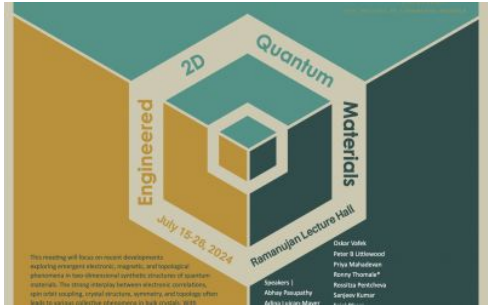
Magnetoimpedance Spectroscopy of Oxides

R. Mahendiran
Department of Physics,
National University of Singapore,
Singapore

E-mail: phyrm@nus.edu.sg



National University
of Singapore

<u>Contributions from</u>

Dr. Amit Chanda
Dr. Ushnish Chaudhuri
Dr. Lee Yang Heng

@ICTS, July15-27, 2024

Spintronics and Magnetic Materials laboratory @ NUS, Physics

What are we doing?

1. Magnetoresistive, Multicaloric, Magnetostrictive, and thermoelectric properties
2. High-frequency magnetoresistance
3. Spin Caloritronics (Nernst and Spin Seebeck effects, Magnetothermopower)
4. Impacts of microwave irradiation on synthesis and physical properties of oxides

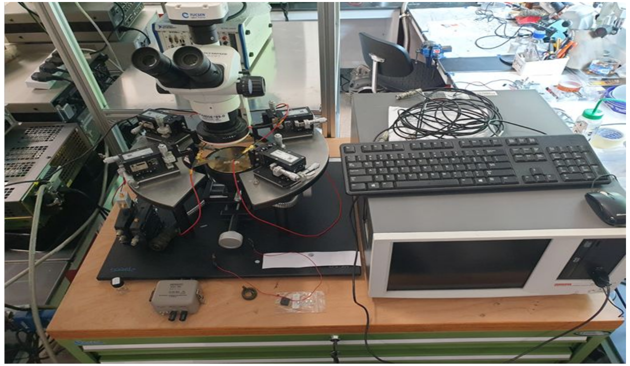
Thermopower & Resistivity

VNA based FMR spectrometer

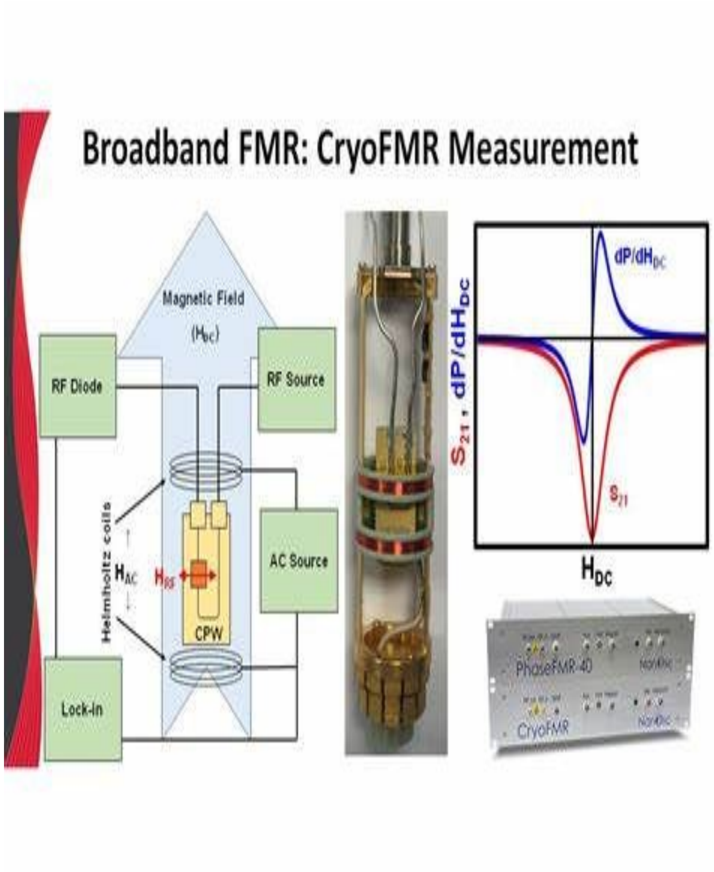
Spin-thermoelectrics,
Magnetoimpedance
Magnetoelectrics

Vibrating Sample magnetometer

Superconducting magnet (PPMS)



Cryogenic FMR spectrometer



Broadband FMR: CryoFMR Measurement

f = 2 to 18 GHz, T = 400 - 5 K

Capacitance dilatometer

T = 310 -5 K

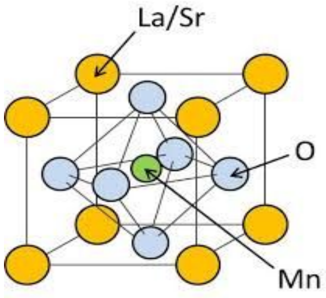
Synthesis by microwave irradiation & sintering

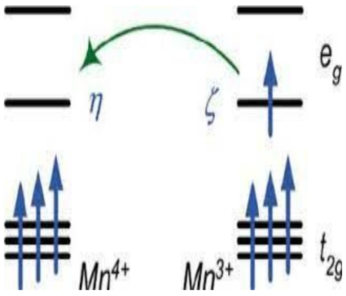
Outline

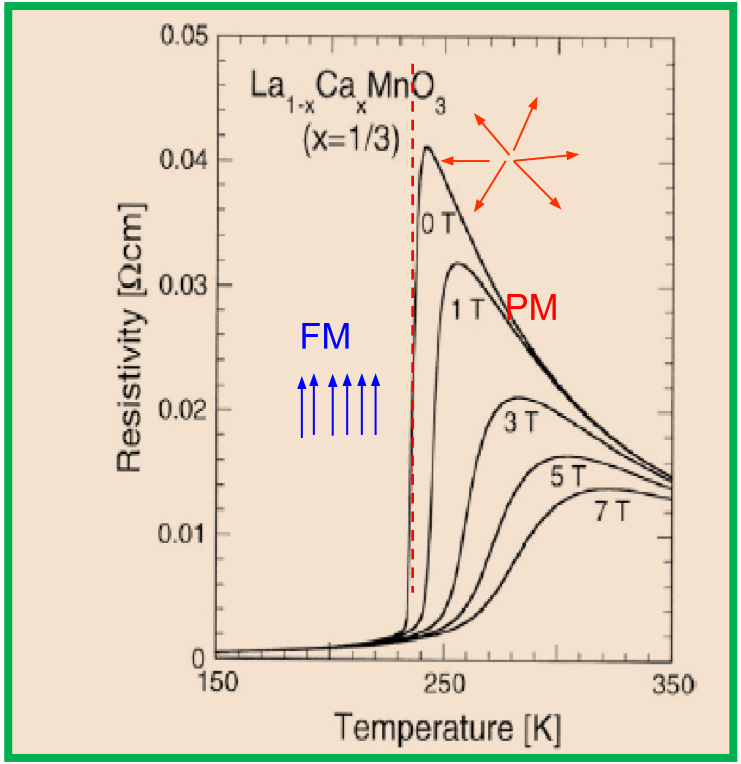
- Magnetoimpedance (MI) in metallic Oxides: $R_{0.6}Sr_{0.4}MnO_3$ (R = Nd, Pr, La)
- MI in insulating Oxides: La_2NiMnO_6, YIG, and also DPPH
- Relevant to recent studies in magnetic/nonmagnetic bilayers

Colossal DC Magnetoresistance in Mn-Perovskites

LaMn^{3+}O$_3$:Antiferromagnetic Insulator (T$_N$ = 140 K)

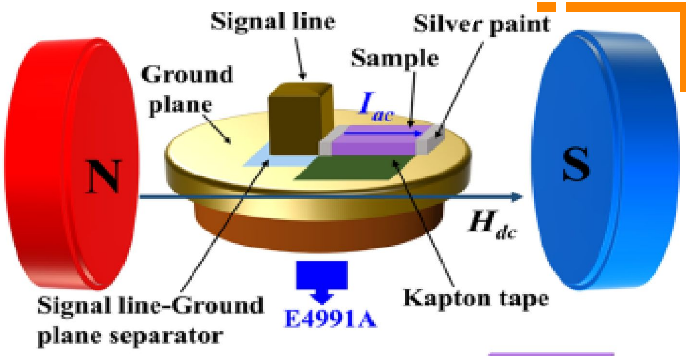






- ○ MR is negative and isotropic
- ○ CMR for H = 30-50 kOe but smaller -MR (< 5%) for H < 1 kOe.
- ○ MR with direct-current (dc MR) is widely reported.
- ○ Small polaron hopping for T > T$_C$ to metallic conduction for T < T$_C$.
- ○ How does electron hopping, hence, the MR respond to an oscillating current?

Experiment: Magnetoimpedance



Electrical Impedance: $Z(f,H_{dc}) = R(f, H_{dc})$ $jX(f, H_{dc})$

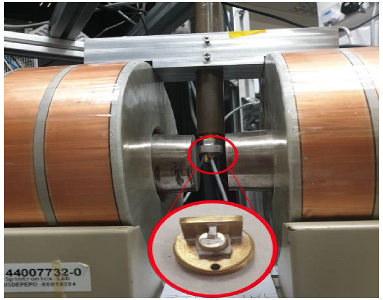
Ac Resistance

Reactance

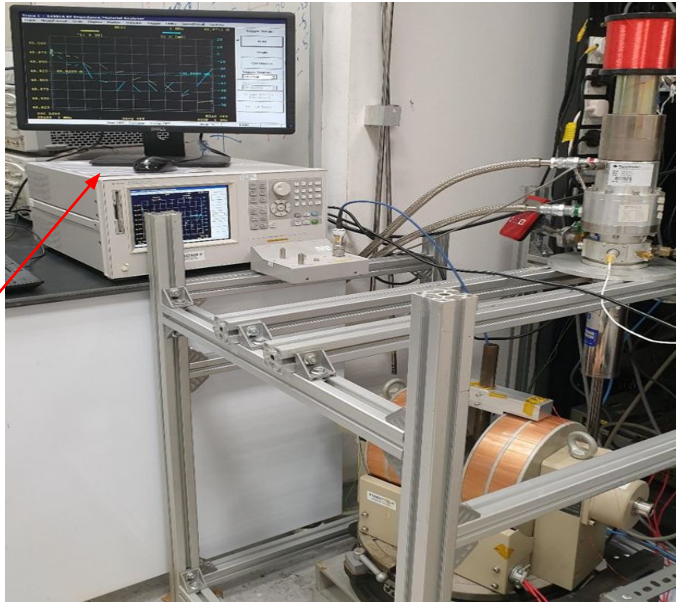
f = 1 MHz to 3 GHz

Ac Magnetoresistance : $MR_{ac} = \Delta R/R = [R(H)-R(0)]/R(0)$

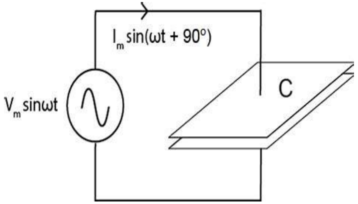
Magnetoreactance : $MX = \Delta X/X = [X(H)-X(0)]/X(0)$



Agilent E4991 RF Impedance analyzer

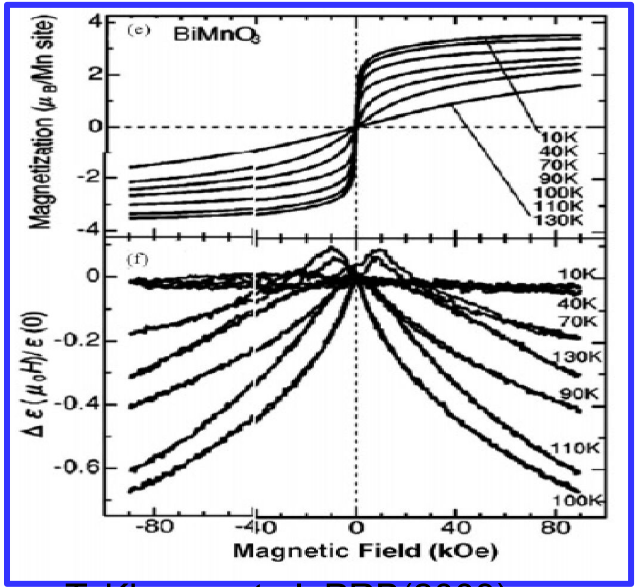


Magnetocapacitance in a multiferroic oxide: BiMnO$_3$



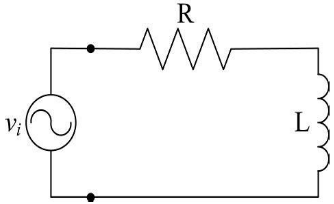
For a leaky capacitor

$Z = [1/R+/i\omega C]^{-1}$



T. Kimura et al. PRB(2003)

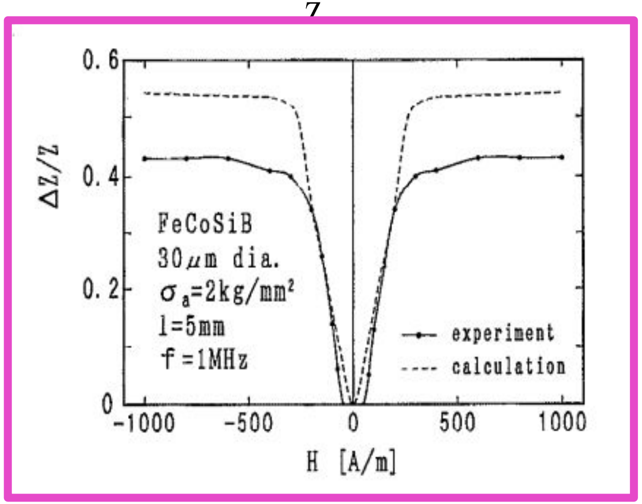
Magnetoimpedance in amorphous ferromagnets



For a metal

$Z = R+i\omega L$

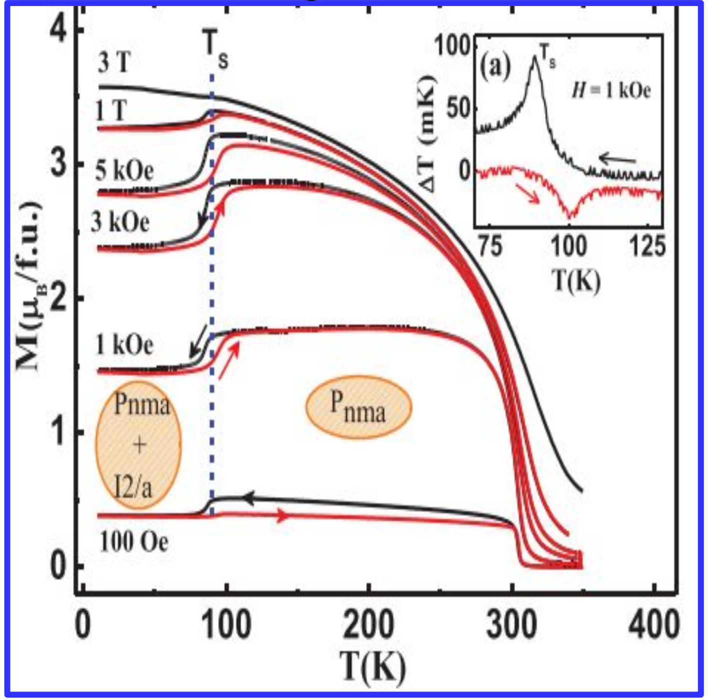
Z

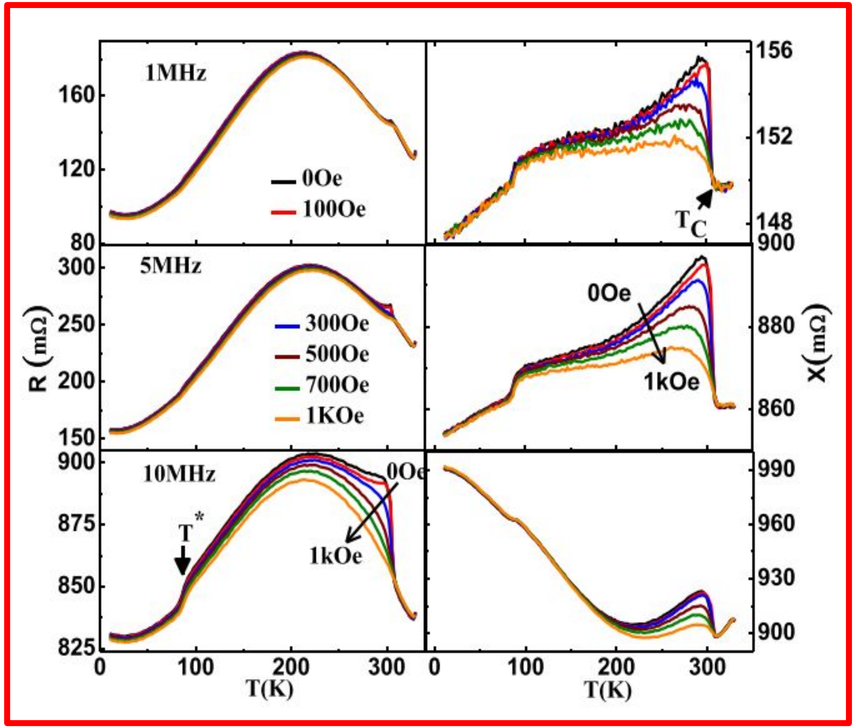


L. Panina, APL, 65,1190 (1994)

AC Electrical resistivity of $Pr_{0.6}Sr_{0.4}MnO_3$ in H = 0 and H < 1 kOe

T_C = 305 K





R. Mahesh et al., J. Appl. Phys. 112, 123915 (2012)

Reactance (X) for f=1 & 5 MHz mimics the ac χ

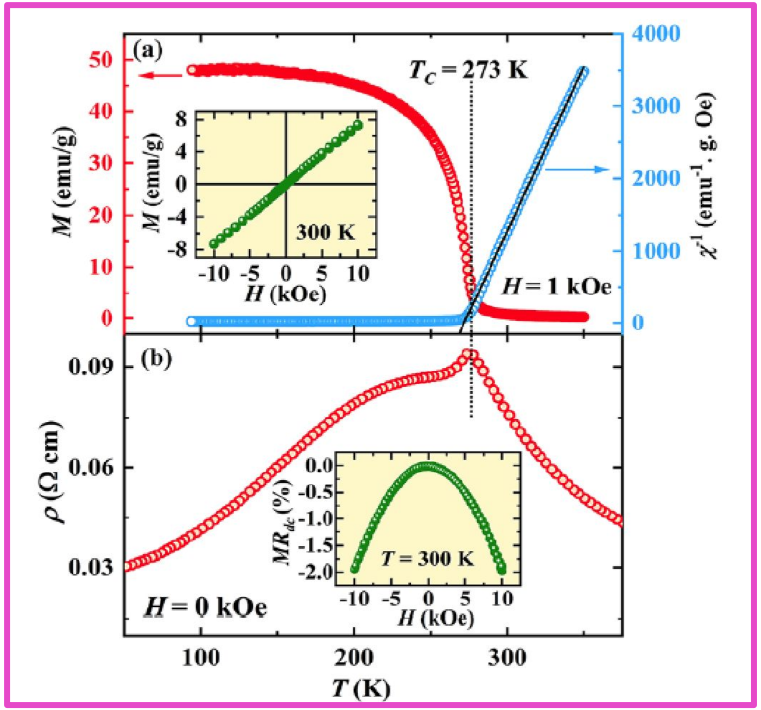
Magnetism and dc electrical resistivity in $R_{0.6}Sr_{0.4}MnO_3$ (R= Pr,Nd)

$Nd_{0.6}Sr_{0.4}MnO_3$

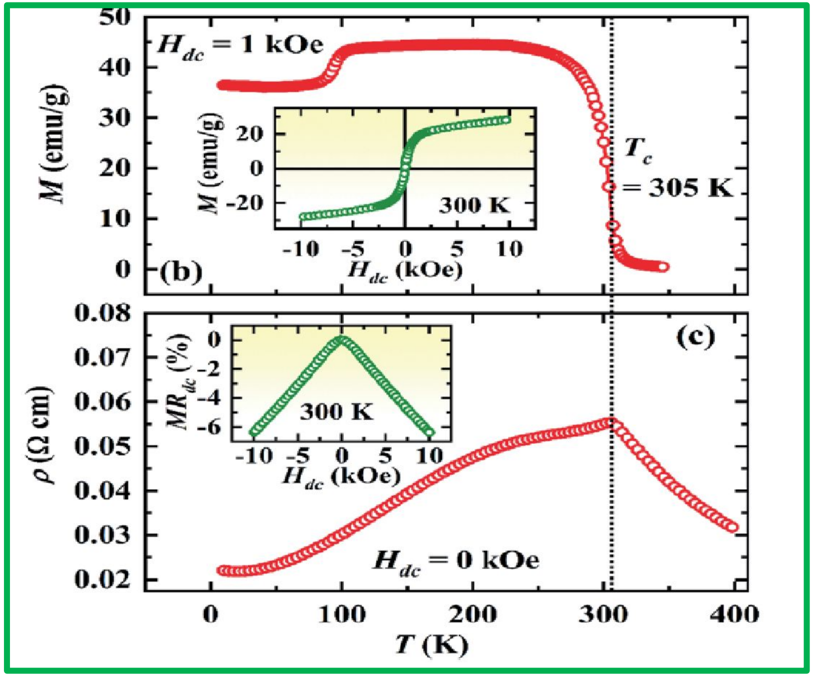
Paramagnetic @ 300 K

$Pr_{0.6}Sr_{0.4}MnO_3$

Ferromagnetic @ 300 K



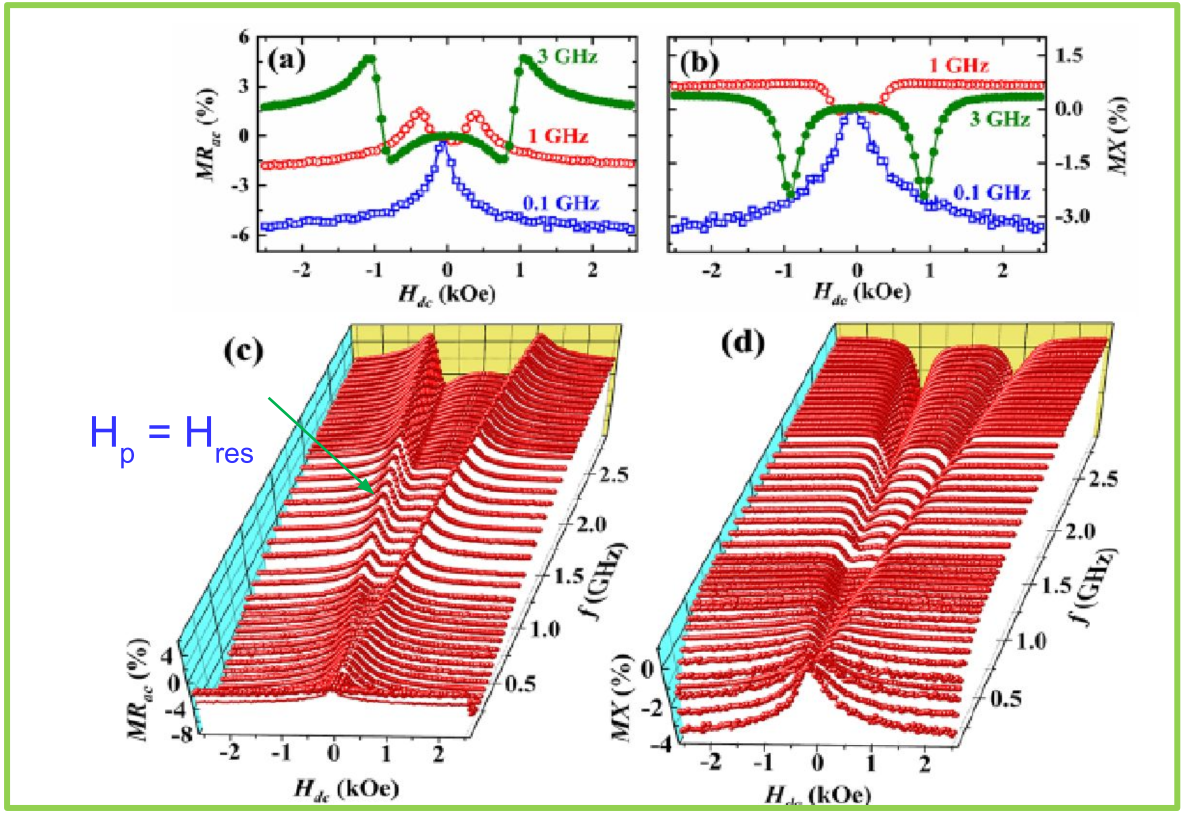
Dc MR = -2% for H = 1 T



Dc MR = -6% for H = 1 T

Ac MR in paramagnetic $Nd_{0.6}Sr_{0.4}MnO_3$

Ac Magnetoresistance Magnetoreactance



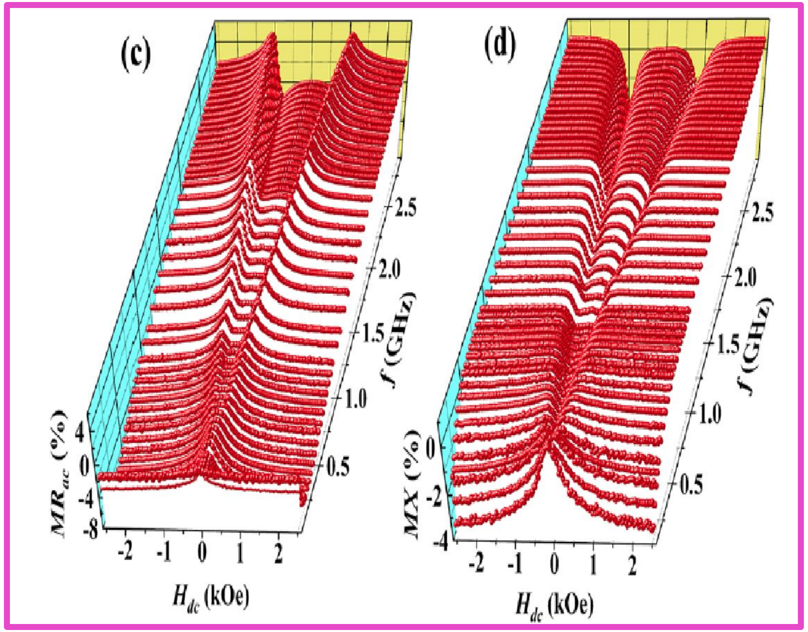
- Single peak in MR at H = 0 for f < 0.9 GHz
- Double peaks at $H_{dc} = \pm H_p$ for higher f,
- H_p shifts up as *f* increases.
- Sign of MR_{ac} changes as f increases
- MR_{dc} = -2% at 10 kOe
- MR_{ac} = +5 % for 2.5 kOe @ 3 GHz

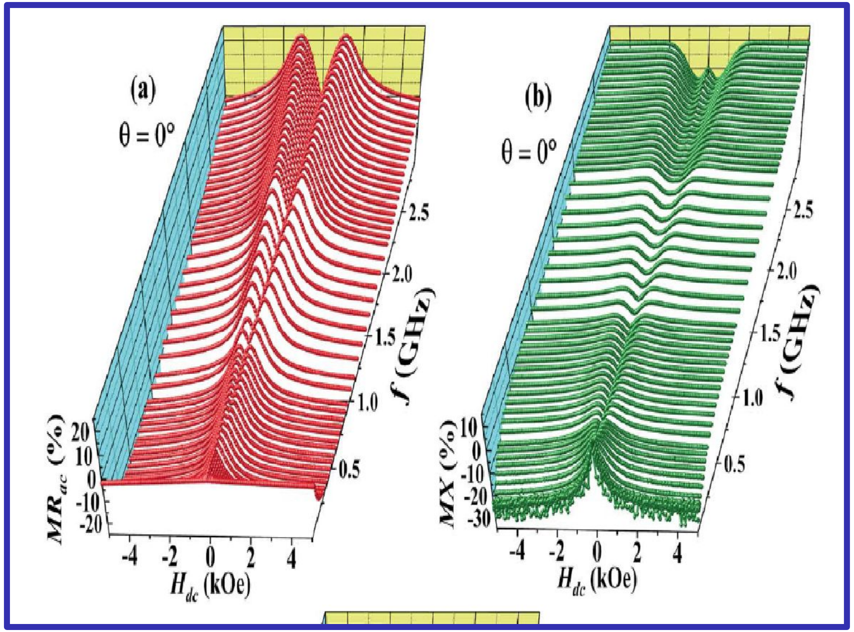
A. Chanda and R. M, J. Phys. Chem. C 124, 18226 (2020)

Ac MR in paramagnetic vs ferromagnetic samples

$Nd_{0.6}Sr_{0.4}MnO_3$ (Paramagnetic)

$Pr_{0.6}Sr_{0.4}MnO_3$ (Ferromagnetic)



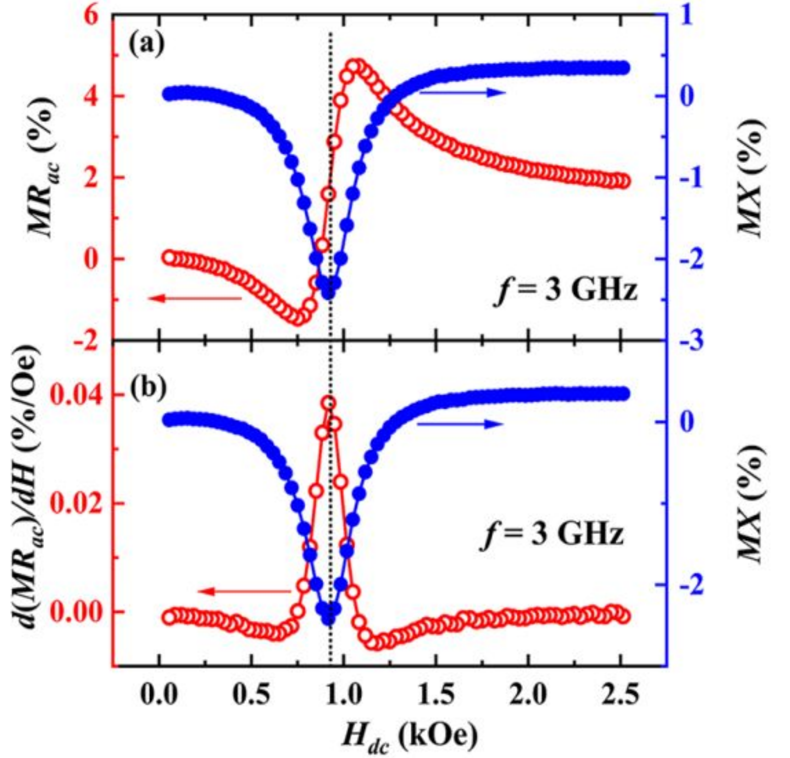


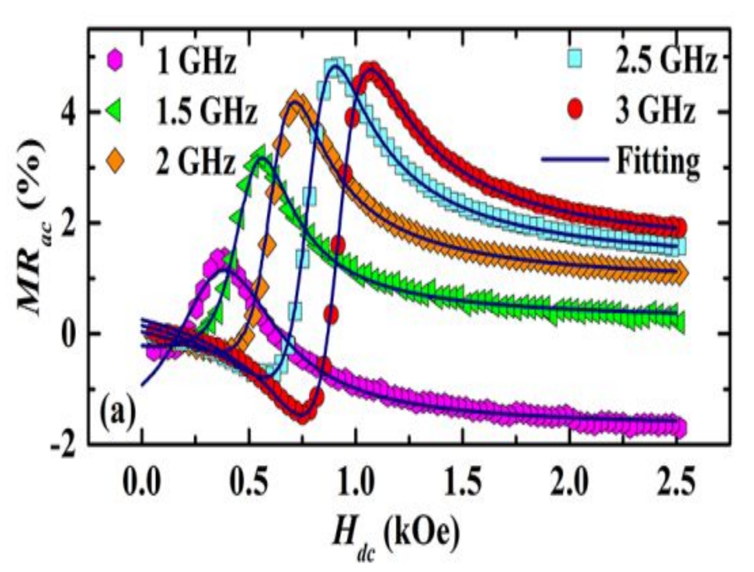
The “canal” in MR_{ac} in FM sample is narrower than in PM sample.

MX shows a distinct peak around the zero field in the FM and PM sample

Features in MR and MX are interconnected

It is sufficient to analyze either MR or MX

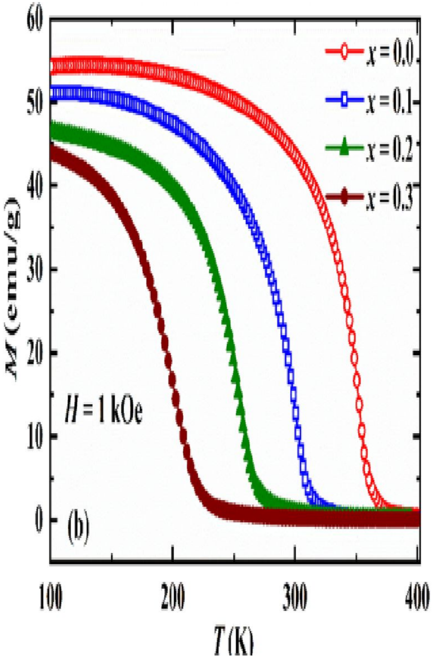




A model fit using 2 Lorentizians

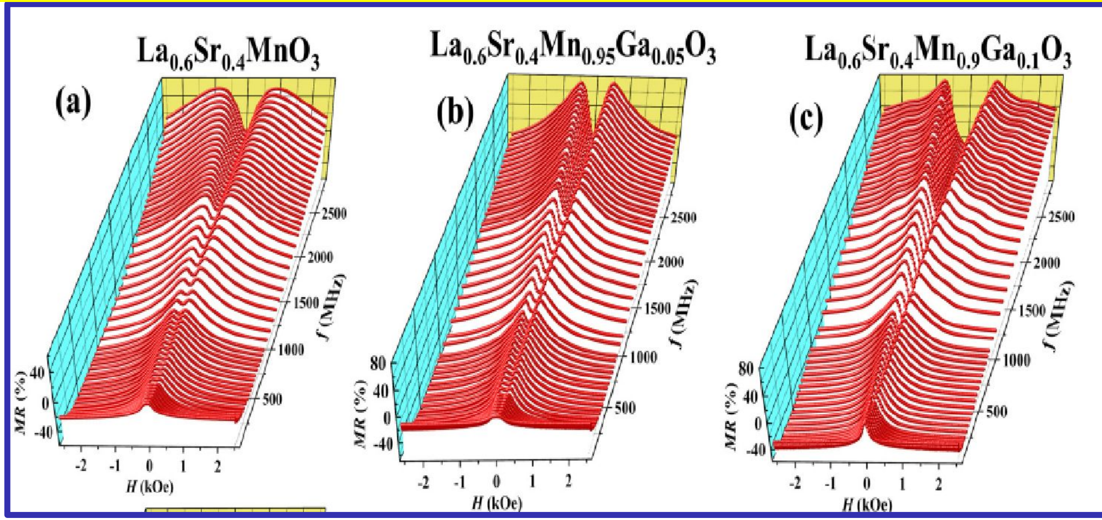
$$\mathrm{MR_{ac}}(H_{\mathrm{dc}}) = R_{\mathrm{sym}}\frac{\left(\frac{\Delta H}{2}\right)^2}{(H_{\mathrm{dc}}-H_{\mathrm{res}})^2+\left(\frac{\Delta H}{2}\right)^2} + R_{\mathrm{asym}}\frac{\frac{\Delta H}{2}(H_{\mathrm{dc}}-H_{\mathrm{res}})}{(H_{\mathrm{dc}}-H_{\mathrm{res}})^2+\left(\frac{\Delta H}{2}\right)^2} + R_0$$

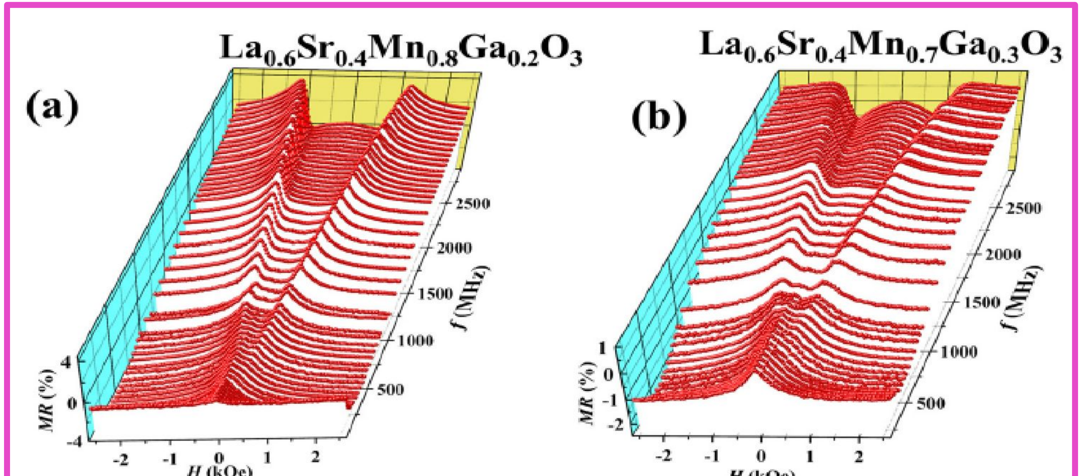
Tuning T_C & Ac MR in $La_{0.6}Sr_{0.4}Mn_{1-x}Ga_xO_3$ (x = 0 to 0.3)



FM@300K

- ○ Giant +ve MR (~ 80%) at low fields !!!
- ○ -ve MR also increases to ~40% !!!



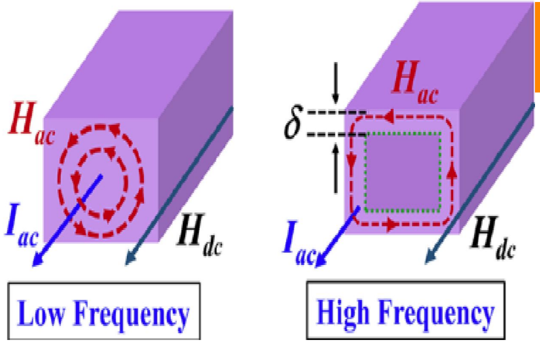


PM@ 300K

A. Chanda et al, J. Appl. Phys. 126, 08395 (2019

What is the origin of ac magnetoresistance?

Impedance of ac current carrying conducting slab $Z = R + iX$



Ac magnetic field is created transverse to the current flow, which interacts with the magnetic moments of the sample.

Skin depth decreases as ω increases $\delta = \sqrt{(2\rho)/(\omega\mu_t)}$

Impedance at high frequencies: $Z = R_{dc}\left[(1+i)/\delta\right] = R_{dc}t(1+i)\left(\frac{\omega\mu_0\mu_t}{2\rho}\right)^{\frac{1}{2}}$

Since transverse permeability (μ) is complex $\mu_t = \mu_t' - i\,\mu_t''$

High frequency impedance $Z = \sqrt{\omega\rho(\omega, H)}\left[\sqrt{\mu_R} + j\sqrt{\mu_X}\right]$

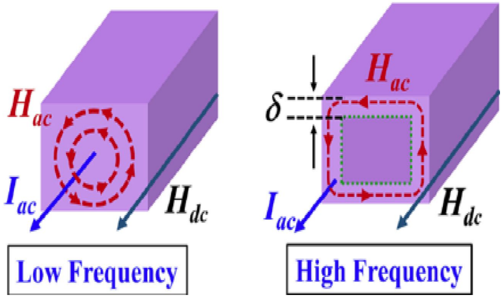
where $\mu_R = |\mu_t| + \mu_t''$, $\mu_L = |\mu_t| - \mu_t''$

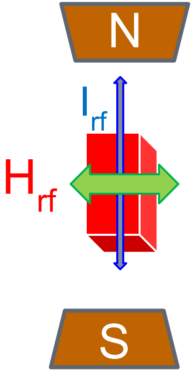
$$R = (\omega\rho)^{1/2}\sqrt{|\mu_t| + \mu_t''}$$

$$X = (\omega\rho)^{1/2}\sqrt{|\mu_t| - \mu_t''}$$

Thus, ac MR is influenced by H-dependence of μ_t !!!
The peak in ac MR may be caused by magnetic resonance in the sample

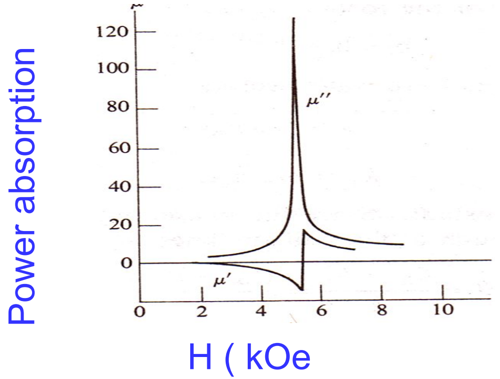
Key point: Ac magnetic field in magnetoimpedance is transverse to dc field



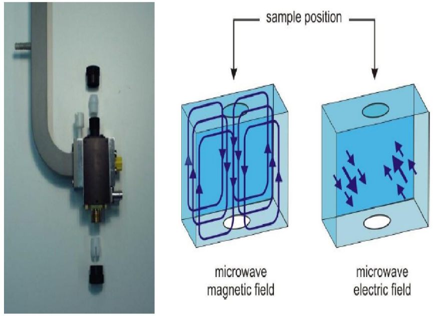


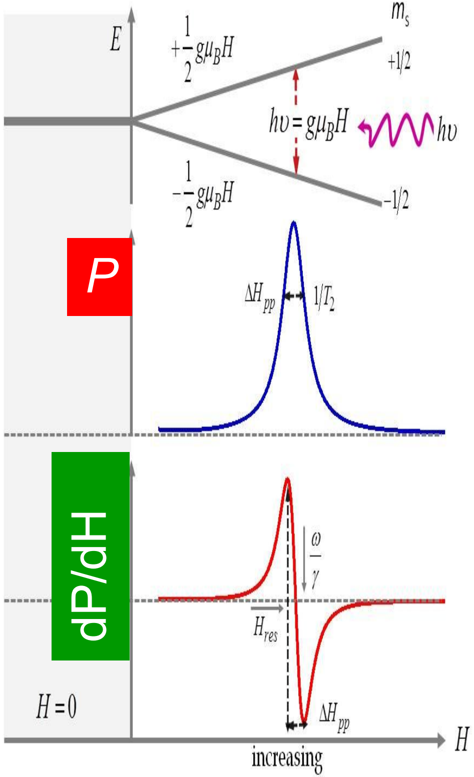
$\parallel$ H_{dc}

$\perp$ H_{dc}

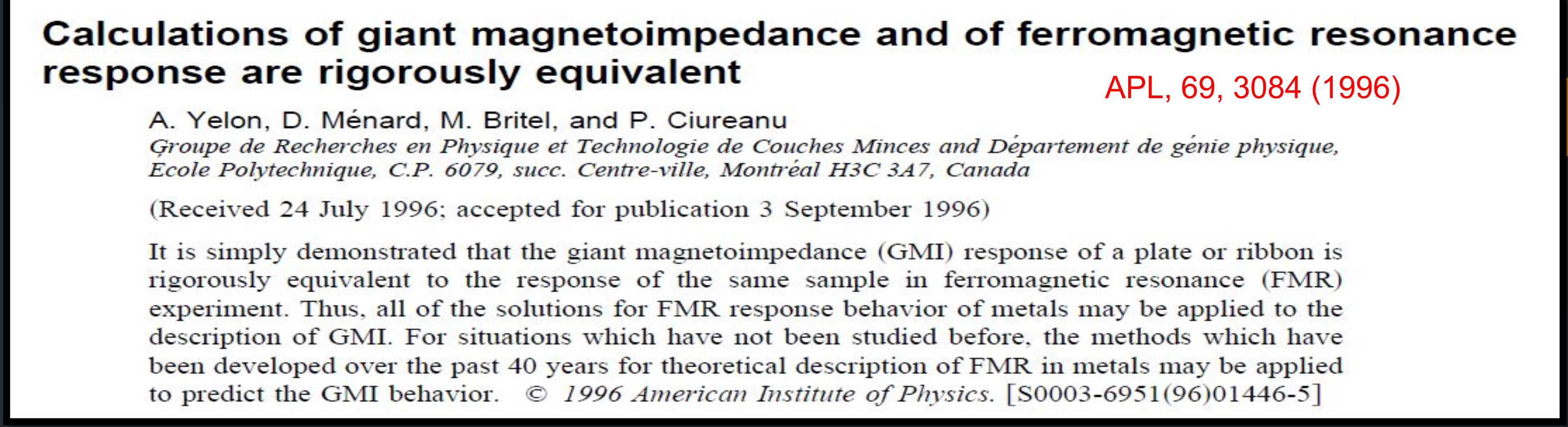


Power absorbed $P \propto \mathrm{Re}(Z) H_{rf}^{2}$





Power absorption $P = (1/2)V\omega\mu'' H_{rf}^{2}$



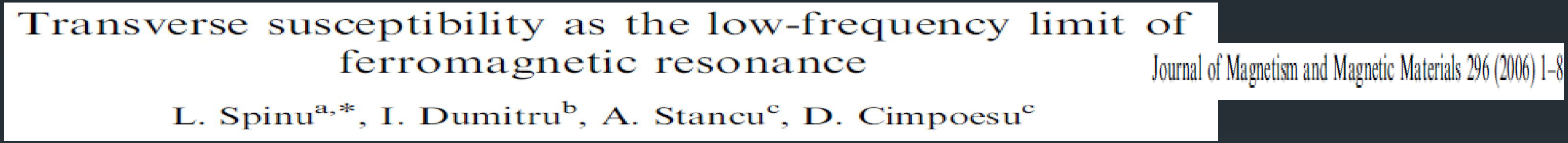
Calculations of giant magnetoimpedance and of ferromagnetic resonance response are rigorously equivalent

APL, 69, 3084 (1996)

A. Yelon, D. Ménard, M. Britel, and P. Ciureanu
Groupe de Recherches en Physique et Technologie de Couches Minces and Département de génie physique, Ecole Polytechnique, C.P. 6079, succ. Centre-ville, Montréal H3C 3A7, Canada

(Received 24 July 1996; accepted for publication 3 September 1996)

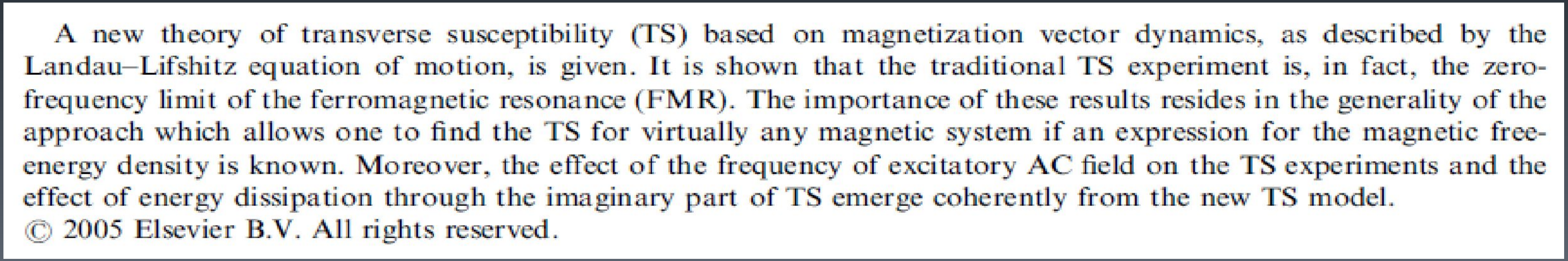
It is simply demonstrated that the giant magnetoimpedance (GMI) response of a plate or ribbon is rigorously equivalent to the response of the same sample in ferromagnetic resonance (FMR) experiment. Thus, all of the solutions for FMR response behavior of metals may be applied to the description of GMI. For situations which have not been studied before, the methods which have been developed over the past 40 years for theoretical description of FMR in metals may be applied to predict the GMI behavior. © *1996 American Institute of Physics.* [S0003-6951(96)01446-5]



Transverse susceptibility as the low-frequency limit of ferromagnetic resonance

Journal of Magnetism and Magnetic Materials 296 (2006) 1–8

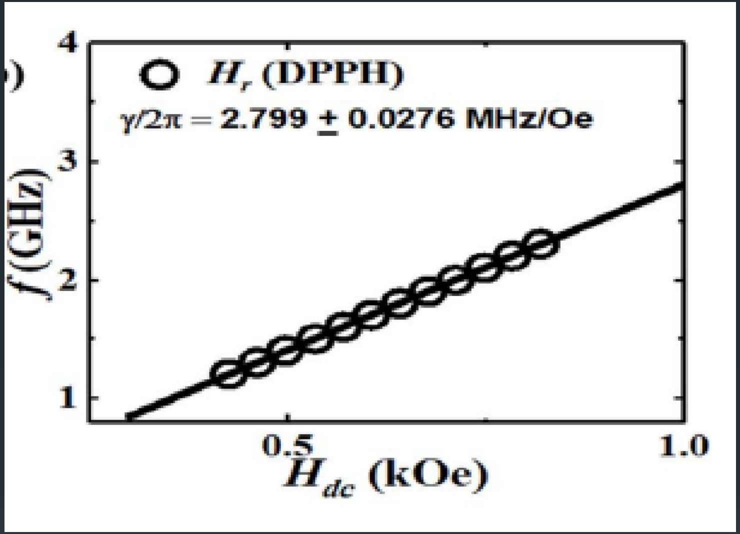
L. Spinu[a,*], I. Dumitru[b], A. Stancu[c], D. Cimpoesu[c]



A new theory of transverse susceptibility (TS) based on magnetization vector dynamics, as described by the Landau–Lifshitz equation of motion, is given. It is shown that the traditional TS experiment is, in fact, the zero-frequency limit of the ferromagnetic resonance (FMR). The importance of these results resides in the generality of the approach which allows one to find the TS for virtually any magnetic system if an expression for the magnetic free-energy density is known. Moreover, the effect of the frequency of excitatory AC field on the TS experiments and the effect of energy dissipation through the imaginary part of TS emerge coherently from the new TS model.

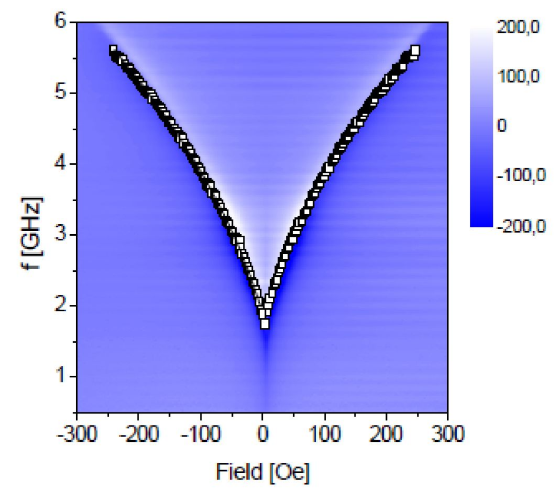
Typical ESR and FMR spectra

Electron spin resonance in DPPH



$$f_r = \left(\frac{\gamma}{2\pi}\right) H_{dc}$$

Ferromagnetic resonance in CoFeB



$$\omega^2 = \mu_0{}^2 \gamma^2 \left(\mathrm{H_{res}} + \mathrm{H_k}\right)\left(\mathrm{H_{res}} + \mathrm{H_k} + \mathrm{M_s}\right)$$

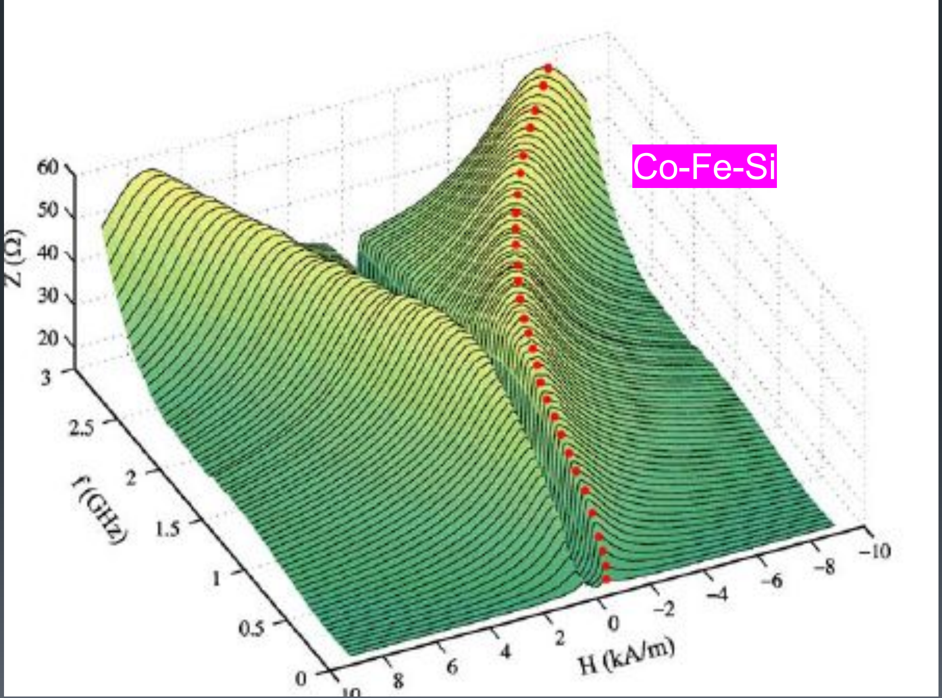
Kittel’s equation

JOURNAL OF APPLIED PHYSICS **99**, 103904 (2006)

Transition from quasistatic to ferromagnetic resonance regime in giant magnetoimpedance

J. M. Barandiarán, A. García-Arribas,[a] and D. de Cos
Departamento de Electricidad y Electrónica, Universidad del País Vasco, Apartado 644, 48080 Bilbao, Spain

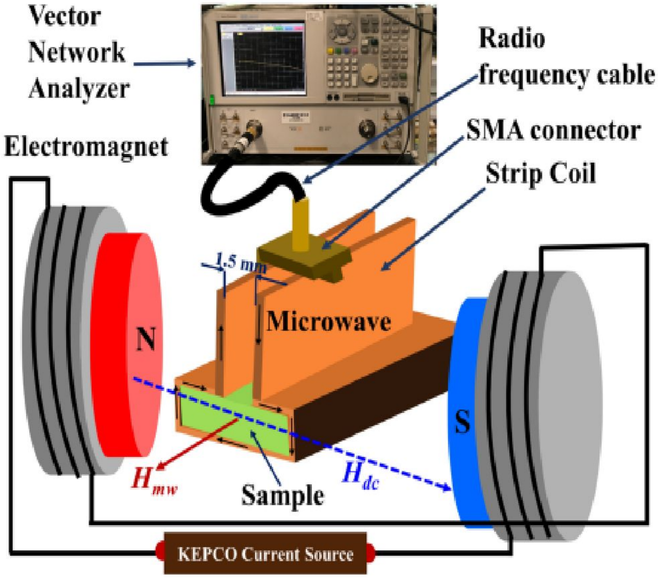
Total Impedance (Z)



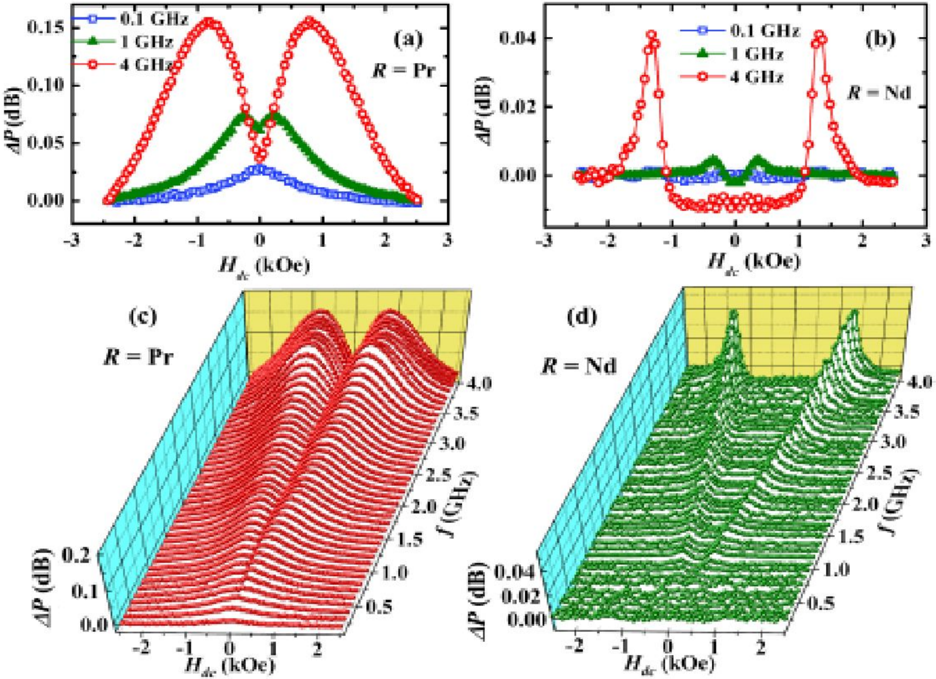
$$P = \mathrm{Re}(Z)H^2_{rf}$$

Barandiaran et al. J. Appl. Phys. 99, 103904 (2005)

Proof 1. Measurement of MW power absorption using a Cu-strip coil & VNA

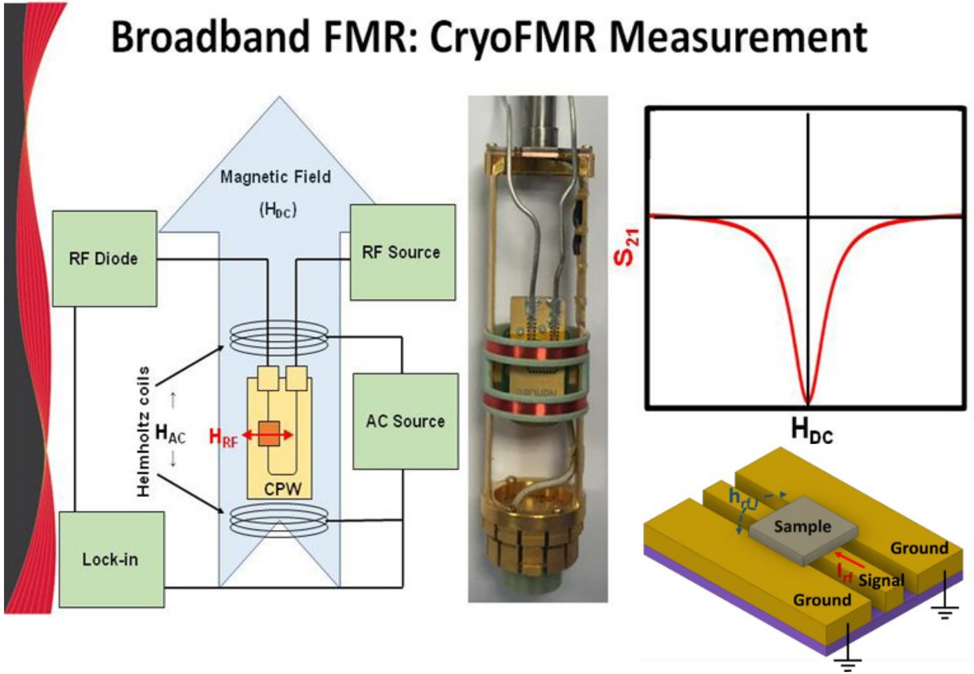


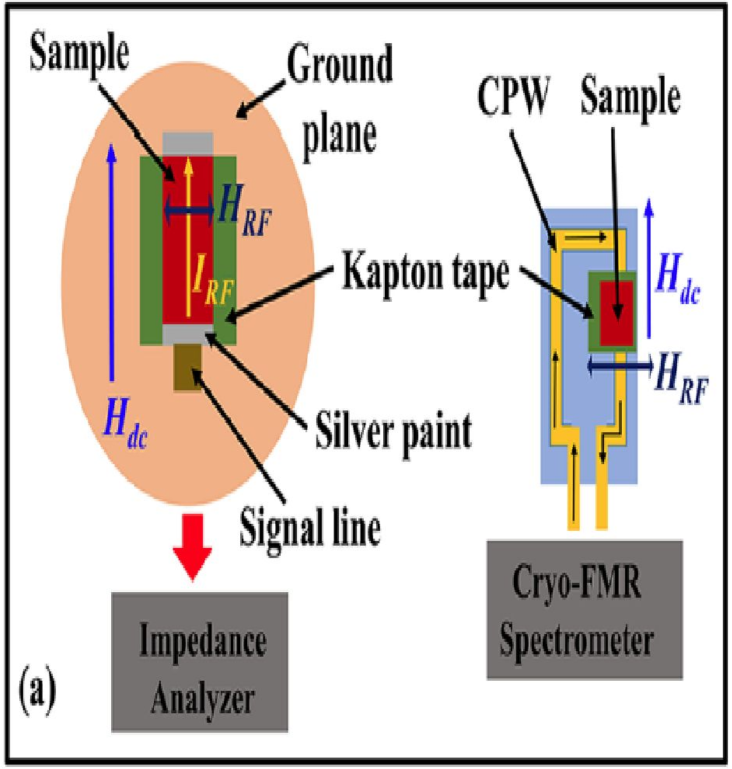
$Pr_{0.6}Sr_{0.4}MnO_3$ $Nd_{0.6}Sr_{0.4}MnO$



$$P(H_{dc}) = P_{max}\frac{\left(\frac{\Delta H}{2}\right)^2}{(H_{dc}-H_{res})^2+\left(\frac{\Delta H}{2}\right)^2}$$

Proof 2: Double confirmation of ESR/FMR with a commercial broadband magnetic resonance spectrometer



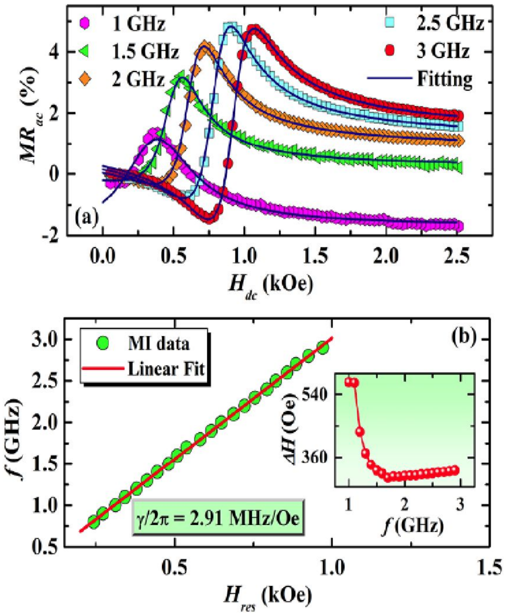


Ac magnetoresistance & broadband ESR

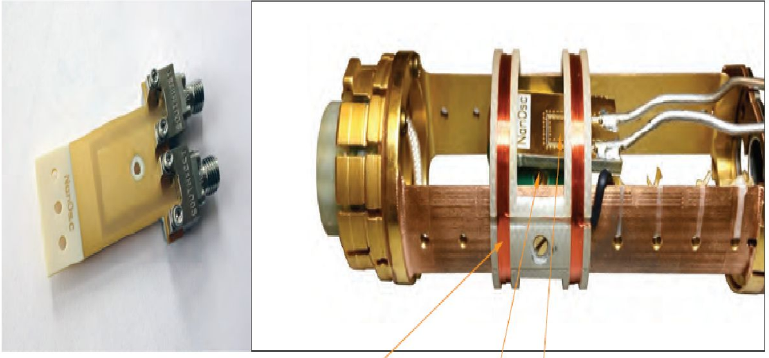
Ac Magnetoresistance

$Nd_{0.6}Sr_{0.4}MnO_3$

Broadband ESR



CryoFMR Probe Insert for PPMS® (VersaLab™)



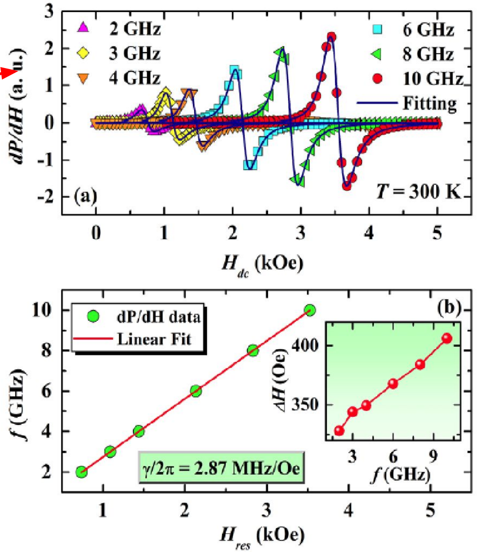
Coplanar Waveguide (CPW) for room temperature PhaseFMR

Helmholtz coils for field modulation

Temperature sensor

CPW for in-plane analysis

*(out-of-plane) CPW also available



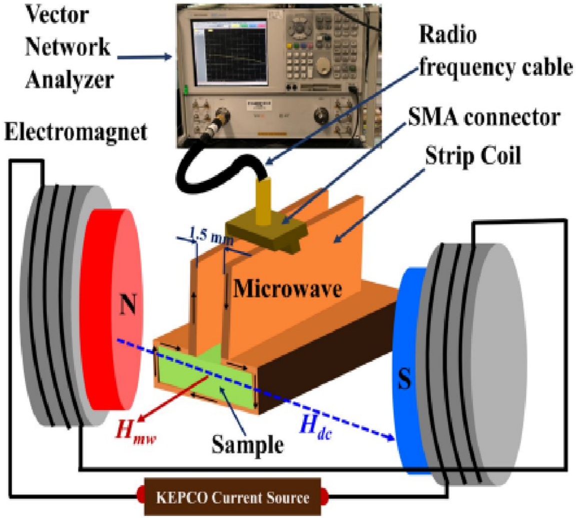
For ESR $f = (\gamma/2\pi)\, H_{res}$

$$\mathrm{MR}_{\mathrm{ac}}(H_{\mathrm{dc}}) = R_{\mathrm{sym}} \frac{\left(\frac{\Delta H}{2}\right)^2}{(H_{\mathrm{dc}} - H_{\mathrm{res}})^2 + \left(\frac{\Delta H}{2}\right)^2} + R_{\mathrm{asym}} \frac{\frac{\Delta H}{2}(H_{\mathrm{dc}} - H_{\mathrm{res}})}{(H_{\mathrm{dc}} - H_{\mathrm{res}})^2 + \left(\frac{\Delta H}{2}\right)^2} + R_0$$

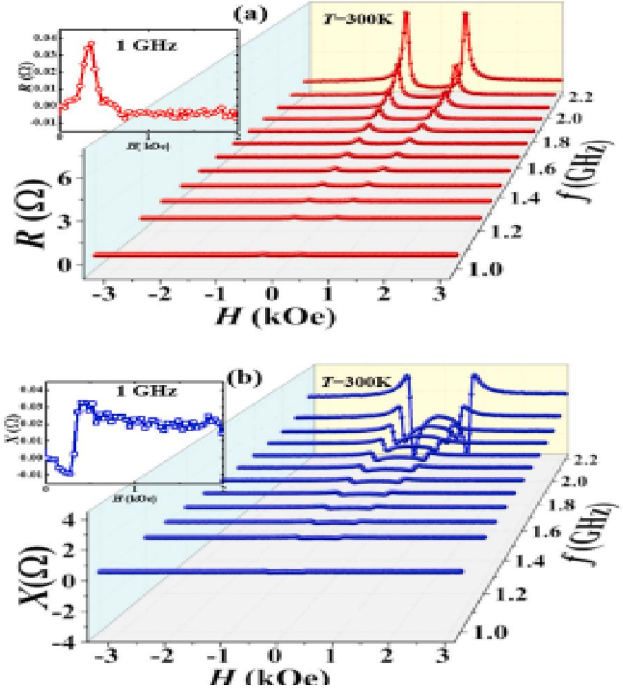
$$\frac{dP}{dH} = P_{\mathrm{sym}} \frac{\frac{\Delta H}{2}(H_{\mathrm{dc}} - H_{\mathrm{res}})}{\left[(H_{\mathrm{dc}} - H_{\mathrm{res}})^2 + \left(\frac{\Delta H}{2}\right)^2\right]^2} + P_{\mathrm{asym}} \frac{\left(\frac{\Delta H}{2}\right)^2 - (H_{\mathrm{dc}} - H_{\mathrm{res}})^2}{\left[(H_{\mathrm{dc}} - H_{\mathrm{res}})^2 + \left(\frac{\Delta H}{2}\right)^2\right]^2} + P_0$$

Can we measure an insulating sample?

Trick 1: Enclose the sample with rectangular copper strip and measure its impedance



La_2NiMnO_6
(T_C = 270 K)



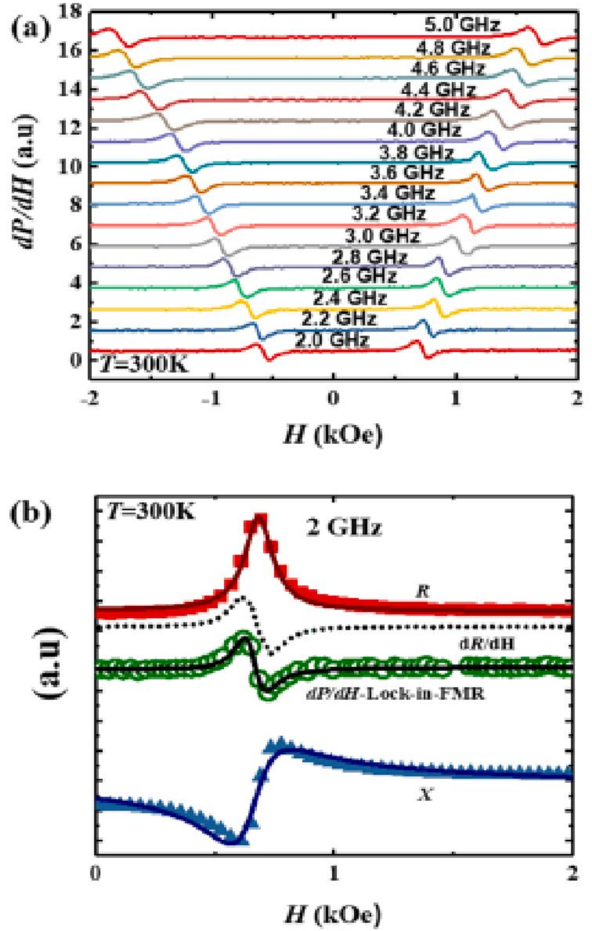
Paramagnetic resonance in La_2NiMnO_6 probed by impedance and lock-in detection techniques

Ushnish Chaudhuri[a], Debendra Prasad Panda[b], A. Sundaresan[c], R. Mahendiran[a,*]

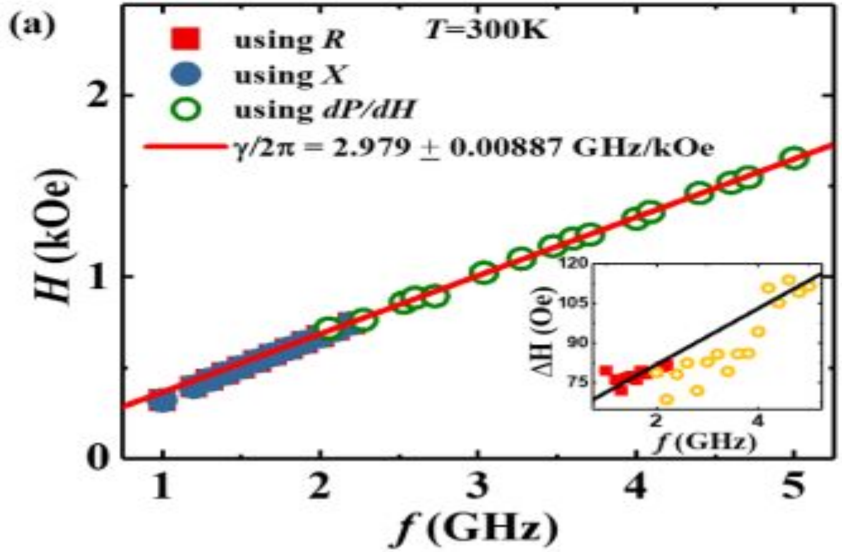
Journal of Magnetism and Magnetic Materials 518 (2021) 167400

$$R \text{ or } X = K_{sym}\frac{(\Delta H)^2}{(H-H_r)^2+(\Delta H)^2} + K_{asym}\frac{(\Delta H)(H-H_r)}{(H-H_r)^2+(\Delta H)^2} + C$$

Broadband ESR & MI in La_2NiMnO_6



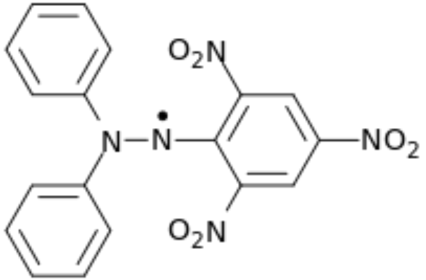
Comparison of 2 different techniques



$$\frac{dP}{dH} = A_{asym}\frac{(\Delta H/2)(H-H_r)}{[(H-H_r)^2+(\Delta H/2)^2]^2} - A_{sym}\frac{(\Delta H/2)^2-(H-H_r)^2}{[(H-H_r)^2+(\Delta H/2)^2]^2} + B$$

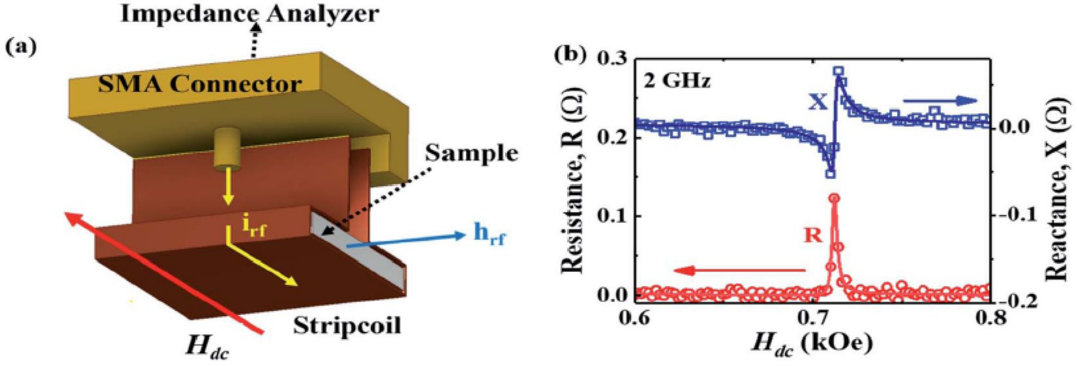
Magnetoimpedance of an insulating organic molecule: DPPH

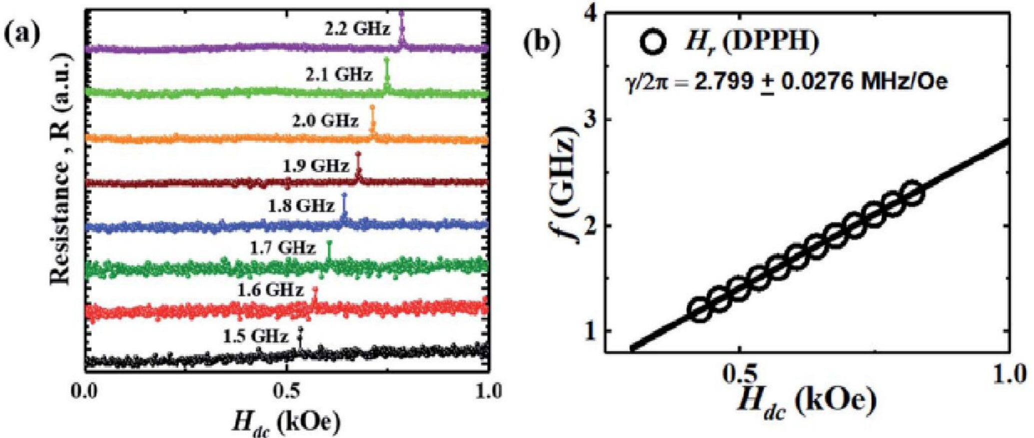
Trick 1: Enclose the sample in a Cu-strip coil



One unpaired spin (free radical) per 41 atoms

DPPH is used as a ESR marker

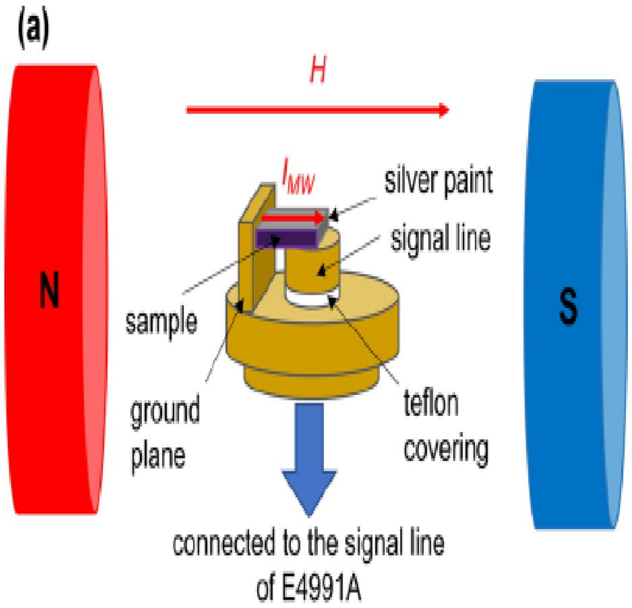


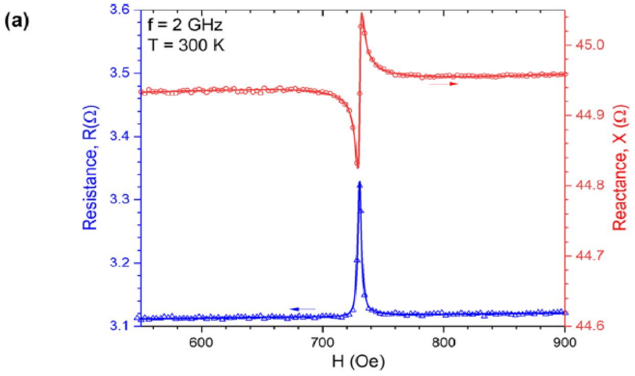


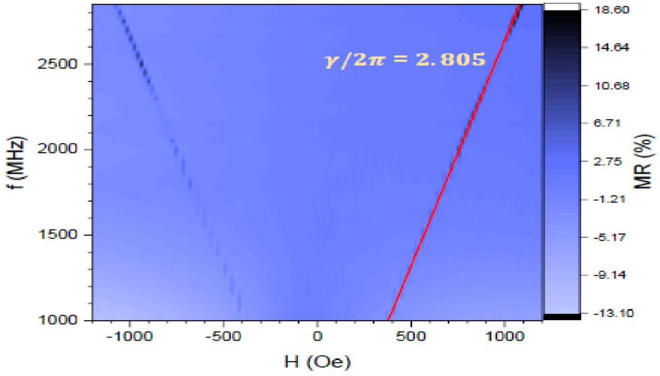
U. Chaudhuri and R. Mahendiran, RSC Adv, 10. 17311 (2020)

Trick 2: How about making a conducting electrode on DPPH?

Coat the top surface and sides of DPPH with Ag paint







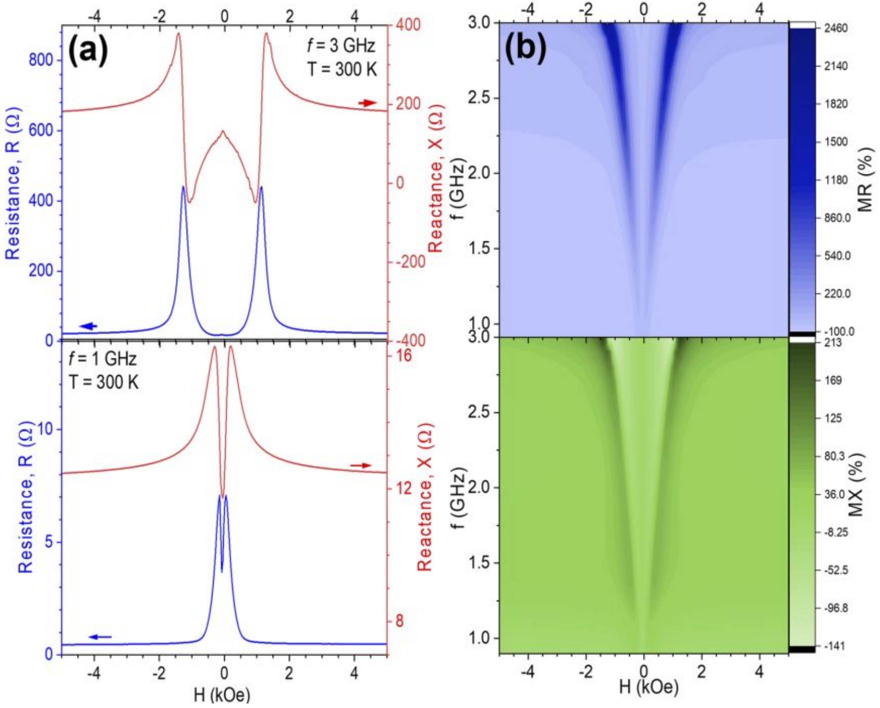
. H. Lee, U. Chaudhuri, and R. Mahendiran, *Electrically detected paramagnetic resonance in Ag-paint covered DPPH*, J. Phys. Chem. C (2021)

Electrical detection of ferromagnetic resonance in bulk YIG sample covered with a layer of silver paint

Yong Heng Lee[a] and R. Mahendiran

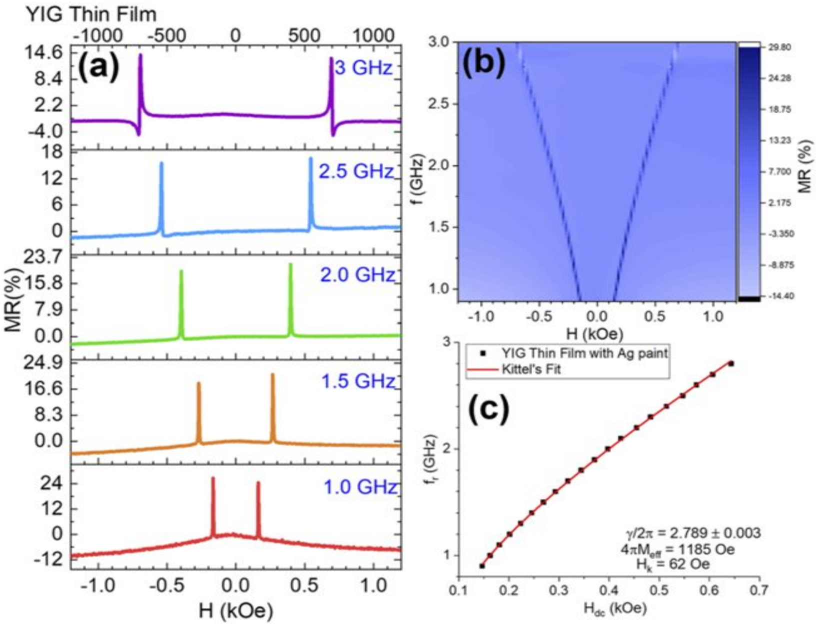
Cite as: AIP Advances 12, 035032 (2022); doi: 10.1063/9.0000351

Bulk ceramic YIG

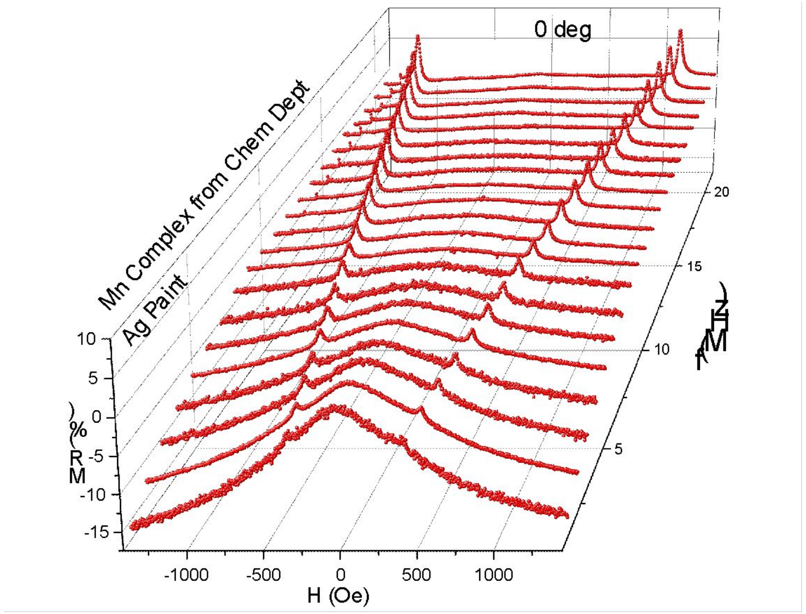


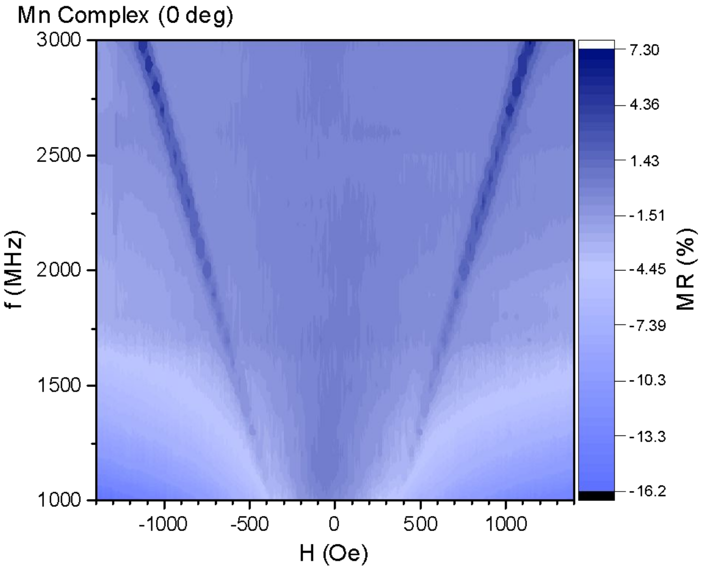
Thin film YIG/GGG

$$f_r = \frac{\gamma}{2\pi}\sqrt{[H_{dc} + H_k][H_{dc} + H_k + 4\pi M_{eff}]}$$



Magnetoimpedance in organic-inorganic complex $(PFB)_2MnCl_4$





$\gamma/2\pi = 2.8$ GHz/kOe

In collaboration with K. P. Loh, Chemistry, NUS

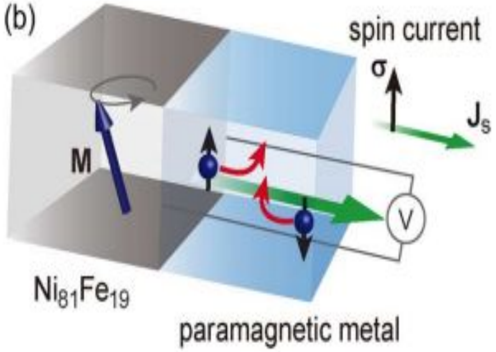
JOURNAL OF APPLIED PHYSICS **102**, 083915 (2007)

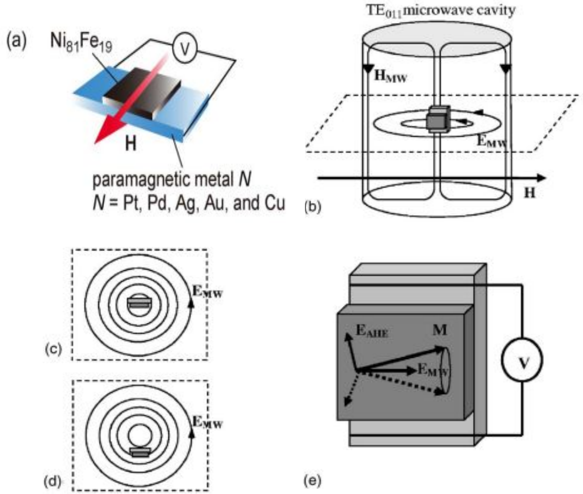
Detection of pure inverse spin-Hall effect induced by spin pumping at various excitation

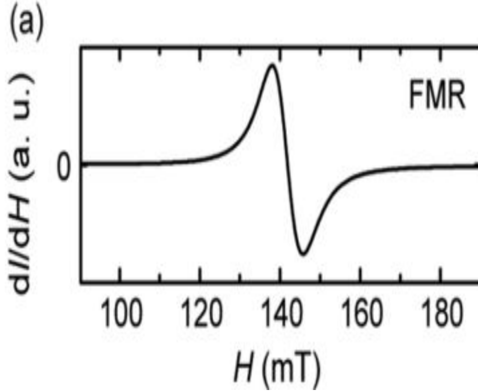
H. Y. Inoue,[a] K. Harii, K. Ando, K. Sasage, and E. Saitoh
Department of Applied Physics, Keio University, Hiyoshi, Yokohama 223-8522, Japan

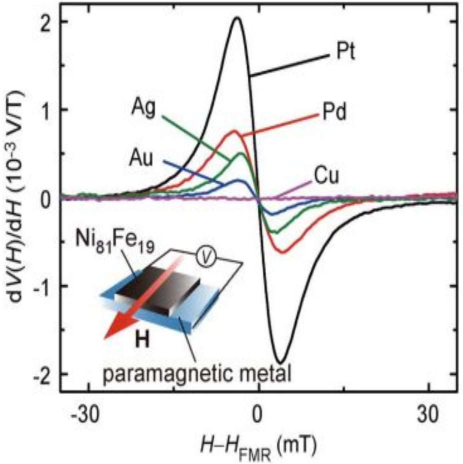
Electrical detection of FMR via inverse spin Hall effect(ISHE)

Sample is inside a microwave cavity





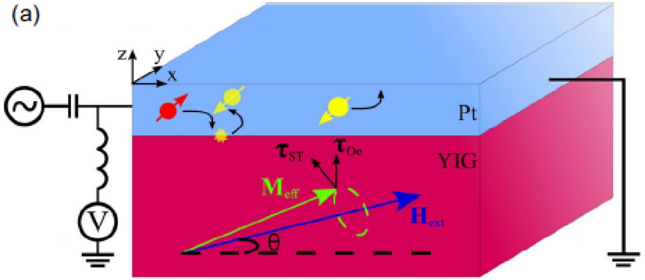


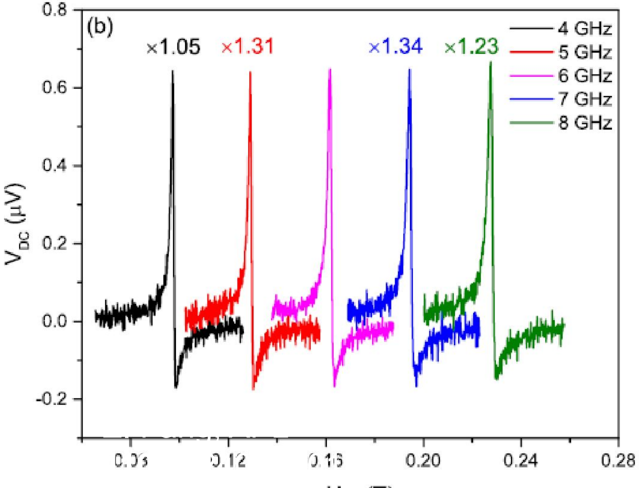


ISHE: Spin current pumped from NiFe into Pt film is converted into a charge current

Current-driven FMR in FM/NM bilayers: Detection by Inverse Spin Hall effect

YIG/Pt





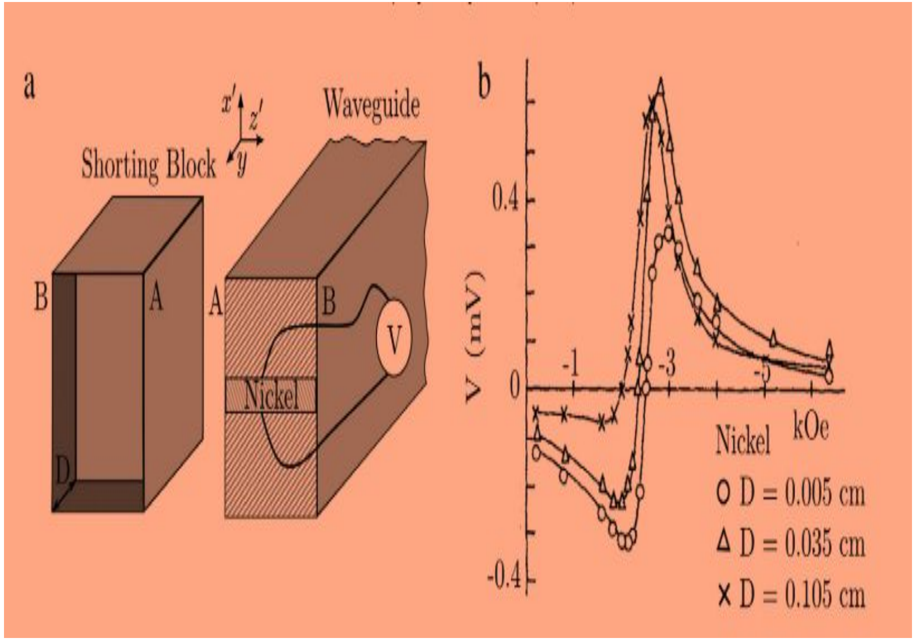
RF current through non-magnetic "Pt" injects pure spin current in the FM layer, which transfer spin angular momentum to M in FM layer and causes spin precession

Back flow of spin current to Pt induces dc voltage in the Pt due to Inverse spin Hall effect (ISHE) .

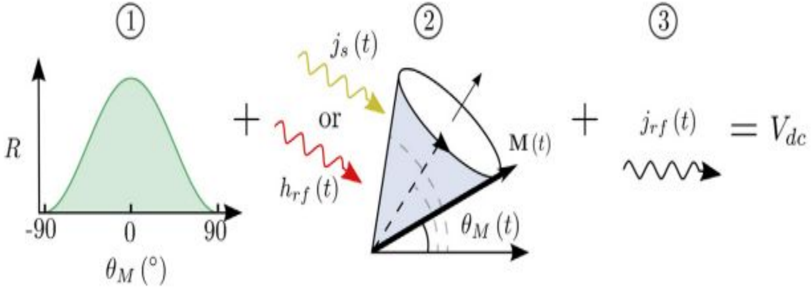
Our experiment does not need a layer of high-spin orbit coupling normal metal.

Will ac ISHE or ac spin Hall MR larger than dc counterparts?

Electrical detection of FMR in a single layer film : DC voltage at FMR in NiFe film due to spin rectification



H.J. Juretschke, DC detection of spin resonance in thin metallic films, J. Appl. Phys. 34, 123 (1963

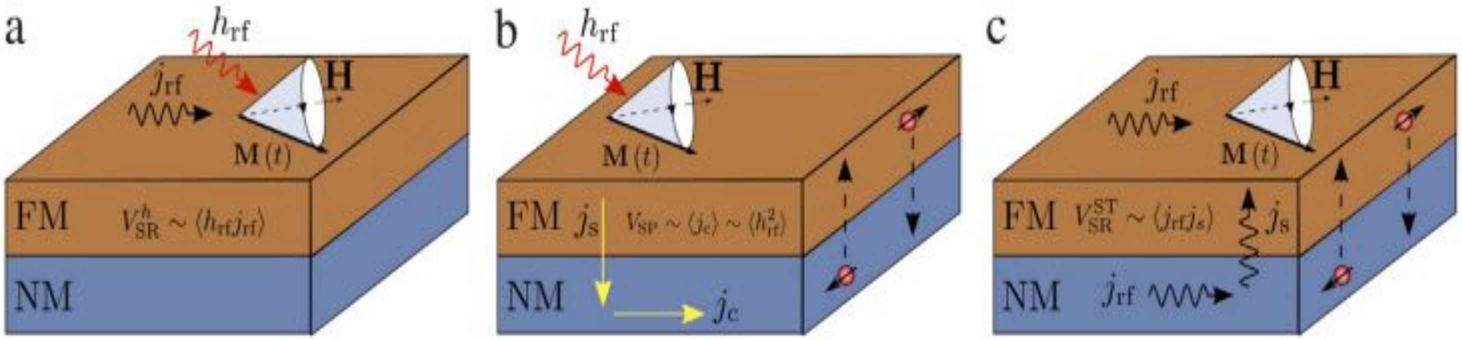


Nonlinear coupling between dynamic magnetoresistance and induced rf current

$$V = \langle \mathrm{Re}\,\{I\,(t)\}\,\mathrm{Re}\,\{R\,[\mathbf{H}\,(t)]\}\rangle = \frac{1}{2} I_0 \mathbf{h}_0 \cdot \nabla R\,(\mathbf{H}_0)\cos\Phi\,.$$

Multiple sources of dc voltage in a bilayer

M. Harder et al. / Physics Reports 661 (2016) 1–59



Spin rectification

Spin pumping

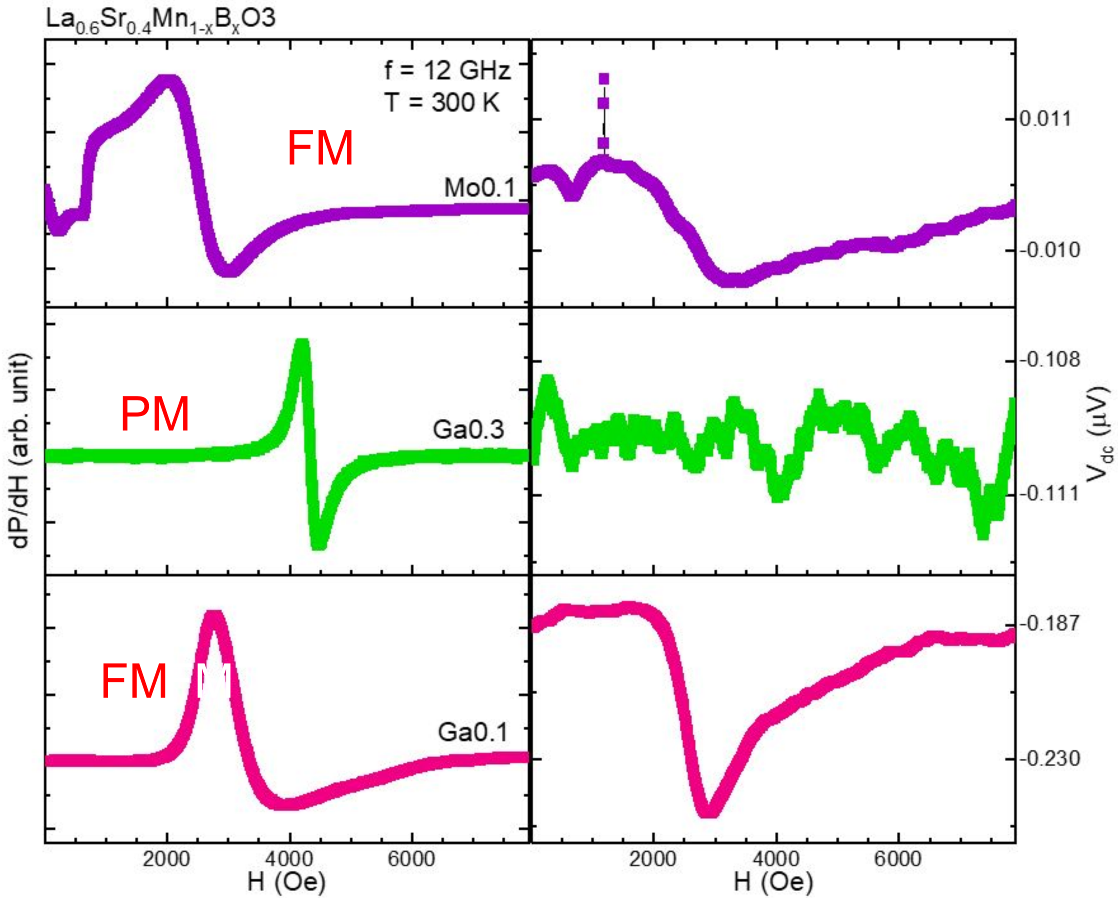
Spin Transfer Torque

Nonlinear coupling between dynamic MR and I_{rf}

ISHE dominates

Nonlinear coupling between dynamic MR and I_{rf}

DC voltage at FMR/ESR in bulk ceramic sample (In progress



Simultaneous FMR and dc voltage detection

Summary

- Ac MR >> dc MR.

- Ac MR shows fingerprints of ESR/FMR.
- They were verified also by VNA-based MW power absorption, and broadband spectroscopy

- Ac MR is a simple, but less exploited technique. It can be explored for domain wall resonance and domain mode resonance in non-saturated regions.

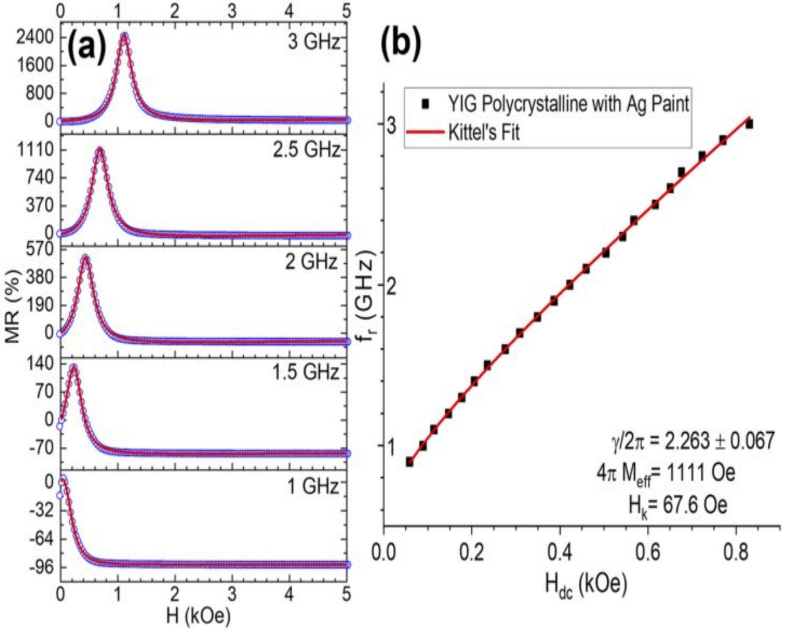
Feature directions:

- Origins other than Oersted field? Spin-Transfer Torque?

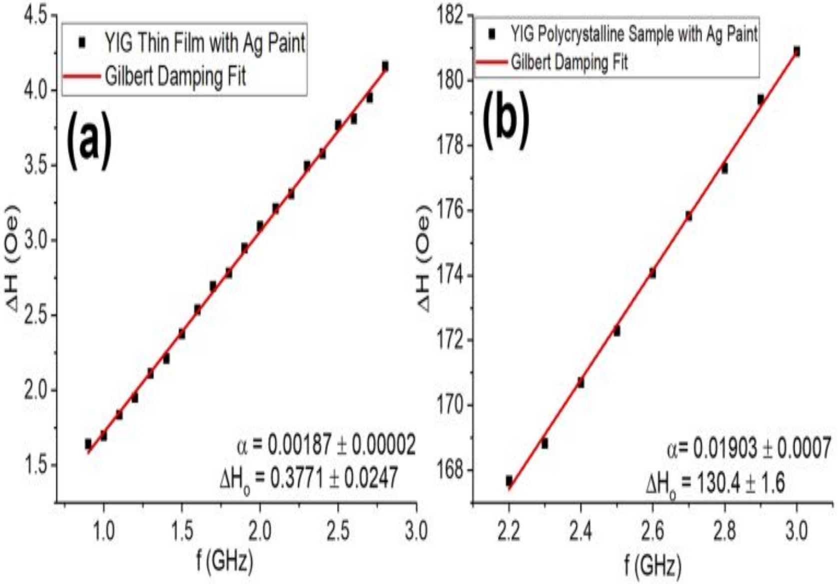
- Does the ac spin Hall effect play any role?


Thank You!
shutterstock.com • 1153070891


WELCOME
Collaboration



$$f_r = \frac{\gamma}{2\pi}\sqrt{[H_{dc} + H_k][H_{dc} + H_k + 4\pi M_{eff}]}$$

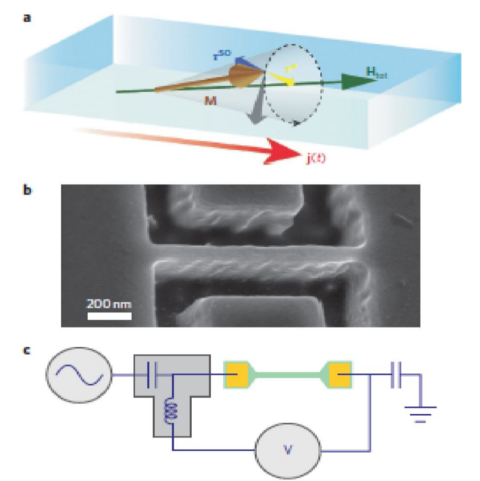


$$\Delta H = \Delta H_0 + \frac{2\pi f}{\gamma}\alpha$$

2. Spin-orbit torque driven FMR in a single layer

Spin-orbit-driven ferromagnetic resonance

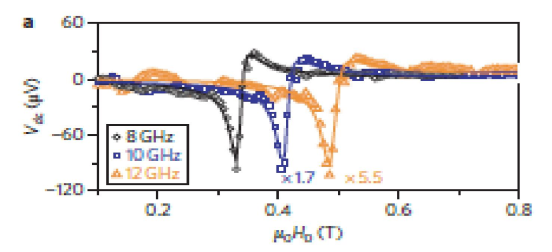
Room-temperature spin-orbit torque in NiMnSb



$Ga_{0.95}Mn_{0.05}As$ (T_C = 120 K)

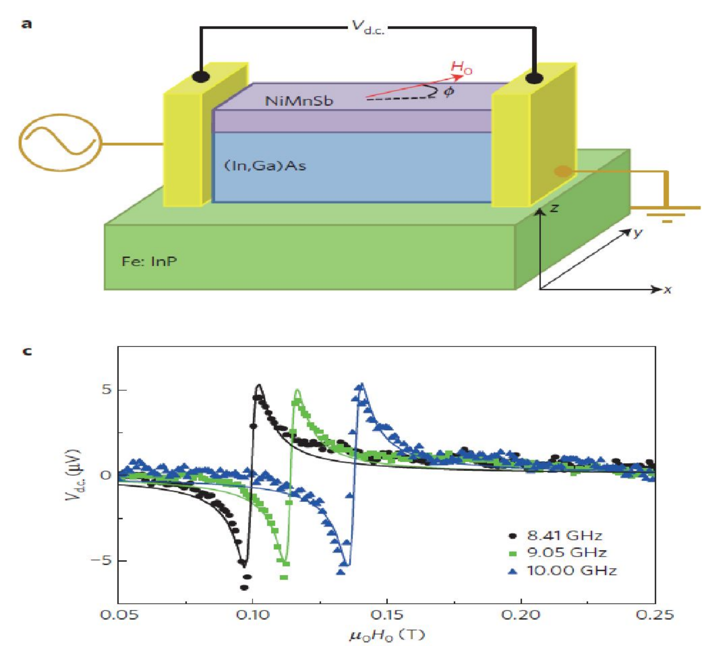
SOC coupling in non-centrosymmetric bulk sample can drive FMR

-Similar to our MI ?



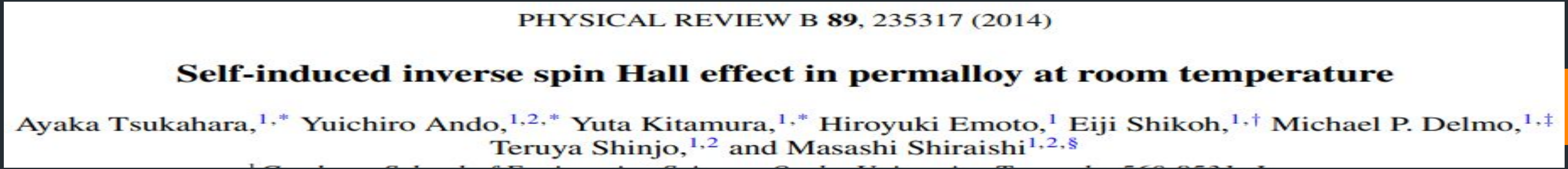
D. Fang

NATURE NANOTECHNOLOGY | VOL 6 | JULY 2011 | www.nature.com/naturenanotechnology



C. Ciccarelli

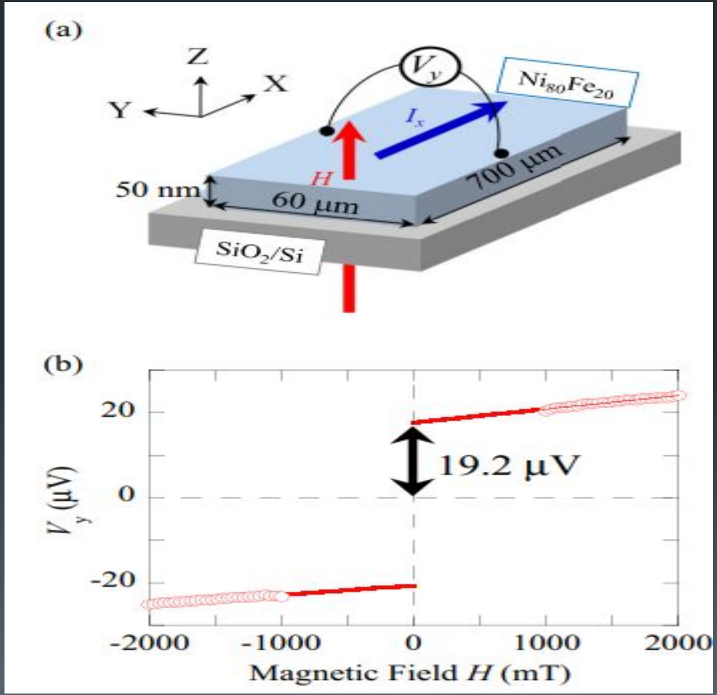
NATURE PHYSICS | VOL 12 | SEPTEMBER 2016 | www.nature.com/naturephysics

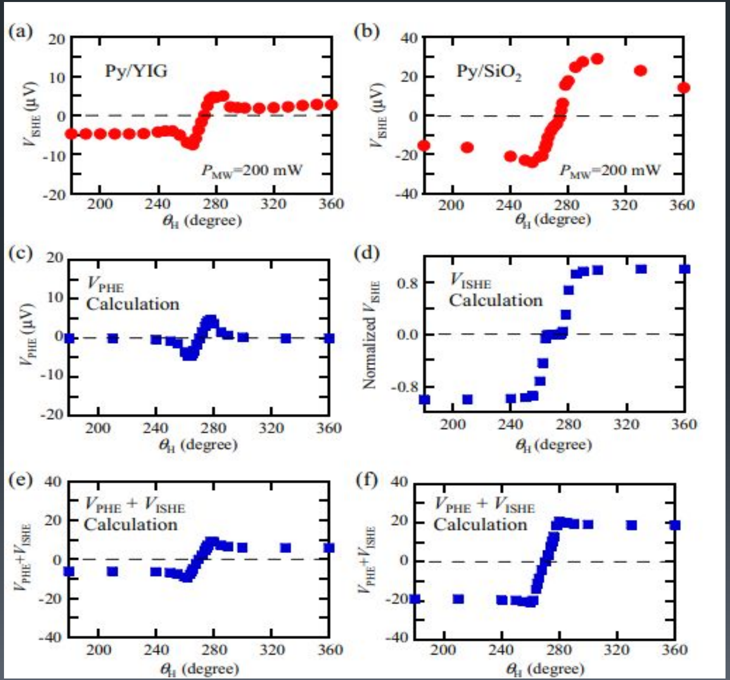


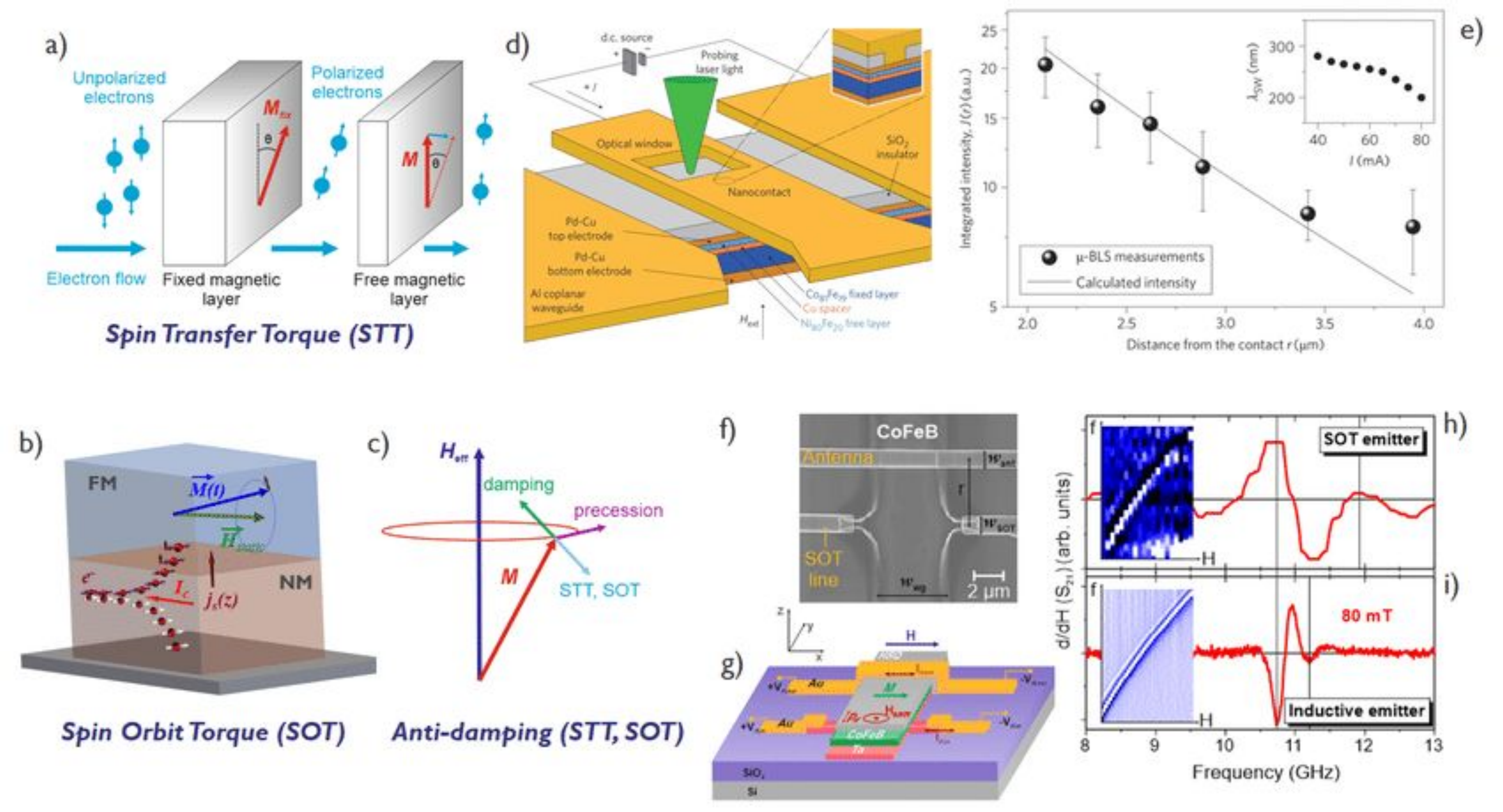
PHYSICAL REVIEW B **89**, 235317 (2014)

Self-induced inverse spin Hall effect in permalloy at room temperature

Ayaka Tsukahara,[1,*] Yuichiro Ando,[1,2,*] Yuta Kitamura,[1,*] Hiroyuki Emoto,[1] Eiji Shikoh,[1,†] Michael P. Delmo,[1,‡]
Teruya Shinjo,[1,2] and Masashi Shiraishi[1,2,§]





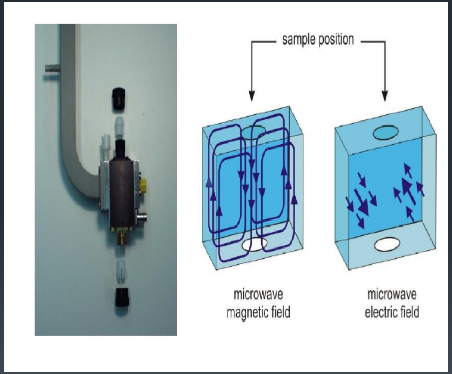


Spin Transfer Torque (STT)

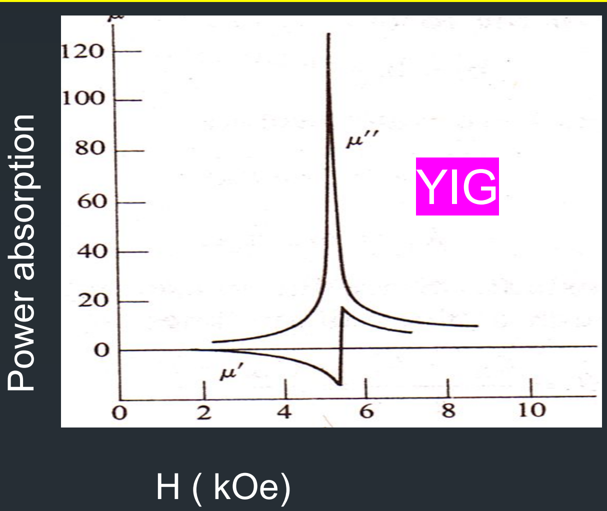
Spin Orbit Torque (SOT)

Anti-damping (STT, SOT)

Double peak in MR : Reminiscence of paramagnetic/ferromagnetic resonance?



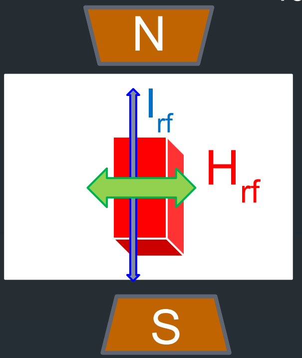
Conventional FMR with sample inside a microwave cavity



Absorbed Power in ESR/FMR:

$$P = (1/2)V\omega\mu'' H_{rf}^2$$

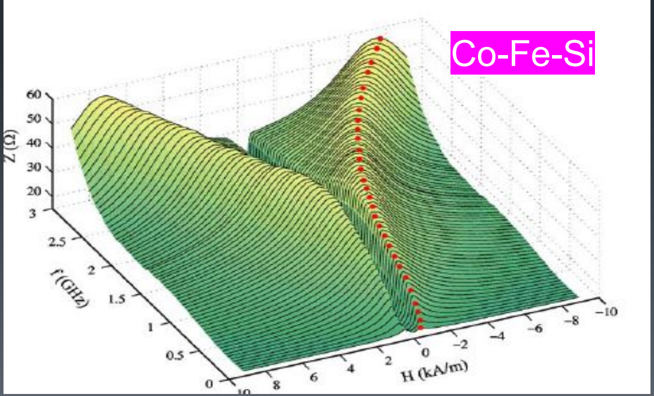
Barandiaran et al. J. Appl. Phys. 99, 103904 (2005)

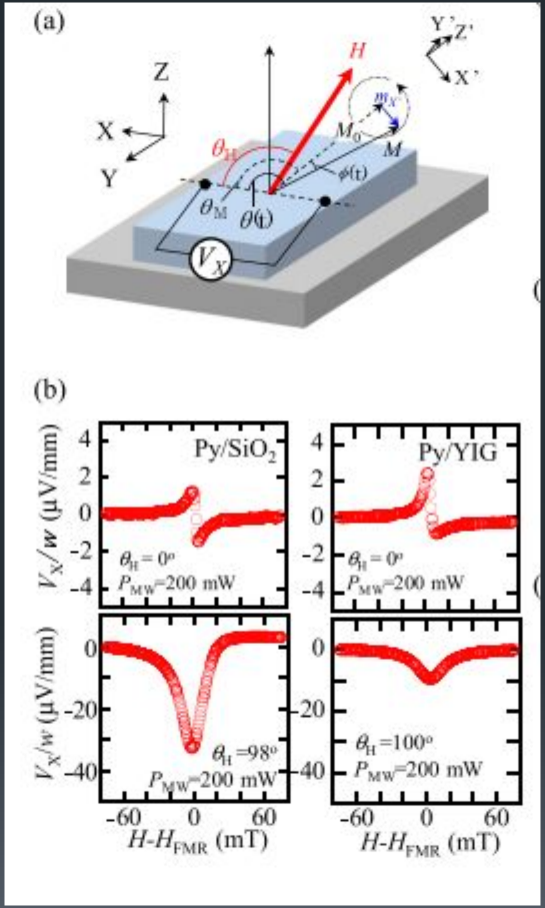


$H_{rf} \perp H_{dc}$

Absorbed Power in MI:

$$P \propto \mathrm{Re}(Z) H_{rf}^{2}$$




(a)
Y'
Z'
X'
Z
X
Y
H
$m_{X'}$
M_0
M
θ_H
$\phi(t)$
θ_M
$\theta(t)$
V_X
(b)
Py/SiO$_2$
Py/YIG
V_X/w (μV/mm)
$\theta_H = 0°$
P_{MW}=200 mW
θ_H =98°
θ_H =100°
4
2
0
-2
-4
-20
-40
-60
60
H-H_{FMR} (mT)