Magnetic Resonance(s) European School on Magnetism, Brno 2019

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This lecture will last 90 minutes $\pm 180.$ Should I rush?



- Dynamics of one (electronic) spin
 - Electron Spin Resonance (ESR)
- Ferromagnetic dynamics
 - Ferromagnetic Resonance (FMR)
 - Uniform, Non uniform Modes
 - Magnetic Objects : Domain Wall, Vortex
- Mossbauer, NMR

Classical Magnetometry = Magnetic Moment m(H,T,angle ...)

An alternative is to detect a Magnetic Resonance to determine :

- resonance field (or resonance frequency)
- amplitude of the resonance
- width of the resonance line

as a function of external parameters (Applied Magnetic Field, Geometry, Temperature ...)

and hopefully to extract interesting magnetic infos

A magnetic atom is characterised by its quantum number J (could be J=S only)

The atom angular momentum is $L = \hbar J$. The atom magnetic moment is $m = g \mu_B J$.

The gyromagnetic factor is the ratio magnetic moment / angular momentum i.e.

$$\gamma = -\frac{g\mu_B}{\hbar} = -\frac{ge}{2m_e}$$

 γ is negative (today), both moments are antiparallel.

UNLIKE the usual choice in spintronics where electrons magnetised up are labelled spin up (and in reality are spin down).

Larmor precession

When a magnetic field H is applied, a Zeeman energy appears :

$$E_z = -\mu_0 \vec{m}.\vec{H}$$

The torque $\vec{\Gamma}$ which is applied to the magnetic moment is :

$$ec{\mathsf{\Gamma}}=ec{m}\wedge\mu_0ec{H}$$

The torque corresponds to a change of angular momentum :

$$\vec{\Gamma} = \frac{d\vec{L}}{dt}$$

Finally : $\frac{d\vec{m}}{dt} = \gamma \vec{m} \wedge \mu_0 \vec{H}$

The equation corresponds to a precession around the applied field : Larmor Precession Angle $(\vec{m}, \vec{H}) = \text{constant}$ (constant energy, no relaxation).

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Electron Spin Resonance (ESR)



In an applied magnetic field H, a spin 1/2 can absorb a photon = electromagnetic wave

$$h\nu = g\mu_B\mu_0H$$

To allow the transition, the ac field from the wave must be

perpendicular to the applied field $(S_z \text{ is not anymore an eigen state})$

$$\frac{d\vec{m}}{dt} = \gamma \vec{m} \wedge \mu_0 \vec{H}$$
$$h\nu = g\mu_B \mu_0 H$$

- if we know ν and g : field sensor, calibration
- if we know ν and H : g sensor, orbital/spin contributions, chemical infos

Applied Field $\mu_0 H_z$, J=S= $\frac{1}{2}$

$$\Delta E = g\mu_B\mu_0 H_z = h\nu$$

When g=2 ν =10 GHz gives resonance at 0.357 Tesla ν =100 MHz gives resonance at 3.57 mTesla

MHz electronics = small field resonance (mT) GHz microwave = Tesla field $\mu_0 H_{res}$ (electromagnets and superconducitng coils)

Measurement

Two types of setups/measurements to get the resonance At fixed field, scan the frequency, detect absorption At fixed frequency, scan the field, detect absorption



The absorption line or its derivative is measured

Extract resonance field, amplitude of absorption, width of the line (peak-to-peak)

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10-100 MHz ESR for practicals Helmholz Coils for the applied field

What to learn from ESR?



Amplitude of the resonance signal : number of resonating species Position of the resonance line :

- shift from free electron g-factor
- environment effects on the orbitals (orbital moment is impacted)
- interaction with nucleus moment (hyperfine splitting of the energy lines)

Line width :

- intrinsic (narrow)
- distribution of g-factors (wider line)
- inhomogeneous field, sample

DPPH calibration



DPPH : 2,2-Diphenyl-1-picrylhydrazyl (free radical) Chemical Formula $C_{18}H_{12}N_5O_6$

For a free electron g = 2.0023 (2 + correction QED) For DPPH (very light atoms, small SO), g = 2.0035 $\gamma = 1.76 \ 10^{11} Hz/T (28.041 \ \text{GHz/T})$

The resonance field at 9.750 GHz for DPPH is 0.347703 T (g=2.0035)

For DPPH, g factor varies from 2.003 to 2.0045 depending on preparation and environment (solvant in particular).

Hyperfine Structure



The electronic spin feels the field of the nuclear spin. It is then a (S=1/2,I=1/2) system : singlet state + triplet state Singlet-triplet degeneracy is lifted when the field is applied

ESR and magnetometry

NV center magnetometry



Balasubramanian et al. NatMat2009



Ta/CoFeB (1.5 nm)/MgO, perpendicular M.

In a ferromagnetic material :

The volume torque acting on magnetisation is : $\vec{\Gamma} = \mu_0 \vec{M} \wedge \vec{H}_{eff}$.

The variation of volume angular momentum is $\frac{d\vec{L}}{dt} = \vec{\Gamma}$

$$rac{dec{M}}{dt} = \gamma ec{\Gamma}$$
, $\gamma < 0$
So, $rac{dec{M}}{dt} = \gamma ec{\mu_0} ec{M} \wedge ec{H}_{eff}$
Similar to Larmor

Experimentally Larmor precession does not last forever and the magnetisation aligns with the field Need relaxation

Landau-Lifshitz (LL) equation (1935) :

$$\frac{d\vec{M}}{dt} = \mu_0 \gamma \vec{M} \wedge \vec{H} + \alpha \frac{\vec{M}}{Ms} \wedge (\vec{M} \wedge \vec{H})$$

Landau-Lifshitz-Gilbert (LLG) equation (1955)

$$\frac{d\vec{M}}{dt} = -\mu_0 \gamma \vec{M} \wedge \vec{H} + \alpha_{LLG} \frac{\vec{M}}{Ms} \wedge \frac{d\vec{M}}{dt}$$

 α_{LLG} has no unit Typically 0.001 (low damping) to 0.1 (fast relaxation)

Mathematically, one can transform LL into LLG : same dynamics.

Relaxation of the magnetic moment

The field is applied along z. In NMR, Bloch equations (1946) are used and take into account two relaxations The relaxation of m_z : longitudinal relaxation Characteristic time T_1

$$rac{dec{m_z}}{dt} = \gamma ec{m} \wedge \mu_0 ec{H} - rac{m_z - m_{sat}}{T_1}$$

Energy needs to be transferred out, to the lattice (spin-lattice relaxation time)

The relaxation of m_x and m_y : transverse relaxation, in Bloch equations : Characteristic time T_2

$$\frac{d\vec{m}_x}{dt} = \gamma \vec{m} \wedge \mu_0 \vec{H} - \frac{m_x}{T_2}$$

Energy stays in the spin system (spin-spin relaxation time)

In ferromagnets, LL and LLG introduced a phenomelogical constant (isotropic) relaxation, characterised by damping constant α .

Its value is measured from resonance experiments or relaxation experiments

Its minimum theoretical value is not zero (intrinsic damping) can be evaluated from band structure calculation.

Low alpha material



Co₂FeAl, effect of annealing. Low damping when well ordered.

- intrinsic
- inhomogeneous sample
- eddy currents
- Spin Transfer Torque
- Spin Pumping

FMR : allows to compare series of samples (with thickness or growth conditions)

Easier to estimate homogeneity than Magnetometry (think of a M(H) loop)

skin depth (conductivity dependent) Typically 100 nm at 10 GHz

negligible for ultrathin films (the field penetrates) eddy current : losses, heating (microwave oven).

Water-based large samples (bio environment) \Rightarrow need to go to lower frequency for ESR



Spin Transfer Torque : acts as an extra contribution to LLG

$$rac{dec{M}}{dt} = \mu_0 \gamma ec{M} \wedge ec{H} + (lpha_{LLG} + lpha_{STT}) rac{ec{M}}{Ms} \wedge rac{dec{M}}{dt}$$

 $\alpha_{\textit{STT}} \propto j.\textit{P} = \text{electron}$ flow. spin polarisation

- Increase damping (faster relaxation, less ringing)
- Decrease (Cancel) damping (spin pumping, STT oscillators)

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Landau-Lifschitz-Gilbert equation

$$\frac{d\boldsymbol{M}}{dt} = \gamma \boldsymbol{M} \times \boldsymbol{H}_{eff} + \alpha \boldsymbol{M} \times (\boldsymbol{M} \times \boldsymbol{H}_{eff})$$
(1)

 H_{eff} the effective field :

$$m{H}_{eff} = -rac{1}{\mu_0}rac{\partial E}{\partial m{M}}$$

The effective field includes contributions from the applied field

(Zeeman energy), the demagnetising field (shape anisotropy), magnetocrystalline and exchange energies.

Once the external parameters are known (geometry, field).

- Write the Total energy : $E_Z + E_d + E_{mc} + E_A$
- Look for \vec{M} equilibrium position
- Determine the shape of the energy minimum (curvature)
- Determine the natural frequency of the oscillations around the equilibrium position

Resonance Fields

Linear development around the equilibrium position of \vec{M} Free energy F (spherical coordinates), $\vec{M} = M_s \vec{u}_r$:

$$dF = -\mu_0 d\vec{M}.\vec{H}_{eff} = d\vec{M}.\frac{\partial F}{\partial \vec{M}}$$

$$Heff_{\theta} = -\frac{1}{\mu_0 M_s} \frac{\partial F}{\partial \theta}, Heff_{\phi} = -\frac{1}{\mu_0 M_s sin\theta} \frac{\partial F}{\partial \phi}$$

(all steps in Baselgia et al. PRB 1988)

$$\left(\frac{\omega^2}{\gamma^2}\right) = \frac{1}{M_s^2} \left(F_{\theta\theta} \left(\frac{F_{\phi\phi}}{\sin^2\theta} + \frac{\cos\theta}{\sin\theta}F_{\theta}\right) - \left(\frac{1}{\sin\theta}F_{\theta\phi} - \frac{\cos\theta}{\sin^2\theta}F_{\phi}\right)^2\right)$$

Line-width FWHM :

$$\Delta H = rac{lpha}{d\omega/dH} rac{\gamma}{M_s} (F_{ heta heta} + rac{1}{sin^2 heta} F_{\phi\phi})$$

Uniform film, not patterned Only Zeeman + Shape anisotropy :

Field in-plane :
$$B_{res} = \frac{\sqrt{(\mu_0 M)^2 + 4* \frac{\omega^2(1+\alpha^2)}{\gamma^2} - \mu_0 M}}{2}$$

At 10 GHz : $B_{res} = \frac{\sqrt{(\mu_0 M)^2 + 4*0.34^2} - \mu_0 M}{2}$

Example : Soft Film



FMR gives M (magnetisation) magnetometry (VSM-SQUID) gives m (magnetic moment) Useful to compare

Uniform vs Non Uniform modes



Uniform Mode : all spin precess together



Non Uniform Mode : FMR can excite non uniform modes

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A coupled Bilayer can give rise to acoustic (in phase) and optical (out-of-phase) modes

Cavity (fixed f, scanning H) vs BroadBand (scanning f)

- X-band (10 GHz), generator, waveguide, detection scheme (diode) ...
- Broadband Source (VNA), waveguide, VNA

Experimental set-ups



Fixed frequency, Resonating Cavity FMR

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| Band | Frequency (GHz) | Wavelength (cm) | B_{res} (T) free electron |
|------|-----------------|-----------------|-----------------------------|
| S | 2-4 (3) | 10 | 0.11 |
| Х | 8-12 (9.5) | 3 | 0.34 |
| K | 18-27 | 1.3 | 0.8 |
| Q | 35 | 0.86 | 1.3 |
| W | 75-110 (95) | 0.31 | 3.3 |

Experimental set-ups



9.8 GHz (X-band) resonant cavity (mode TE_{102})

Older Technology : Klystrons

it is a type of vacuum tube. A beam of electrons is shaped into ac current and then coupled to transfer its energy to an electromagnetic wave.

Newer Technology : Gunn diode

Solid State technology. Negative differential resistance which makes total resistance zero and creates oscillations.







rectangular cavity. Applied field is along short direction. E_{rf} is minimum in center, B_{rf} is maximum. (! pumping direction)

- RF diode + lock-in
- integrated analyser (power meter)
- Electric detection (small sample)

10 GHz cavity Best sensitivity : 10^{12} spins (narrow line, not hyperfine splitted). If the FMR resonance line has a Lorentzian shape :

$$f(B) = \frac{A}{\frac{(B-B_r)^2}{D^2} + 1}$$

Line amplitude : A. Line center : B_r . FWHM=2D

$$\frac{df}{dB} = \frac{-2A(B-B_r)}{D^2(\frac{(B-B_r)^2}{D^2}+1)^2}$$

Peak to peak width of the line derivative (FMR measurement)

$$\Delta B_{pp} = \frac{2D}{\sqrt{3}} = \frac{FWHM}{\sqrt{3}}$$

Choosing the applied field is often necessary Frequency must be swept to find the resonance Generator, waveguide, sample holder must accept a broadband signal.



Vector Network Analyser (wave out, wave in). The phase of the electric wave can be analysed (not only the intensity).

Connecting the Sample



Signal is transmitted using coaxial cables, coax connectors, microbonding to striplines / coplanar waveguide

Connecting the Sample



RF Probes can also be used to mechanically connect the sample.

Raw signal



the signal is an electromagnetic wave. It can be reflected at many interfaces.

A cable is a resonating object.

Evaluating the electric field at the sample is a complicated task

Electric Detection of FMR



Bolometric response : Heating of the sample when Wave is absorbed.

Domain Wall-FMR



A domain wall in a potential minimum is equivalent to a mass in a potential well

Some nuclei carry a nuclear spin I Since $\frac{m_p}{m_e}$ =2000 for a nucleus $\gamma_n = -\frac{ge}{2m_n}$ is 2000 times smaller compared to γ_e NMR frequencies (MHz) are 2000 smaller compared to EPR frequencies (GHz) Similar interest to go to high frequency to improve resonance line resolution Co/Cu multilayer : where is Co?



NMR in Spin-echo mode. Sensitivity 10¹⁶ Co spins Environment (Co-Cu mixing) and strain (line shift) See P. Mendels' presentation ESM2011 for more details about NMR

Mossbauer : Principle



Mossbauer : Fe57



6 allowed transitions when a gamma photon is absorbed (ground state I=1/2, excited state I=3/2)

Mossbauer

FeRh : Are all Fe equivalent?



Need Fe⁵⁷ ($S_n=1/2$) in the sample (only 2% in natural Fe). here 150nm thick films Get local field on the nucleus (line splitting) Get local field direction (ratio of lines intensities) laurent ranno@neel.cnrs.fr Magnetic Resonances ESM2019Brno 57 / 58 Bland-Heinrich Book series (Ultrathin Magnetic Structures I,II, III)

Hillebrands, Ounadjela/Thiaville Book series (Spin Dynamics in Confined Magnetic Structures)

Review (70 pages) on metallic films FMR : M. Farle, Rep. Prog. Phys. 61, 755 (1998)