Lecture 2

The Microscopic Picture of Magnetic Materials

 We will now revisit the experimentally observed magnetic behaviours and try to understand them from a microscopic point of view

Esse material se destina a uso interno e educacional e não deve ser compartilhado. Fica proibida a sua distribuição sob qualquer forma, assim como a postagem em redes sociais, em sites da internet, e equivalentes.

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- Imagine a classical gas of molecules each with a magnetic dipole moment
- In zero field the gas would have zero magnetization



- Applying a magnetic field would tend to orient the dipole moments
- Gas attains a magnetization



- Very high fields would saturate magnetization
- Heating the gas would tend to disorder the moments and hence decrease magnetization



- Theoretical model
- Non-interacting moments
- Boltzmann statistics
- Dipole interaction with B
- Yields good model for many materials
- Examples: ferrous sulfate crystals, ionic solutions of magnetic atoms



- Classical model yields Langevin function
- Quantum model yields Brillouin function



Exchange InteractionDirect exchange

Direct exchange operates between moments, which are close enough to have sufficient overlap of their wavefunctions. It gives a strong but short-range coupling which decreases rapidly as the ions are separated. An initial simple way of understanding direct exchange is to look at two atoms with one electron each. When the atoms are very close together the Coulomb interaction is minimal when the electrons spend most of their time in between the nuclei. Since the electrons are then required to be at the same place in space at the same time, Pauli's exclusion principle requires that they possess opposite spins. According to Bethe and Slater the electrons spend most of their time in between neighboring atoms when the interatomic distance is small. This gives rise to antiparallel alignment and therefore negative exchange. (antiferromagnetic).



If the atoms are far apart the electrons spend their time away from each other in order to minimize the electron-electron repulsion. This gives rise to parallel alignment or positive exchange (ferromagnetism)





Bethe-Slater curve

 The exchange Heisenberg energy, suitably scaled, replaces the Weiss molecular field constant in the mean field theory of ferromagnetism to explain the temperature dependence of the magnetization

$$\mathbf{E}_{p} = -\mathbf{J}_{ex} \mathbf{S}_{i} \times \mathbf{S}_{i+1}$$



Differences in exchange energy of transition metals as due to the ratio of the interatomic distance a to the radius r of the 3d electron shell

Ferromagnetism



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 materials tend to form
 magnetic domains
- Each domain is magnetized in a different direction
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- Applying a field changes domain structure
- Domains with magnetization in direction of field grow
- Other domains shrink



 Applying very strong fields can saturate magnetization by creating single domain



- Removing the field does not necessarily return domain structure to original state
- Hence results in magnetic
 hysteresis

Magnetic domain walls

$\uparrow \uparrow \uparrow \uparrow \uparrow \uparrow \uparrow \land \checkmark \rightarrow \rightarrow \searrow \downarrow \downarrow \downarrow \downarrow \downarrow \downarrow$

Wall Thickness "t"

Wall thickness, t, is typically about 100 nm

Single domain particles



< t

 Particles smaller than "t" have no domains

Antiferromagnetism



- In some materials, exchange interactions favour antiparallel alignment of atomic magnetic moments
- Materials are magnetically ordered but have zero remnant magnetization and very low χ
- Many metal oxides are antiferromagnetic

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Ferrimagnetism



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- Results in net magnetization
- Example: magnetite, maghemite

Small Particle Magnetism

Stoner-Wohlfarth Particle



 Magnetic anisotropy energy favours magnetization along certain axes relative to the crystal lattice

Easy axis of magnetization

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- Uniaxial single domain particle
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Thermally Activated Jump (Classical Behaviour!!)



• Relaxation time:
$$\tau = \tau_0 \exp\left(\frac{K_a V}{k_B T}\right)$$

• theoretical predictions: $\tau_0 = 10^{-9} \div 10^{-10}$ (see later)

Demagnetization rate of an assembly of uniaxial particles

$$-\frac{dM}{dt} = f_0 M e^{-KV/kT} = \frac{M}{\tau}$$

 f_0 : frequency factor ($\approx 10^9 \text{ sec}^{-1}$) τ : relaxation time

Turn-off external field at t = 0 with M_i

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For Co ($K = 4.5 \times 10^6$ ergs/cm³) at room temp. (T = 300 K)

D = 68 Å (V = 1.6 × 10⁻¹⁹ cm³)
$$\frac{1}{\tau} = 10^9 \cdot e^{-(4.5 \times 10^6 \times 1.6 \times 10^{-19} / (1.38 \times 10^{-16} \times 300))} \approx 279.9 \frac{1}{\text{sec}}$$

 $\tau \approx 3 \times 10^{-2} \text{ sec}$

An assembly of such particles would reach thermal equilibrium state ($M_r = 0$) almost instantaneously. No hysteresis

Magnetization Relaxation Two Regimes: **Standard Magnetic** Measurements: $t_m \approx 100$ s $\tau < t_m$ $\tau > t_m$ $\tau > t_m$ Mössbauer: $t_m \approx 10^{-8}$ s Measuring time Blocked Superparamagnetic (time needed to do a measurement) For *t_m*≈ 100 s: Define a critical volume at n constant T (e.g., $RT \equiv T_0$) by $V_{crit} \approx \frac{25k_BT}{K_a}$ requiring $\tau = t_m$: $\ln 10^2$ $\ln \tau = \ln \tau_0 + \frac{K_a V_{crit}}{k_B T_0} = \begin{cases} 1110 \\ \cdots \\ 1n10^{-8} \end{cases} D_{crit} = \left[\frac{6}{\pi} V_{crit}\right]^{\frac{1}{3}}$ ≈ **10**⁻¹⁰

Magnetic blocking temperature

- The magnetic blocking temp, T_b , is the temp below which moment is blocked
- Blocking temperature depends on particle size and timescale of observation
- Larger particles have higher blocking temperatures
- The longer the observation time, the more likely it is that the moment will be observed to flip

Fluctuation timescales, τ



Effect of applied field on single domain particles



- Applying field along easy axis favours moment aligned with field
- Above T_b this results in moment spending more time in lower well
- Particle exhibits time averaged magnetization in direction of field

Superparamagnetism



 Unblocked particles that respond to a field are known as superparamagnetic

Superparamagnetism



- Response of superparamagnets to applied field described by Langevin model
- Qualitatively similar to paramagnets
- At room temperature superparamagnetic materials have a much greater magnetic susceptibility per atom than paramagnetic materials

Superparamagnetism



Superparamagnets are often ideal for applications where...

a high magnetic susceptibility is required

 zero magnetic remanence is required
Ferromagnetic Resonance

FMR is a spectroscopic technique to probe the magnetization of ferromagnetic materials.

Landau-Lifshitz-Gilbert equation:

$$\frac{\partial \vec{M}}{\partial t} = -\gamma (\vec{M} \times \vec{H}_{eff}) + \frac{G}{\gamma M_s^2} \left[\vec{M} \times \frac{\partial \vec{M}}{\partial t} \right]$$





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• peak-to-peak linewidth (apparent or true) ΔB_{pp}	Dynamics of the spin system
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Source: Janusz Typek, Institute of Physics, West Pomeranian University of Technology, Szczecin, Poland

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The dynamics of the magnetic moment of the particle is described by the Landau– Lifschitz equation, and for uniaxial particles the resonance condition is given by (16)

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where K is the effective anisotropy, $L_2 = 1 - 3(L_1/\xi)$, and $L_1 = \coth \xi - 1/\xi$ ($\xi = \mu H_r/kT$). The magnetic moment per particle (μ) is related to the saturation magnetization (I_s) by $\mu = I_s V$, where V is the particle volume. It must be pointed out that Eq. [2] is valid over a wide temperature range, including high temperatures, where the fluctuation field is typically of the order of the anisotropy field.

FMR resonance field

$$H_{\rm r} = (\omega_{\rm r}/\gamma) - (K/I_{\rm s})(3\,\cos^2\theta - 1).$$

Experimental Results



Angular dependence of the resonance field for a magnetic fluid sample of MnFe2O4 with particles having an average diameter of 6.0 nm. The solid lines represent the best fit of the experimental data according to Eq. [4].

Magnetization Curve

| ifmpan.poznan.pl/~urbaniak/Wyklady2014/urbaniakUAM2014L2_magnetic%20measurements.pdf

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Introduction

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Urbaniak Magnetic materials in nanoelectronics..

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Magnetic sensor can be divided according to different criteria:



Introduction – Hall magnetometer

• Lorentz force acting on electrons in a circuit deflects them perpendicularly to drift direction: $\vec{F}_{Lorentz} = q \vec{E} + q \vec{v} \times \vec{B}$



- The build-up of charges on outer limits of the circuit induces Hall voltage which depends on the field strength and is used to sense it.
- The Hall voltage is given by (t-film thickness, R_H-Hall coefficient*):

$$U_y = R_H \frac{I}{t} B_z$$

 The main figure of interest is field sensitivity of the sensor** (for a given driving current I_c):

$$\gamma_{b} = \frac{U_{y}}{B_{z}} = \frac{R_{H}I_{c}}{t}$$

- Semiconductors are used to obtain high sensitivity combined with temperature stability (InAs)
- The Hall sensors have a limited use at high fields and low temperatures (conductivity quantization)

image from Wikimedia Commons; authors: Peo (modification by Church of emacs)



Jefferson F. D. F. Araujo, et al. J. Magn. Magn. Mater. 426, 159-162 (2017).

- Sensibilidade 3.5x10⁻⁷ Am² ٠
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Magnetômetro Portátil •



Soudabeh Arsalani, et al. J. Magn. Magn. Mater. 475, 458-464 (2019).

Modelo

Modelo Teórico

$$B_{z}(x, y, z) = \frac{\mu_{0}I}{4\pi} \int_{0}^{2\pi} \frac{xasen\varphi}{r^{3}} d\varphi$$
$$B_{z}(x, y, z) = \frac{\mu_{0}m}{4\pi} \left[\int_{-L/2}^{L/2} \int_{0}^{2\pi} \frac{xasen\varphi}{r^{3}} d\varphi dx \right] / \pi a^{2-2\pi}$$







Jefferson F. D. F. Araujo, et al. J. Magn. Magn. Mater. 426, 159-162 (2017).

✤ Automação





Magnetômetro Baixa Temperatura



Jefferson F. D. F. Araujo, A. C. Bruno, and S. R. W. Louro, *Review of Scientific Instruments* 86, 105103 (2015).



- Sensibilidade: 8.5x10⁻⁸ Am²
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- T_{mim}: **6.0 K**

Projeto de Pesquisa



- Sensibilidade: 8.5x10⁻⁸ Am²
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Microscópio Magnético de Varredura •



Desenvolvido de forma pioneira





Jefferson F. D. F. Araujo, et al. J. Magn. Magn. Mater. 499, 166300-9 (2020).





Projetos Futuros



Magnetic Separation



Magnetic separation setup (magnetophoresis device)



Schematic setup of magnetophoresis device (top view) contains three cylindrical cavities, two of them for 2 mL volume tubes and one for 15 mL tube. (b) The MS process for one tube is illustrated.

Magnetophoresis curve and DLS



(a) The magnetophoresis curve of EM10 over a 14 h time period, and (b) DLS of EM10 sample at t_0 before separation, t_1 after 3 min, t_2 after 50 min, and t_3 after 3h of inserting sample in separation system.

Magnetic Particle Spectrometer

Single driving field-based MPS



(a) Time varying sinusoidal magnetic field; (b) MH response curve of SPIONs; (c) time domain magnetization response of SPIONs; (d) power spectrum of collected signal contains higher harmonic components 3*f* (third harmonic), 5*f* (fifth harmonic), etc

Picture from: J. Phys. D: Appl. Phys. 52 (2019) 173001 (17pp)

Magnetization Coil Electrical and Geometrical Specifications

 R_1 = 35 mm R_2 = 41.5 mm R_3 = 52 mm I=160mm N= 1.360 turns L=20.6 mH R=8.7Ω C= 12.3nF





https://www.translatorscafe.com/unitconverter/fr/calculator/series-rlc-impedance/

Magnetic Characteristics

$$\oint _{C} \mathbf{B} \cdot d\mathbf{l} = \mu_0 I$$

$$\mu_0 = 4\pi \times 10^{-7} \frac{N}{A^2}$$
N= 1360 turns
L= 160 mm
B= 10.7 mT/A

Experimental Checks

Search coil positioned at the center of the magnetization coil N= 10 turns R= 5 mm $V_{mag-coil} = 74Vpp$ $V_{search-coil} = 81 mVpp$ $B_{exp} = 0.60 mT/A$

Excitation, detection & monitoring coils

Detection coils, 1st order gradiometer Monitoring coil (center) Excitation coil-

solenoid with two layers of winding.



Resonance of Driving Coil

• The RLC circuit is usually driven at the resonance frequency to optimize power transfer from the power amplifier



Power Amplifier

A power amplifier with low level of distortion must be employed in the excitation circuit.



Dual driving field-based MPS

(e) dual sinusoidal magnetic fields; (f) MH response curve of SPIONs; (g) time domain magnetization response of SPIONs; (h) power spectrum of collected signal contains higher harmonic components $fH \pm 2fL$ (third harmonics), $fH \pm 4fL$ (fifth harmonics).





FIG. 1. The nanoparticle suspension is driven by a time-varying magnetic field provided by the excitation field. The sample magnetization change induces a voltage in a pick-up coil system which is recorded by a DAQ.

Published in: Nicolas Garraud; Rohan Dhavalikar; Lorena Maldonado-Camargo; David P. Arnold; Carlos Rinaldi; *AIP Advances* 7, 056730 (2017) DOI: 10.1063/1.4978003 Copyright © 2017 Author(s)



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 $\mathbf{E} = -\mathbf{m} \mathbf{B} \cos[\theta]$

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- Curie temp is a measure of exchange interaction strength
- Note: exchange interactions much stronger than dipole-dipole interactions



- Ferromagnetic materials tend to form magnetic domains
- Each domain is magnetized in a different direction
- Domain structure minimizes energy due to stray fields



- Applying a field changes domain structure
- Domains with magnetization in direction of field grow
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 Applying very strong fields can saturate magnetization by creating single domain



 Removing the field does not necessarily return domain structure to original state

 Hence results in magnetic hysteresis Magnetic domain walls

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Wall thickness, t, is typically about 100 nm

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Thermal activation



- At low temperature magnetic moment of particle trapped in one of the wells
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Thermally Activated Jump



Thermally Activated Jump (Classical Behaviour!!)



• Relaxation time:
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• theoretical predictions: $\tau_0 = 10^{-9} \div 10^{-10}$ (see later)

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Magnetization Relaxation

Two Regimes:



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Fluctuation timescales, τ



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H=0

 $H=H_1$

 $H=H_2$





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ifmpan.poznan.pl/~urbaniak/Wyklady2014/urbaniakUAM2014L2_magnetic%20measurements.pdf



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M. Urbaniak Magnetic materials in nanoelectronics...





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image from Wikimedia Commons; authors: Peo (modification by Church of emacs)

**some tenths of mV per kA/m for Ic of several mA (www.lakeshore.com/products/Hall-Magnetic-Sensors/pages/Specifications.aspx)



Jefferson F. D. F. Araujo, et al. J. Magn. Magn. Mater. 426, 159-162 (2017).

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Soudabeh Arsalani, et al. *J. Magn. Magn. Mater.* 475, 458-464 (2019).

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Desenvolvido de forma pioneira

Jefferson F. D. F. Araujo, et al. *J. Magn. Magn. Mater.* 499, 166300-9 (2020).







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Magnetic separation setup (magnetophoresis device)



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L=20.6 mH

 $R=8.7\Omega$

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 $\phi = 2.59002 \circ = 0.0452$ rad

ohm (Ω)

millihenry (mH) 🔻

nanofarad (nF)

kilohertz (kHz) 🔹

Partager

ω= 62831.853 rad/s

X_C= 1.29394 kΩ

 $X_{I} = 1.29434 k\Omega$

Q= 148.75165

|Z_{RLC}|= 8.7089 Ω

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https://www.translatorscafe.com/unit-converter/fr/calculator/series-rlcimpedance/

Magnetic Characteristics

$$\oint {}_C \mathbf{B} \cdot d\mathbf{l} = \mu_0 I$$
 $\mu_0 = 4\pi imes 10^{-7} rac{\mathrm{N}}{\mathrm{A}^2}$

L= 160 mm

B= 10.7 mT/A

Experimental Checks

Search coil positioned at the center of the magnetization coil N= 10 turns R= 5 mm $V_{mag-coil} = 74Vpp$ $V_{search-coil} = 81 mVpp$ $B_{exp} = 0.60 mT/A$

Excitation, detection & monitoring coils

Detection coils, 1st order gradiometer

Monitoring coil (center)

Excitation coil-solenoid with two layers of winding.



Resonance of Driving Coil

• The RLC circuit is usually driven at the resonance frequency to optimize power transfer from the power amplifier



Power Amplifier

A power amplifier with low level of distortion must be employed in the excitation circuit.



Dual driving field-based MPS

(e) dual sinusoidal magnetic fields; (f) MH response curve of SPIONs; (g) time domain magnetization response of SPIONs;
(h) power spectrum of collected signal contains higher harmonic components *fH* ± 2*fL* (third harmonics), *fH* ± 4*fL* (fifth harmonics).





FIG. 1. The nanoparticle suspension is driven by a time-varying magnetic field provided by the excitation field. The sample magnetization change induces a voltage in a pick-up coil system which is recorded by a DAQ.

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