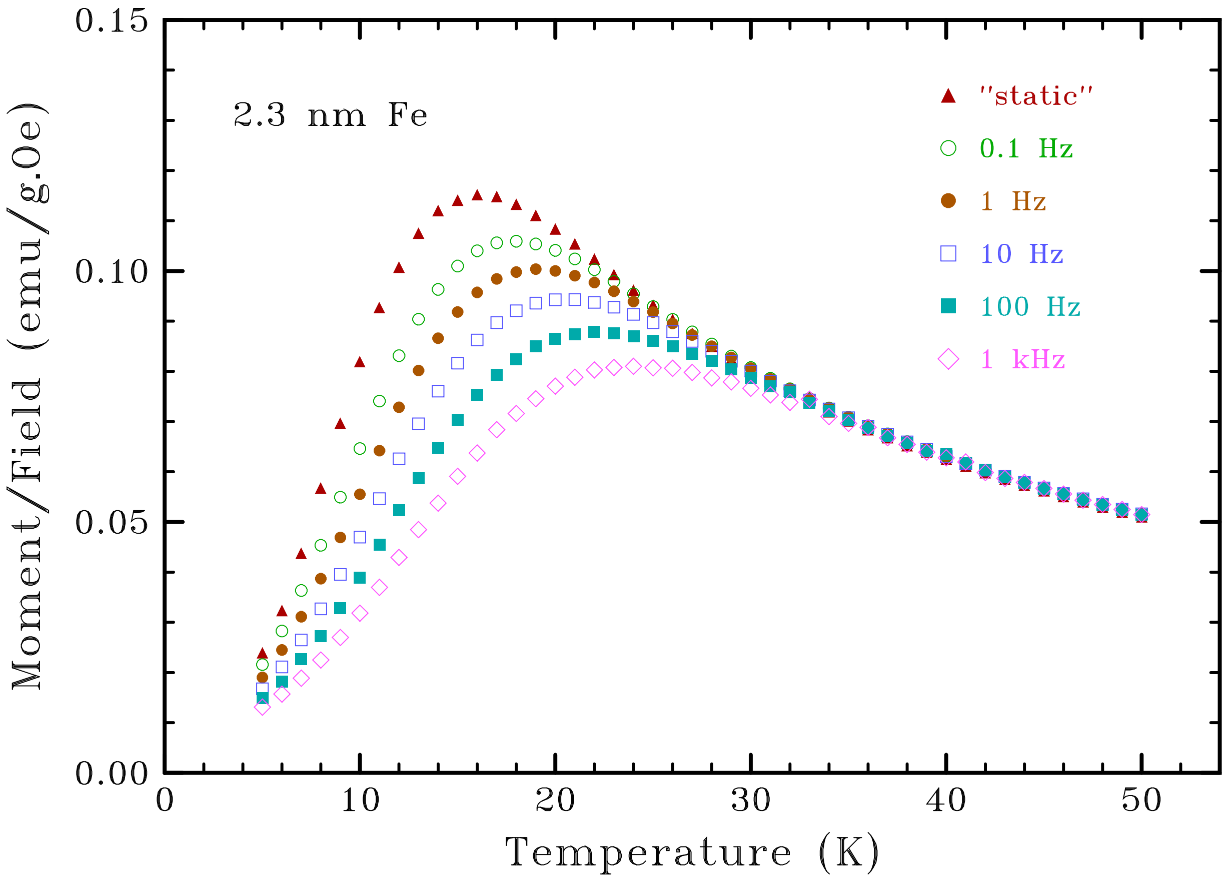


T_B scales with V

- All curves ZFC
- Broad peaks for larger particles expected
- Plotting σ vs. T/T_B peaks would superimpose

Dale.Huber@sandia.gov

AC Susceptibility

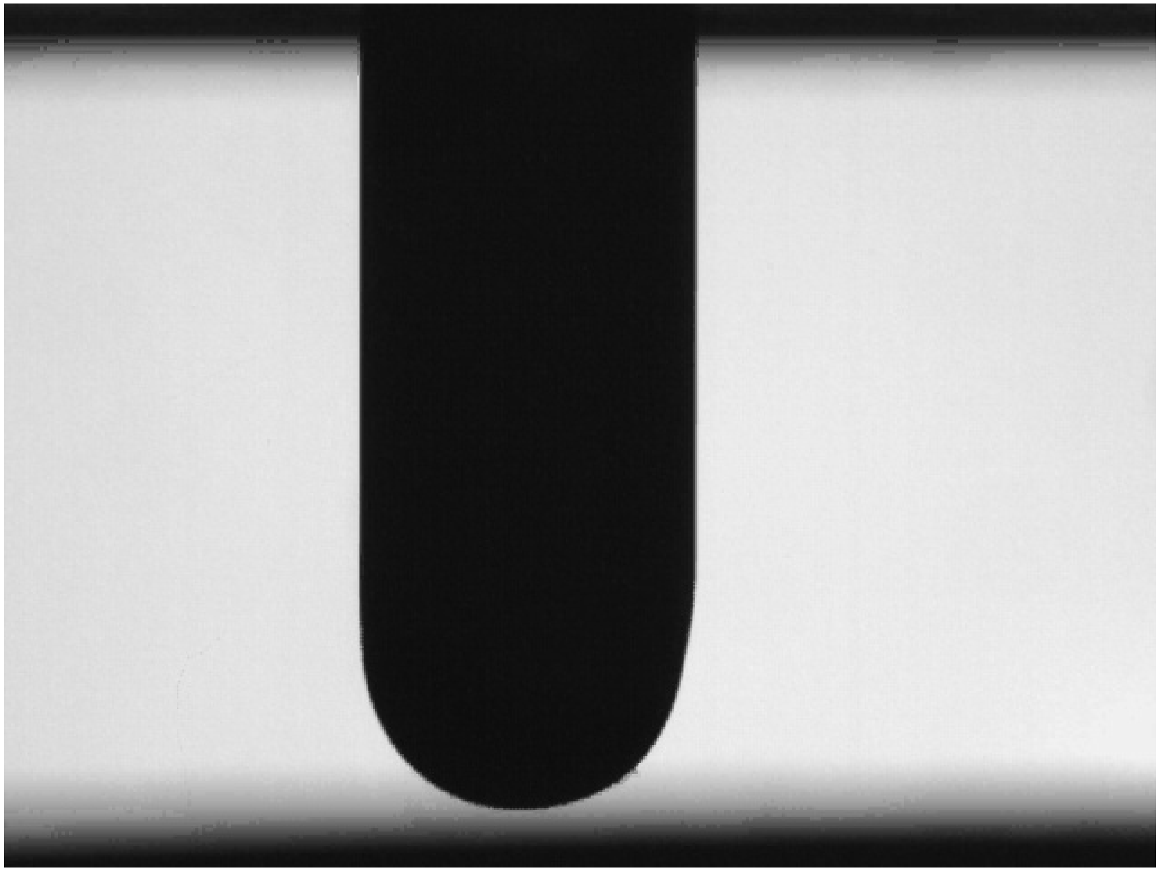


- Spin reorientation time is thermally activated
- AC measurements determine T_B vs. frequency
- "static" measurement time ~ 100 s (0.01 Hz)

Dale.Huber@sandia.gov

Magnetically Structured Composites Horizontal Sheets

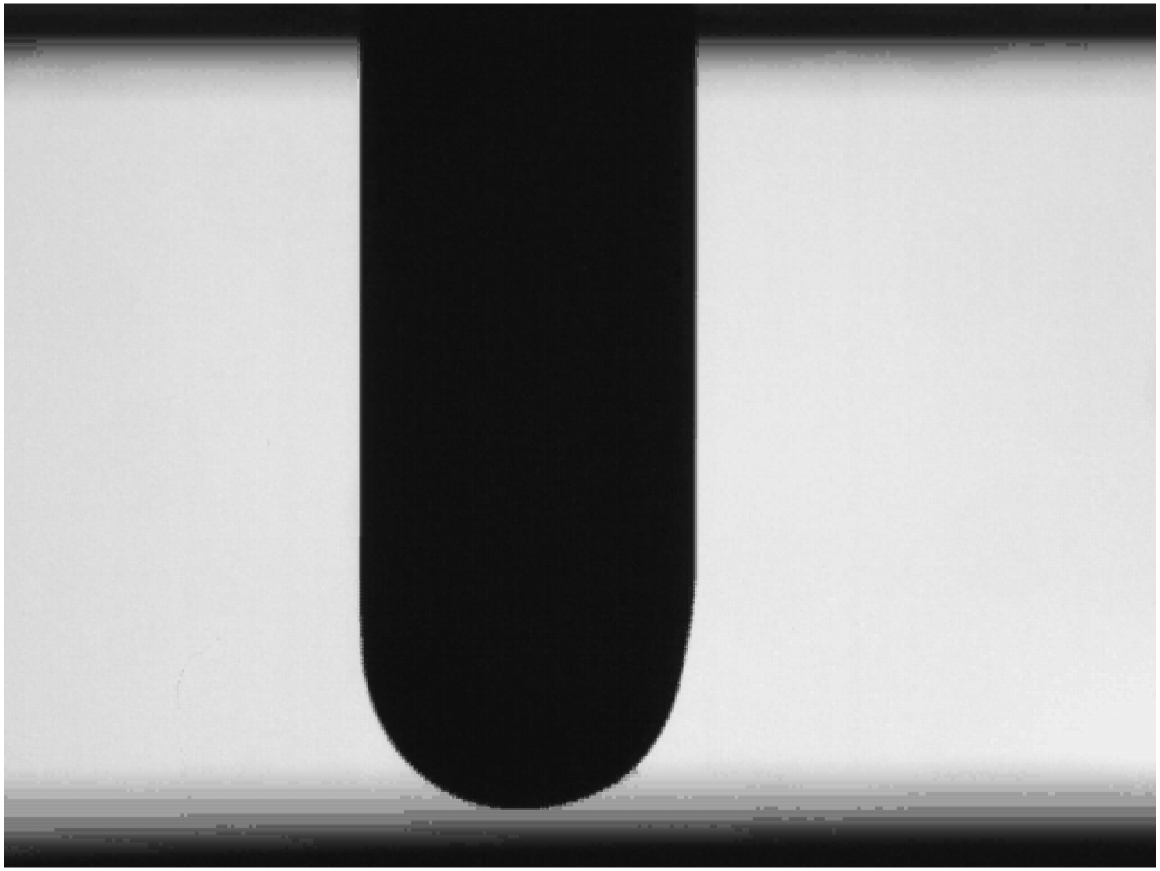
- 3 nested Helmholtz coils allow the application of magnetic fields in x, y, and z axes
- Rotating fields created by applying 2 AC fields with 90° phase lag
- Sample held in a 5 mm glass tube
- 100 G rotating field



With J.E. Martin

Magnetically Structured Composites Vertical Sheets

- 3 nested Helmholtz coils allow the application of magnetic fields in x, y, and z axes
- Rotating fields created by applying 2 AC fields with 90° phase lag
- Sample held in a 5 mm glass tube



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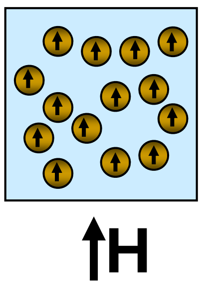
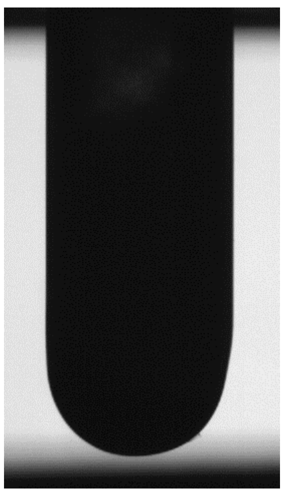
Effect of Structuring on Magnetic properties

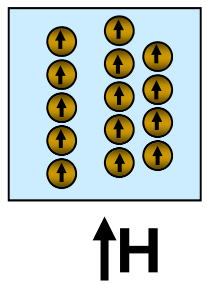
Dispersed particles

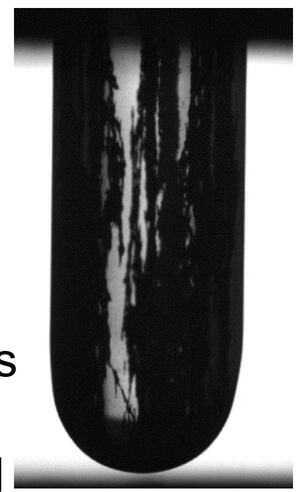




Local field is equal to applied field

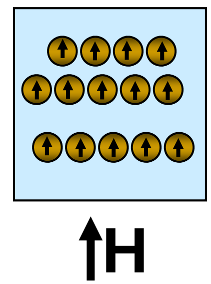
Vertical Structures

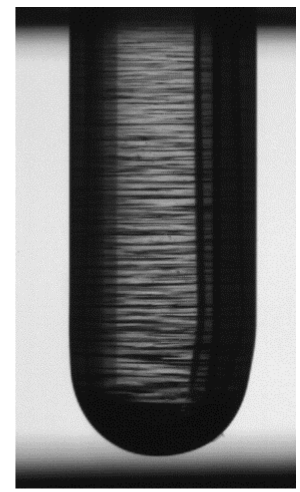




Local fields are reinforced

Horizontal Structures



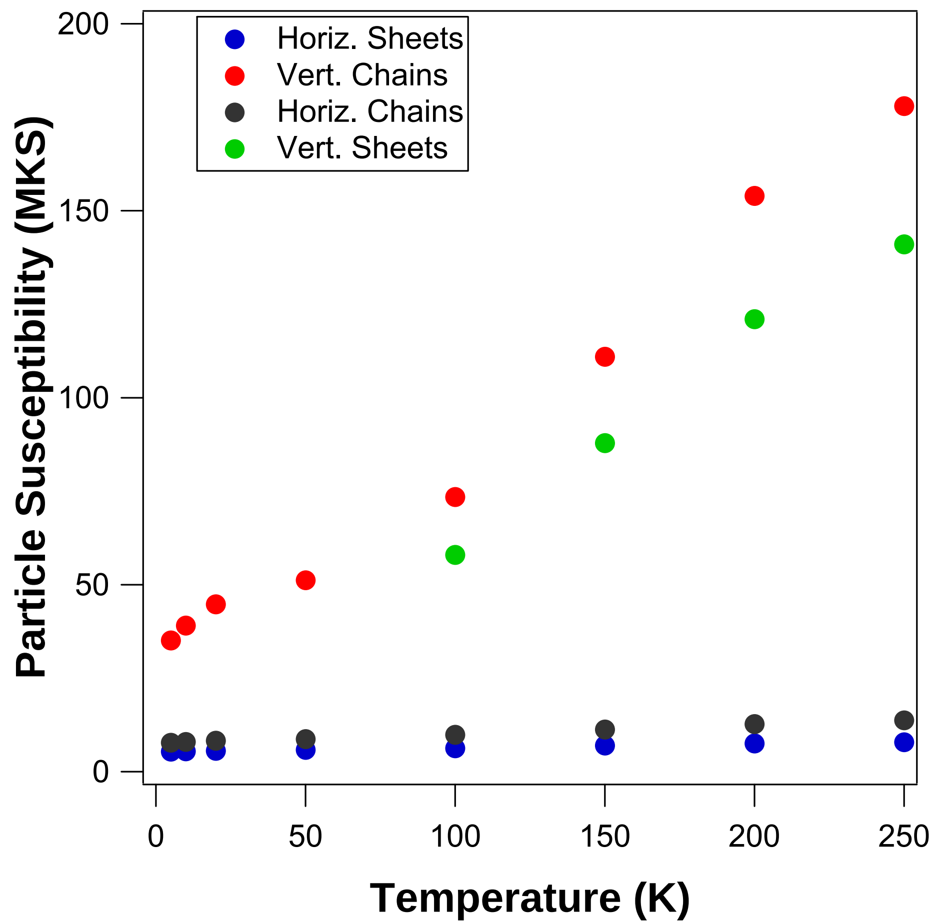


Local fields are weakened

Dale.Huber@sandia.gov

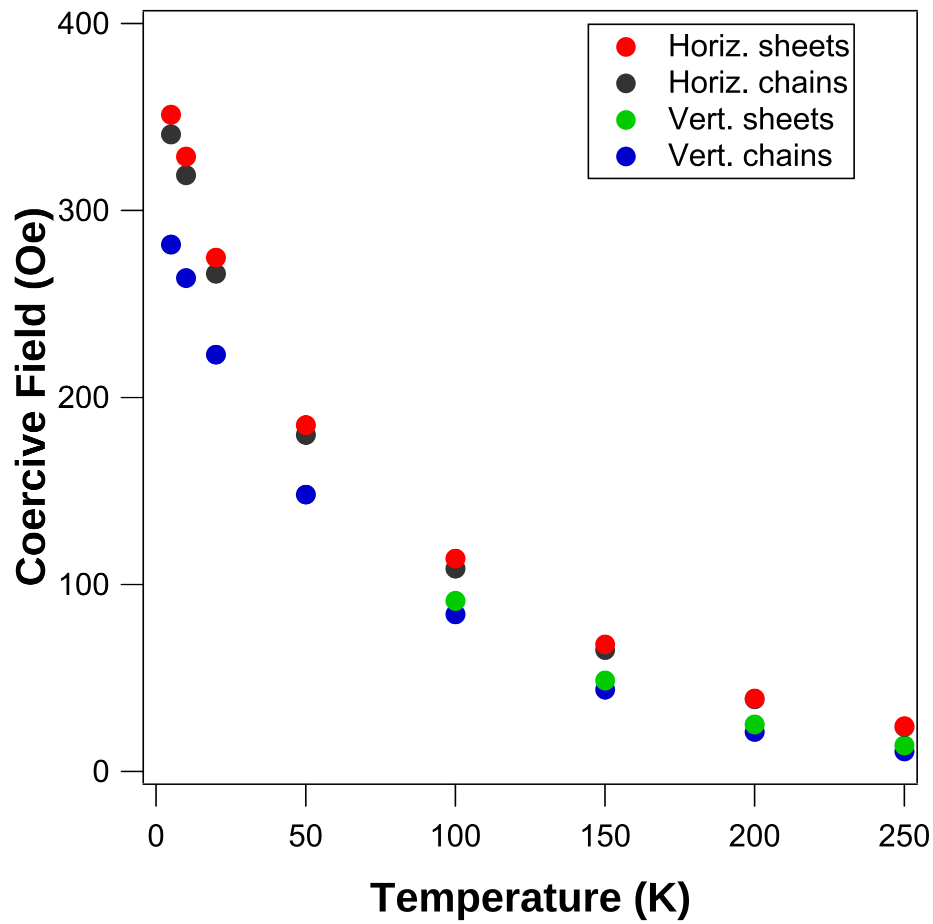
Susceptibility Enhancements in Structured Composites

- Structured with 150 G AC fields
- χ_p as high as 180 (at 250 K) should continue to increase with increasing T
- χ_p limited to 3 for micron scale particles
- Highly anisotropic composites



Coercivity of Structured Composites

- Enhanced susceptibility composites show reduced coercivity
- Cooperative flipping of spins



Dale.Huber@sandia.gov

Optimizing Size

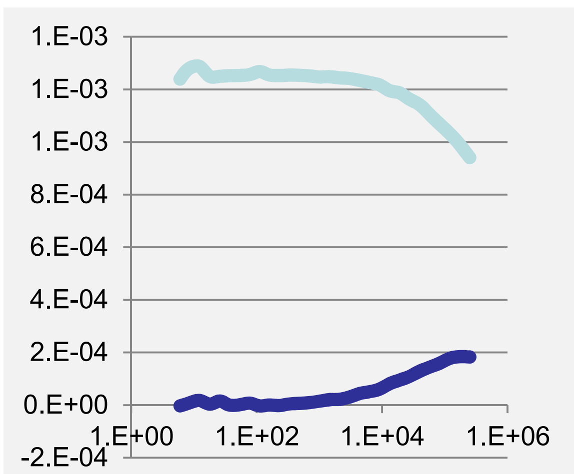
Scalable synthesis

- Microwave assisted thermolysis of $Fe(CO)_5$ in octadecene.
- Reactions can be completed in 5 min.
- Opens the possibility of a flow reactor for production quantities.
- Surfactant used: oleyl amine.

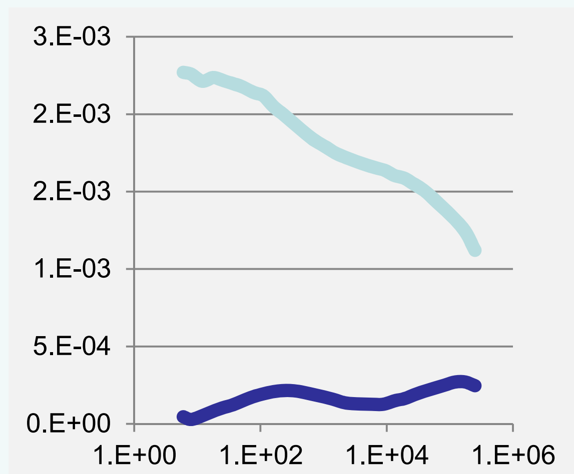
Complex susceptibility vs frequency tells us the maximum size to use

- Real Susceptibility (in phase)
- Imaginary Susceptibility (out of phase or loss)

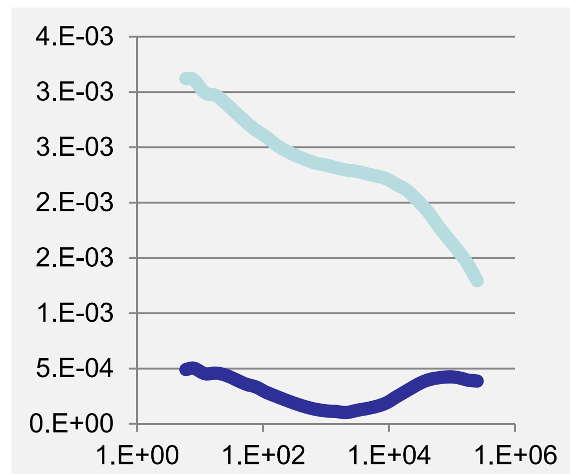
15 nm Fe particles



20 nm Fe particles

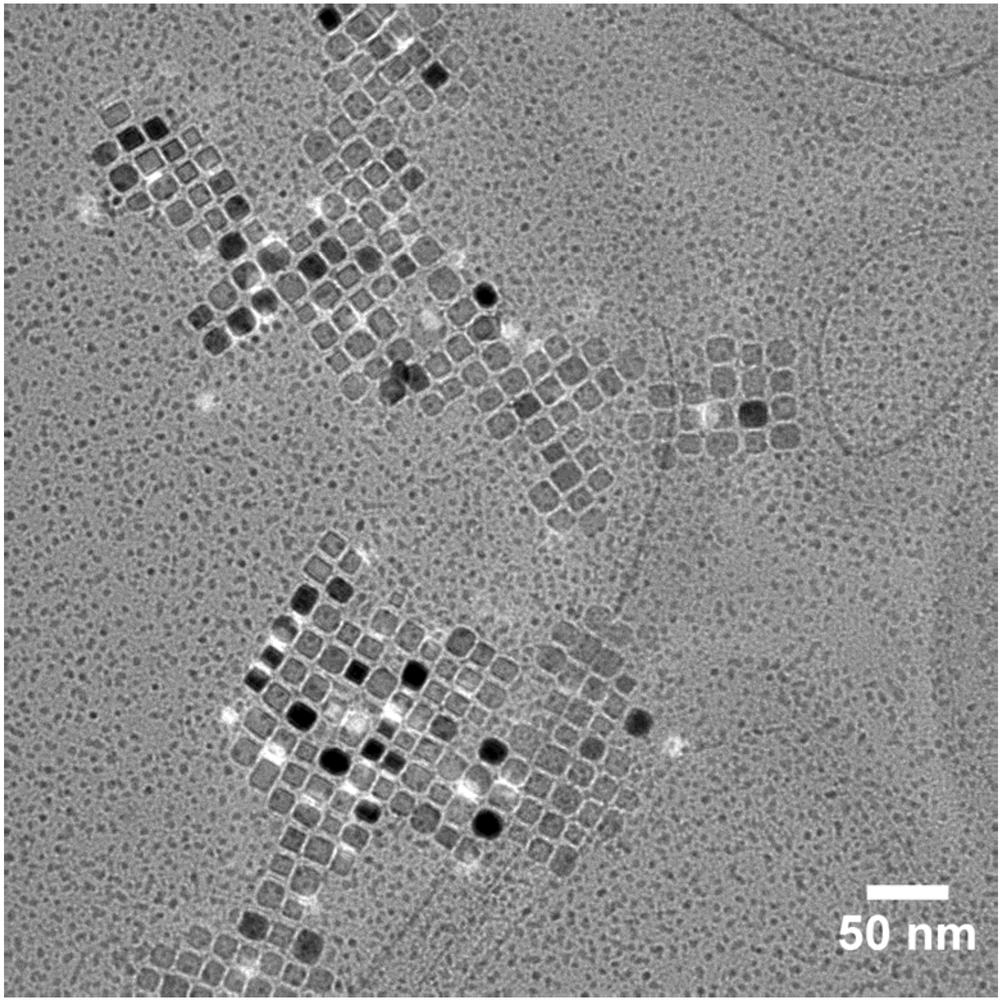


25 nm Fe particles



Dale.Huber@sandia.gov

Cubes on a Simple Cubic Lattice



- Thermolysis of $Fe_3(CO)_{12}$ in octadecene
- Antiferromagnetic ordering between particles?
- Main focus is on avoiding coupling between partices, but it could be beneficial in some cases

Dale.Huber@sandia.gov

Project Status

- Optimizing particles
- Beginning initial experiments with composites
- Small scale test parts within 6 months
  - Allows optimization
- Within 2-3 years we will have full scale part available for testing in real circuit.

- Manufacturing
  - Lab scale: pouring molds
  - Scale up: Injection molding, reactive injection molding