

# **“Recent advances in magnetism at the nanoscale”**

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## Magnetism in nanostructures and applications



## Nanofabrication y advanced microscopies





(ILL año 1987)



# Outline

- Introduction: Magnetic quasiparticles and spin currents
- Magnetic polarons in manganites
- Thin film multilayers and the emergence of new thermospin effects
- Magnetic nanoparticles as nanoheaters and ultrasound emitters
- Conclusions

# Introduction

- Exchange interaction
- Magnetic quasiparticles
- Spin currents

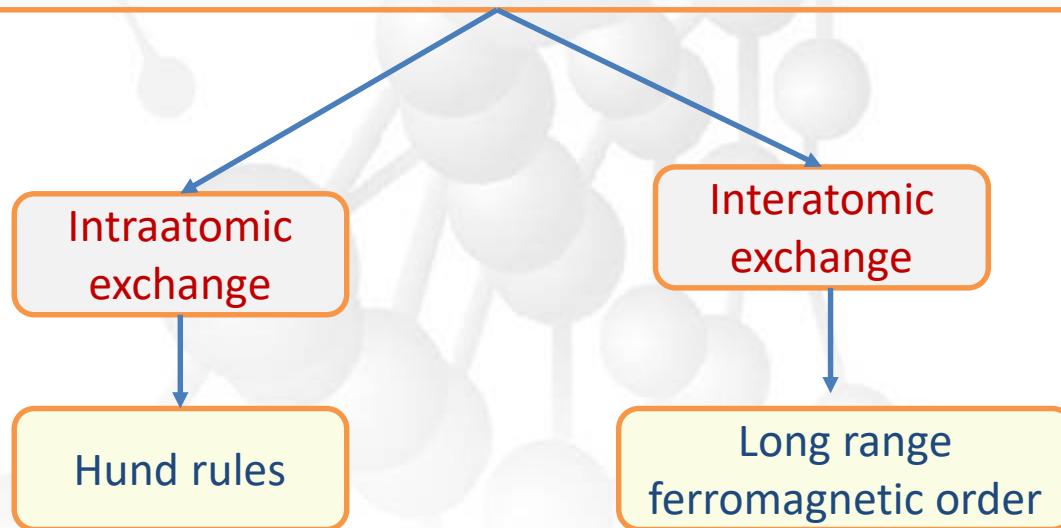


# Exchange interaction

Magnetism constitutes a unique scenario to study the condensed matter from a macro-meso and microscopic point of view

Macroscopic: Maxwell laws

Microscopic: Electrons Coulomb repulsion and Pauli principle



# Going to the smallest: Quasiparticles

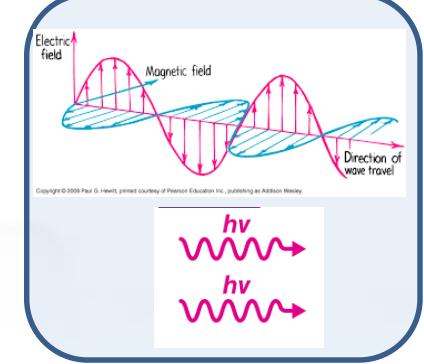
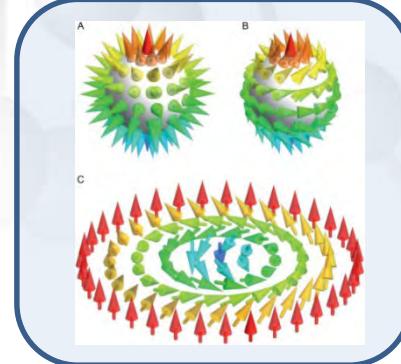
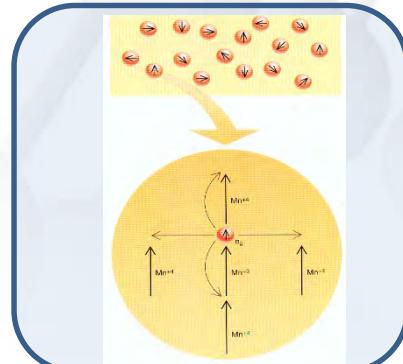
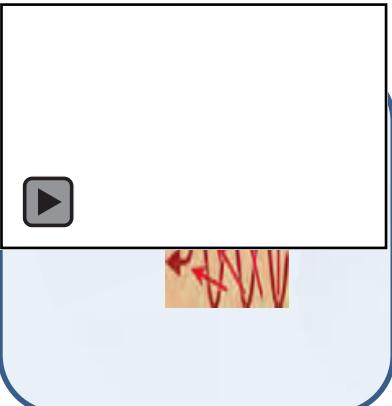
Magnetic quasiparticles: emergent nano-objs resulting from collective excitations

**Magnon**

**Magnetic polaron**

**Skyrmion**

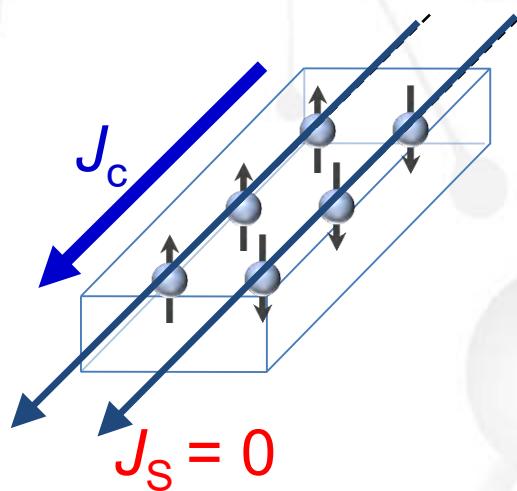
**Photon**



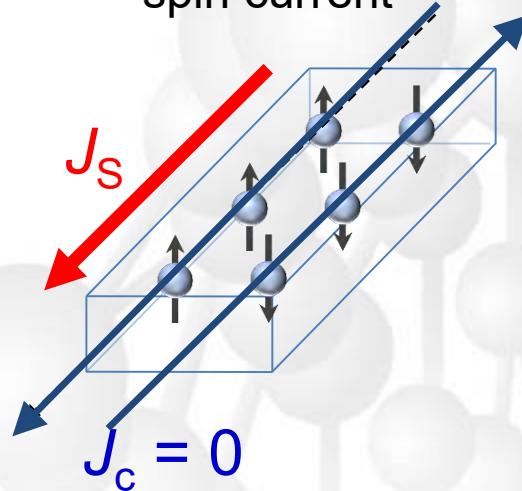
# Charge and spin currents

$J_c$ : charge current

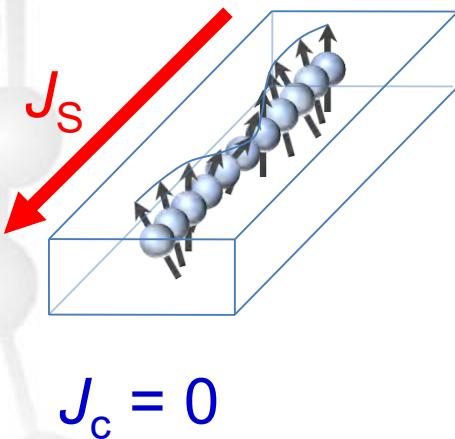
$J_s$ : spin current



Conduction-electron  
spin current



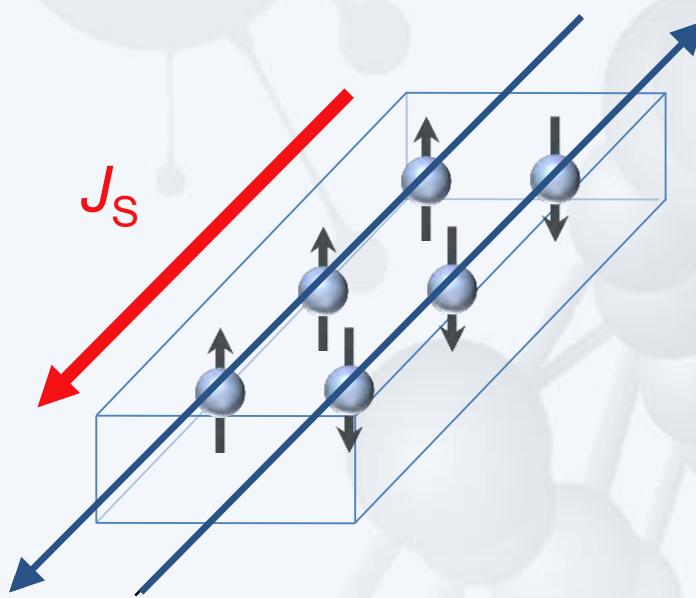
Spin wave (magnons)  
spin current



Spin current: no Joule heating!

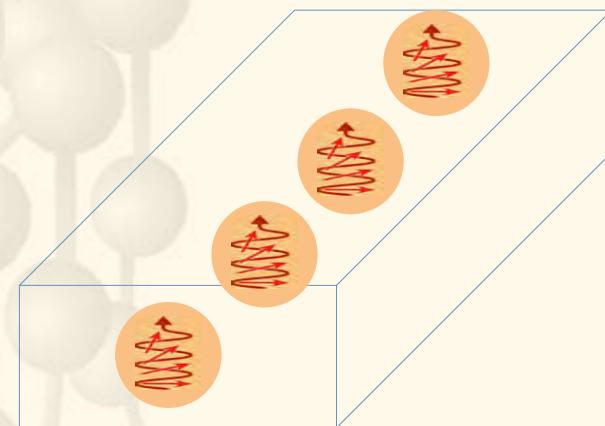
# Pure Spin Currents

Magnetic  
Metal



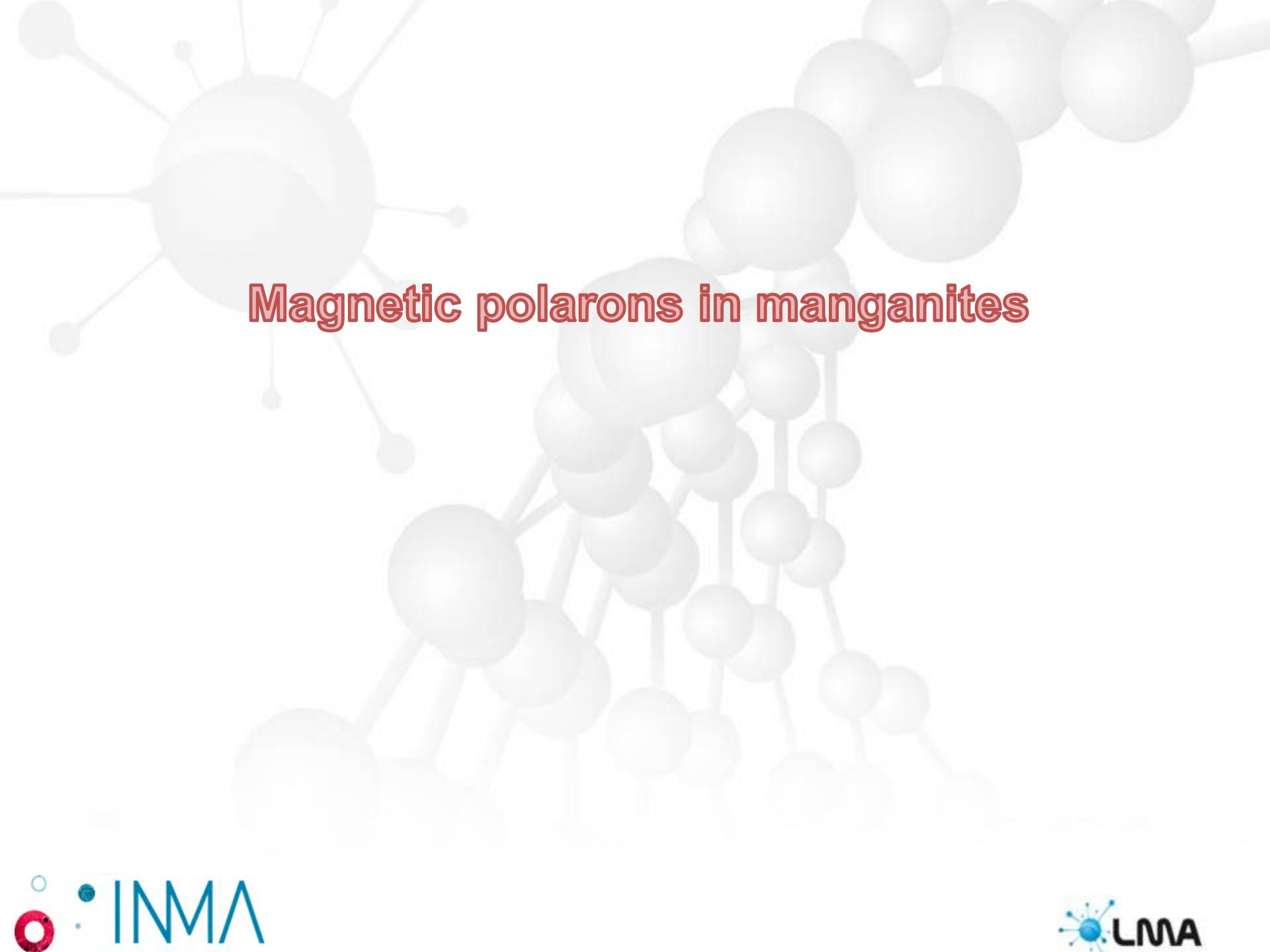
Net electron spin flow

Magnetic  
Insulator

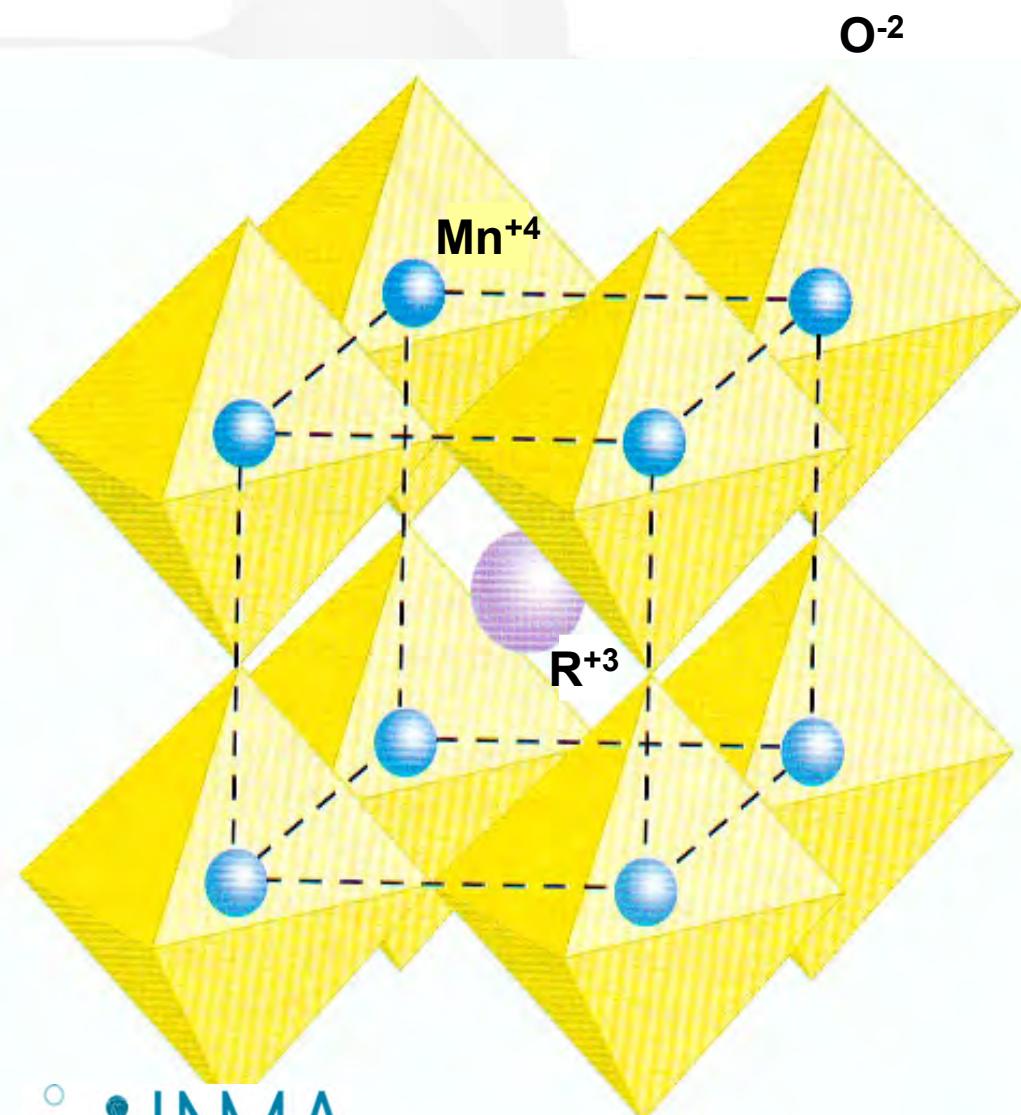


Magnon flow

# Magnetic polarons in manganites



# Manganites structure



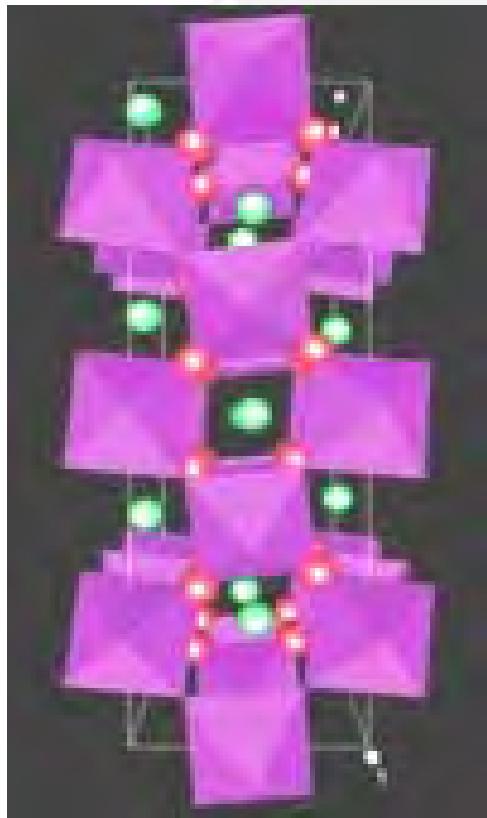
Cubic perovskite structure



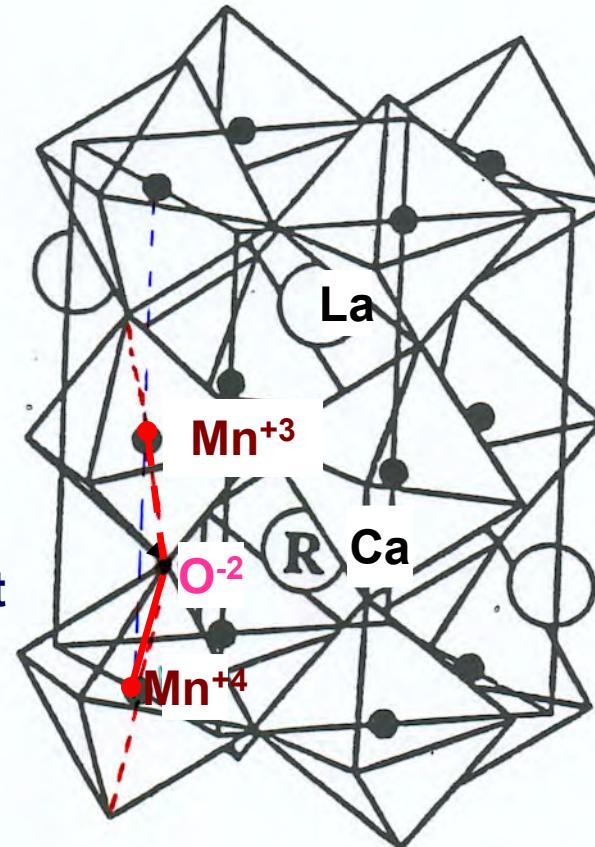
-Octahedral coordination of the Mn ions

-Mn-O-Mn bound angle 180°

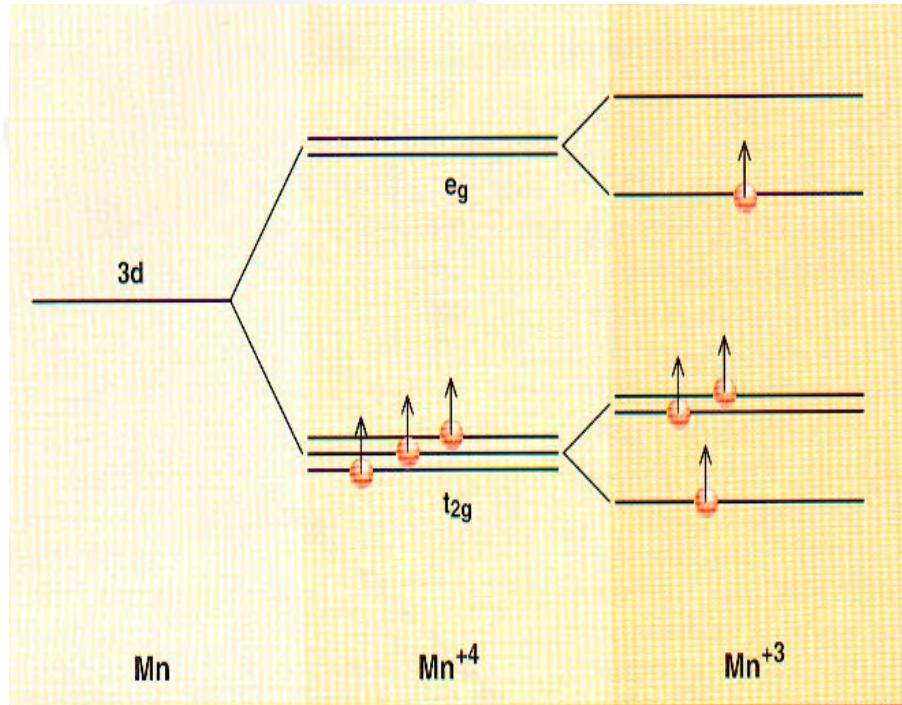
# Mixed valence manganites: Distorted perovskite structure



- Change in the bound angle due to different cation size
- Different ( $\text{La}^{+3}$ ) and ( $\text{Ca}^{+2}$ ) valence gives rise to a mixed valent state of the Mn



# Crystal electric field interaction



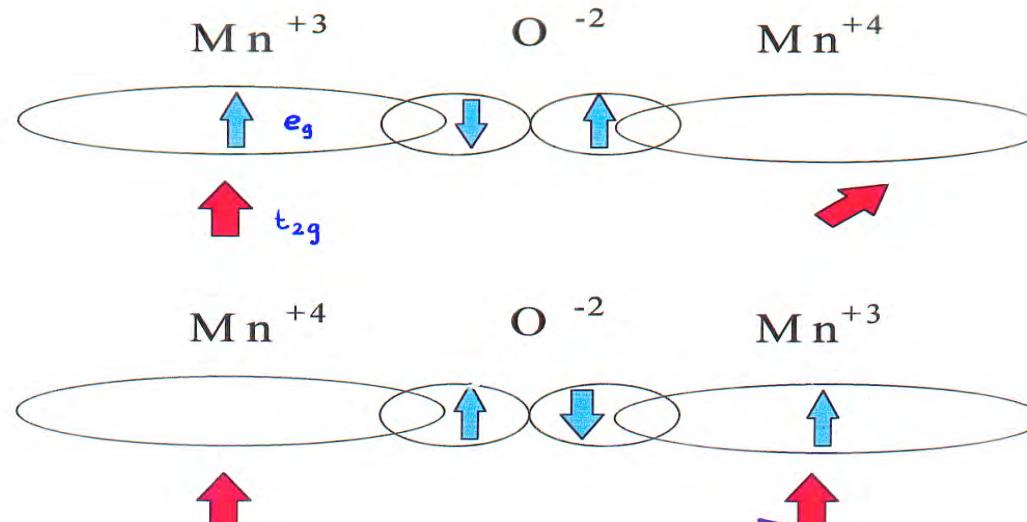
The  $t_{2g}$  electrons are localized on the Mn

The  $e_g$  level is partially occupied by an itinerant electron

Indirect interactions without overlapping of the magnetic ions charge clouds:

- Antiferro and Ferromagnetic superexchange
- Double exchange is ferromagnetic and strong

→ Mixed valence compounds; *ferromagnetic*  
**Mn<sup>+4</sup>-O-Mn<sup>+3</sup> DOUBLE-EXCHANGE**

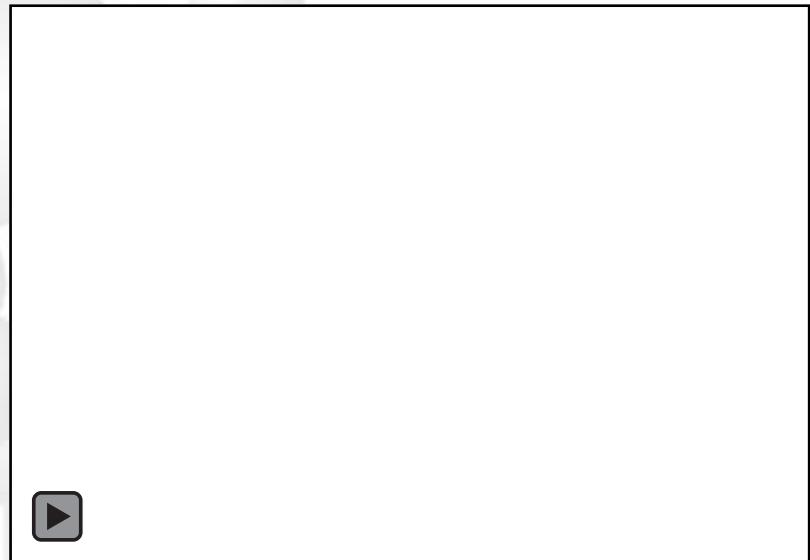


$$t = t_0 \cos[(\Theta)/2]$$

$e_g$  electron travelling in a disorder-order  $t_g$  core angular moments background



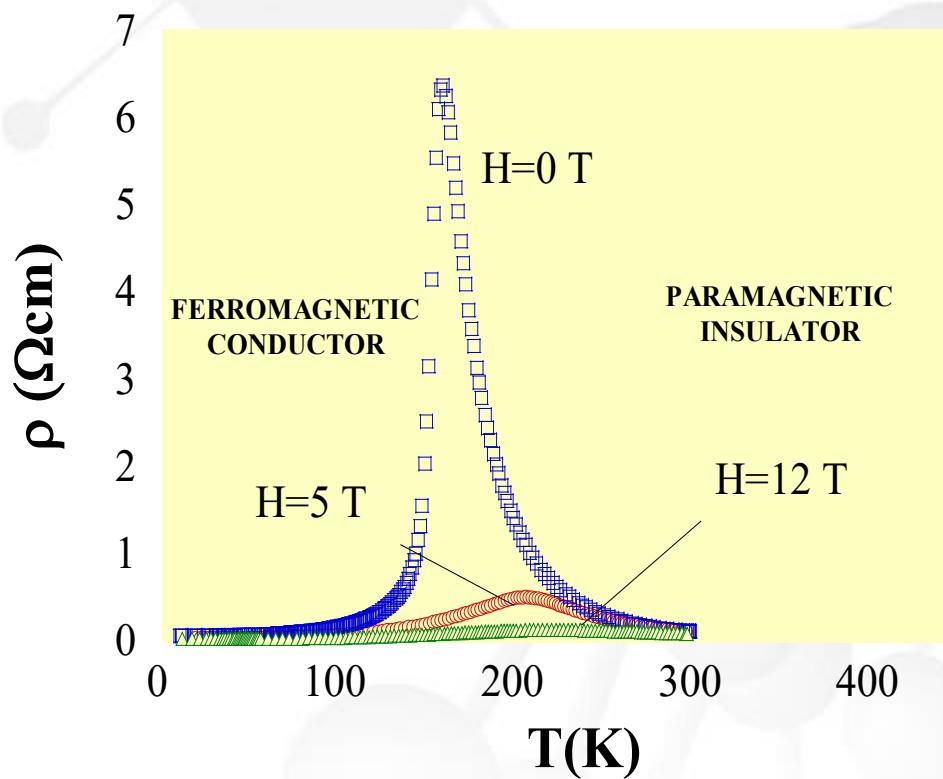
(Paramagnetic phase)



(Ferromagnetic phase)

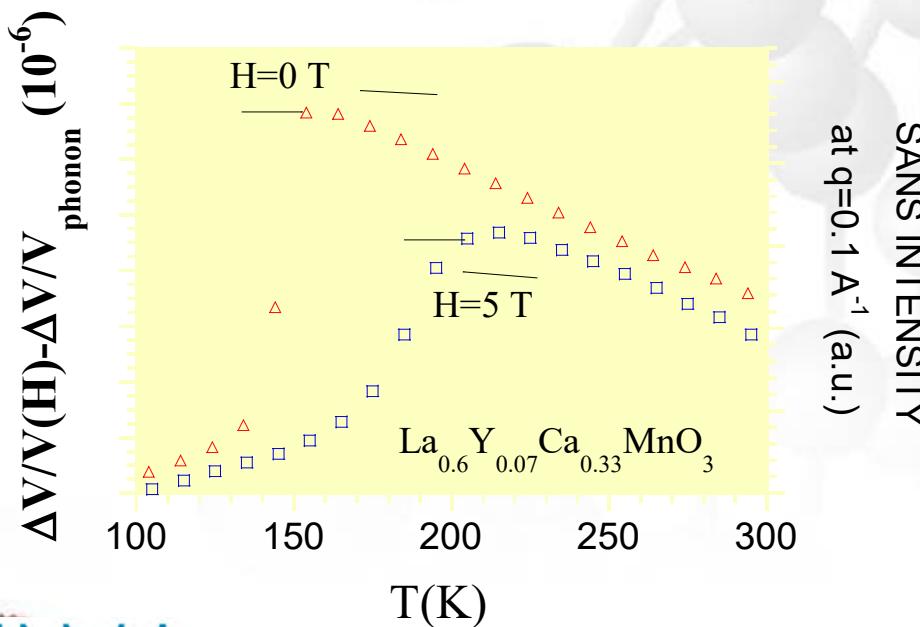
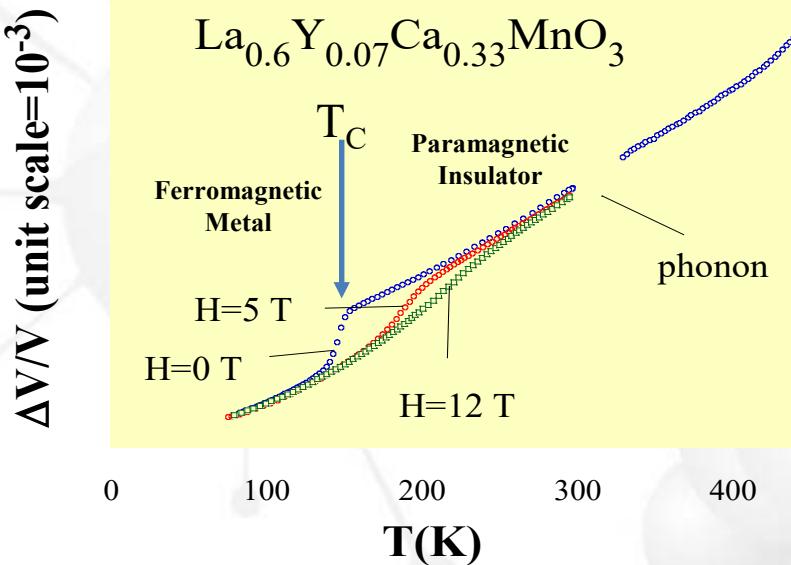
(Dr. Francisco Rivadulla courtesy)

# Colossal magnetoresistance



Magnetic and electric phase transition

Paramagnetic-Ferromagnetic  
Insulator-Metal

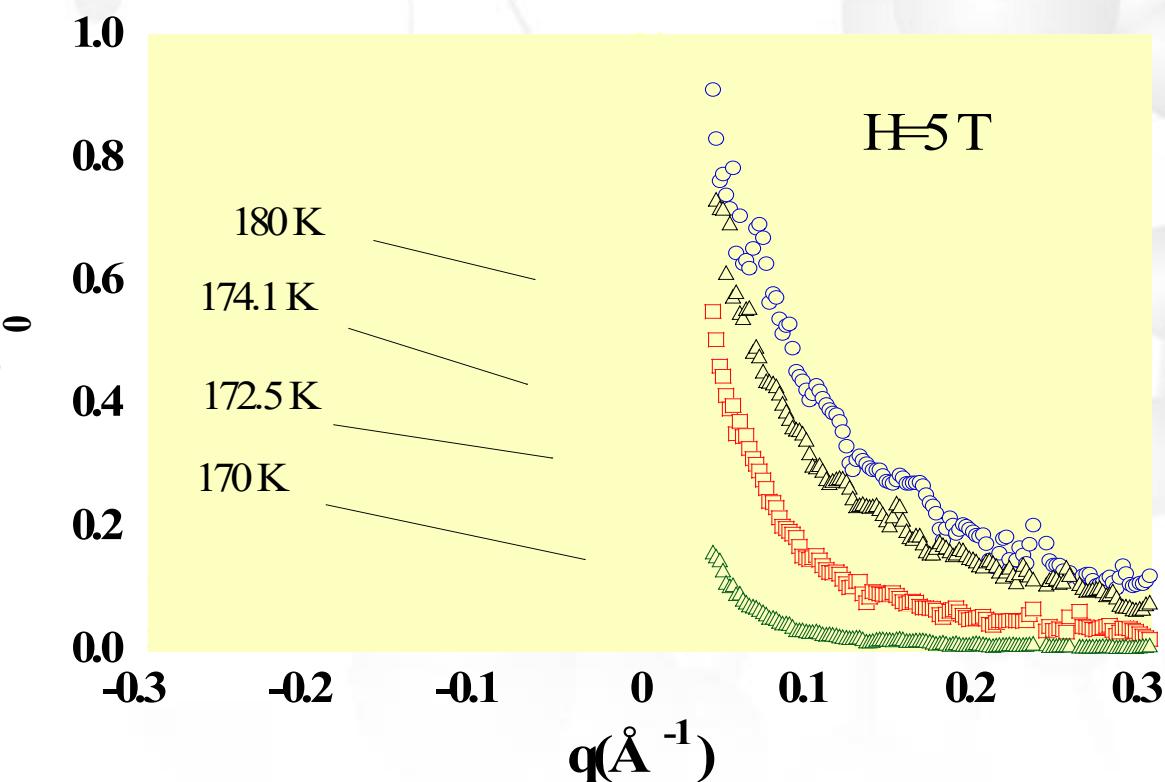
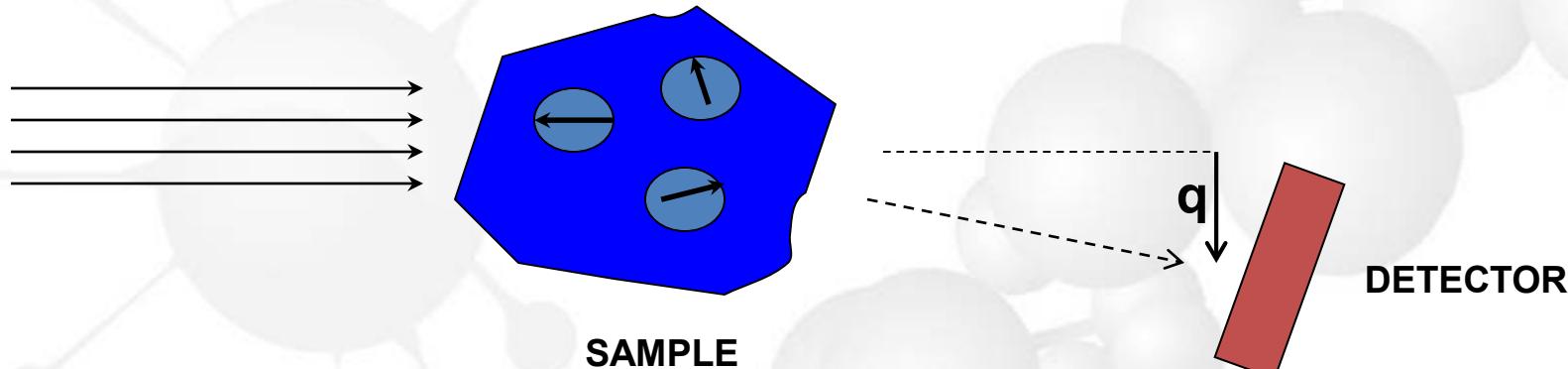


## Anomalous thermal expansion in the paramagnetic phase

M.R. Ibarra et al. Phys. Rev. Lett. 75 (1995) 3541

Small angle neutron scattering: follows the anomalous thermal expansion

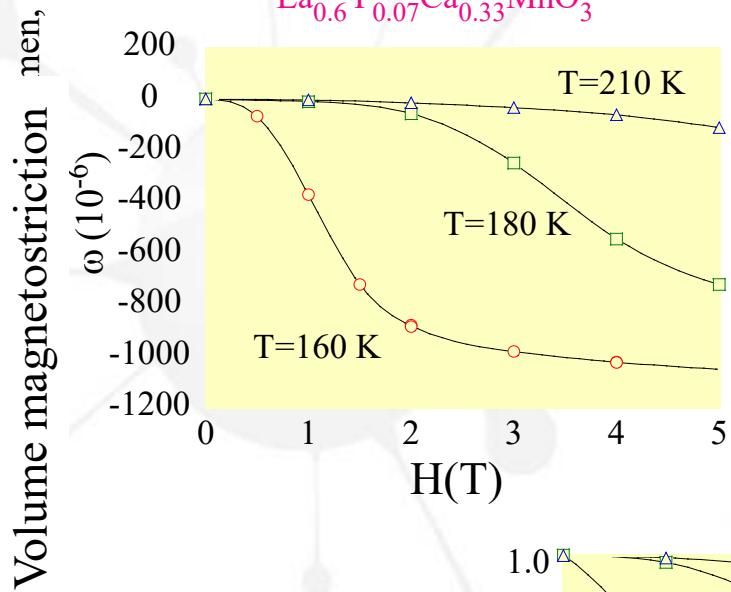
# SMALL-ANGLE NEUTRON SCATTERING (SANS)



$$\langle M(0) \cdot M(r) \rangle \sim [\exp(-r/\xi)]/r$$

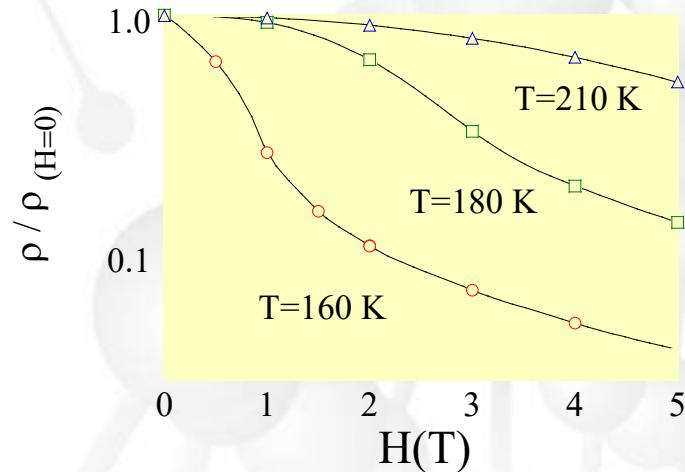


$$I = I_0 / [q^2 + (1/\xi^2)]$$

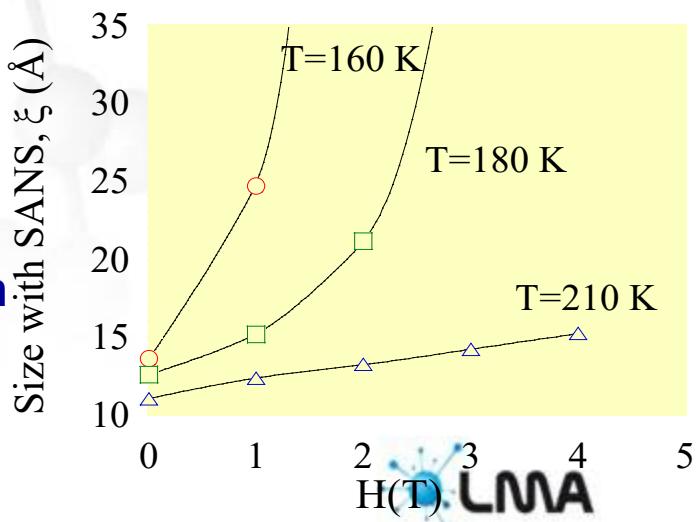


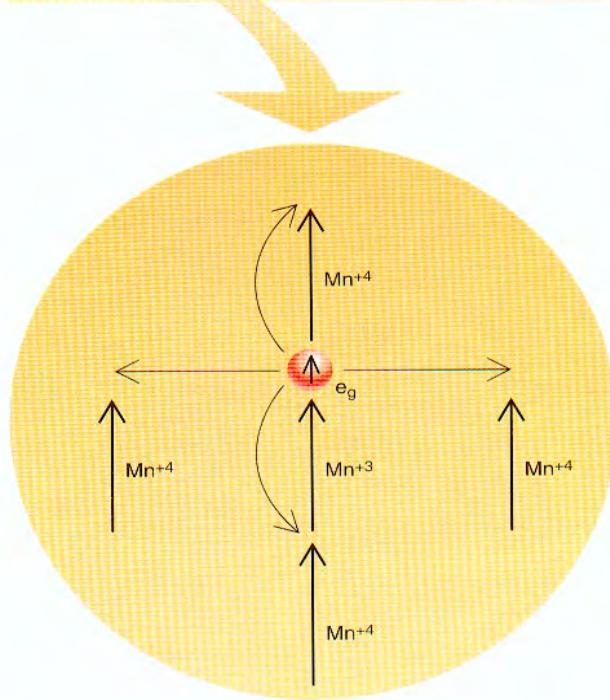
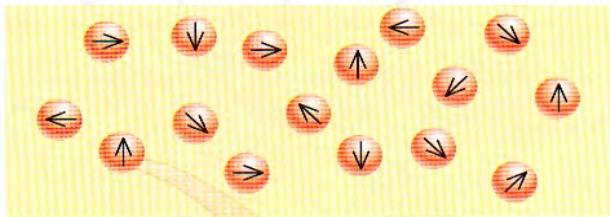
The magnetostriction in paramagnetic phase is quadratic with the field

Magnetoresistance follow the volumen deformation



The cluster size increase with the applied field and





# Magnetic Polaron

New dynamic phase segregation

De Teresa J.M. , Ibarra  
M.R.et al. Nature 386  
(1997) 256

-Hopping intra cluster  $\tau_h < 10^{-9}$  s.

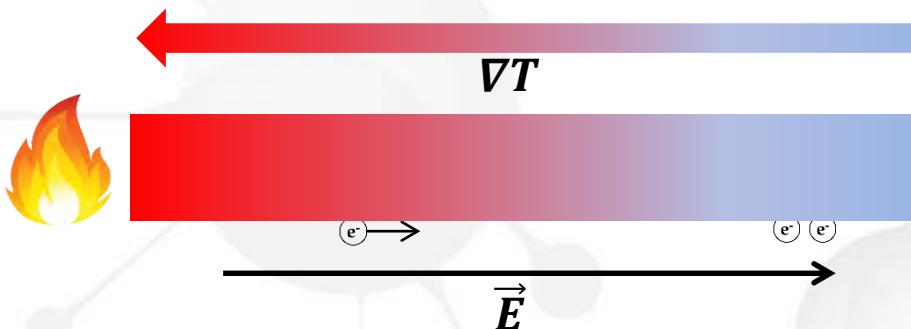
-Polaron average life time  $\tau_p > 10^{-5}$  s.





**Thin film nanostructures as multilayer constitutes  
the emergence of new thermospin effect**

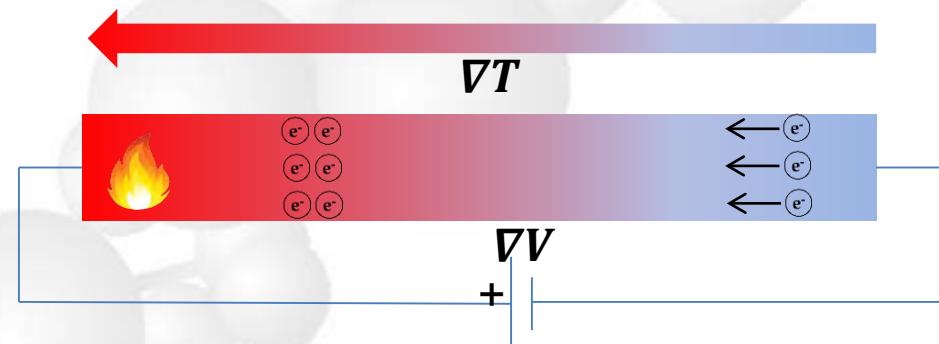
# Thermoelectric effects



$$\vec{J} = \sigma(\vec{E} - S \nabla T) = 0$$

$$\text{Seebeck effect: } S = \frac{\vec{E}}{\nabla T}$$

Thermoelectric power generation



$$\text{Peltier effect: } \Pi = S T$$

Thermoelectric cooling

## Figure of merit

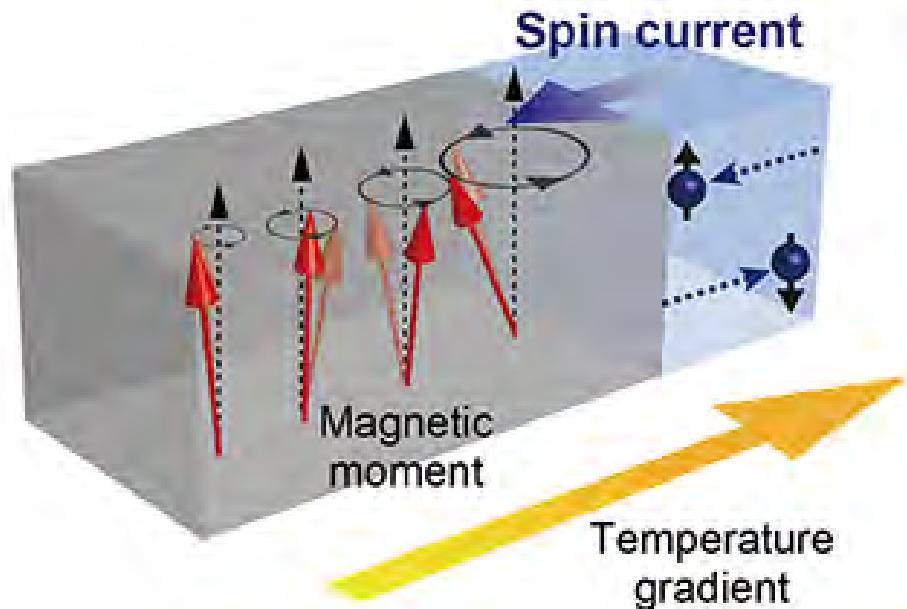
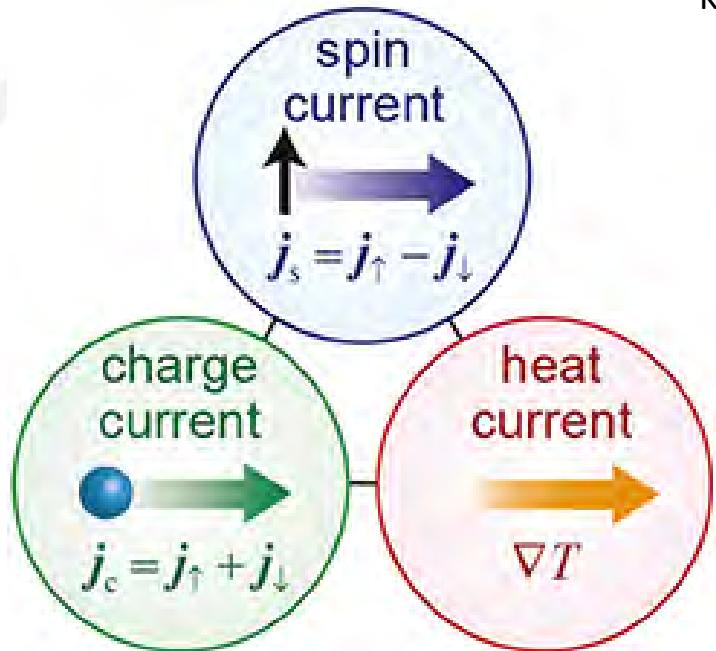
$$ZT = \frac{S^2 \sigma}{\kappa} T$$

$$\kappa = \kappa_e + \kappa_l \quad \kappa_e = L \sigma T$$

$$L = \frac{\pi^2}{3} \left( \frac{k_B}{e} \right)^2 = 2.44 \times 10^{-8} \text{ W}\Omega/\text{K}^2$$

# Spin Seebeck effect effect: Spin current generation by heat

K. Uchida et al. Nature 455, 778 (2008)

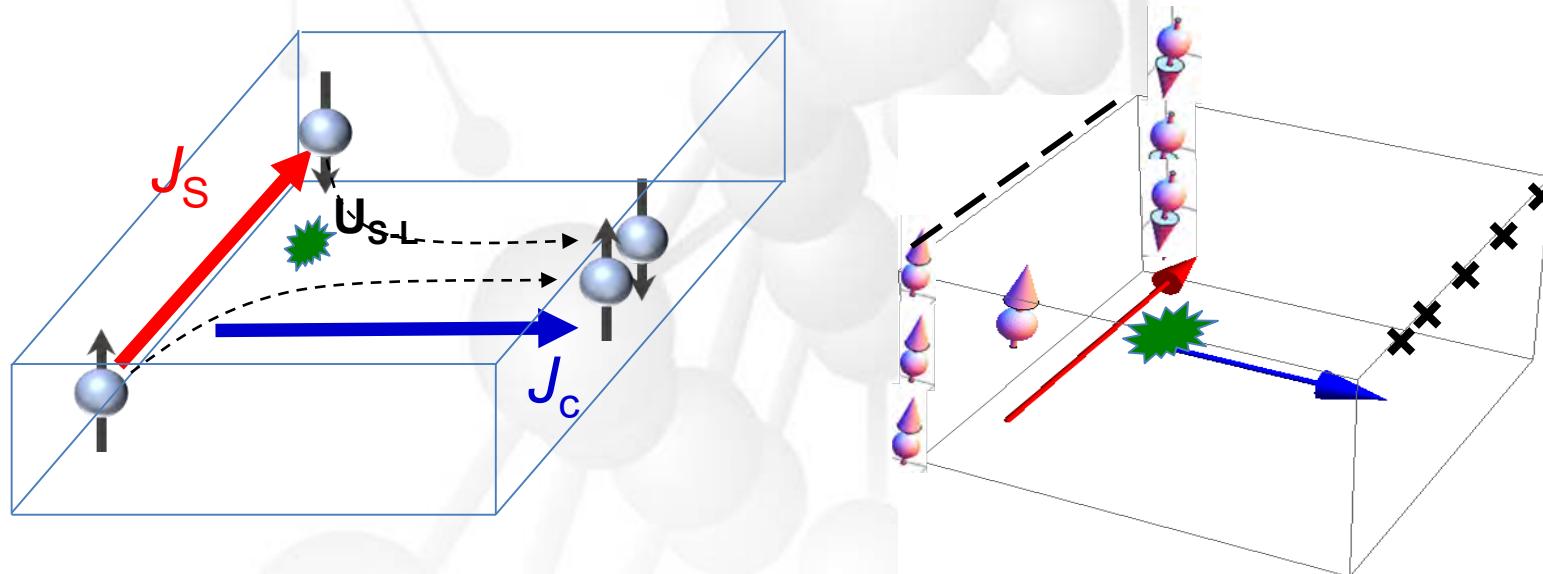


$$I_S = -G_S \frac{k_B}{\hbar} (T_F - T_N)$$

J. Xiao et al. PRB **81**, 214418 (2010)  
H. Adachi et al. PRB **83**, 094410 (2011),  
Rep. Prog. Phys. **76**, 036501 (2013)

# Inverse Spin Hall effect (ISHE)

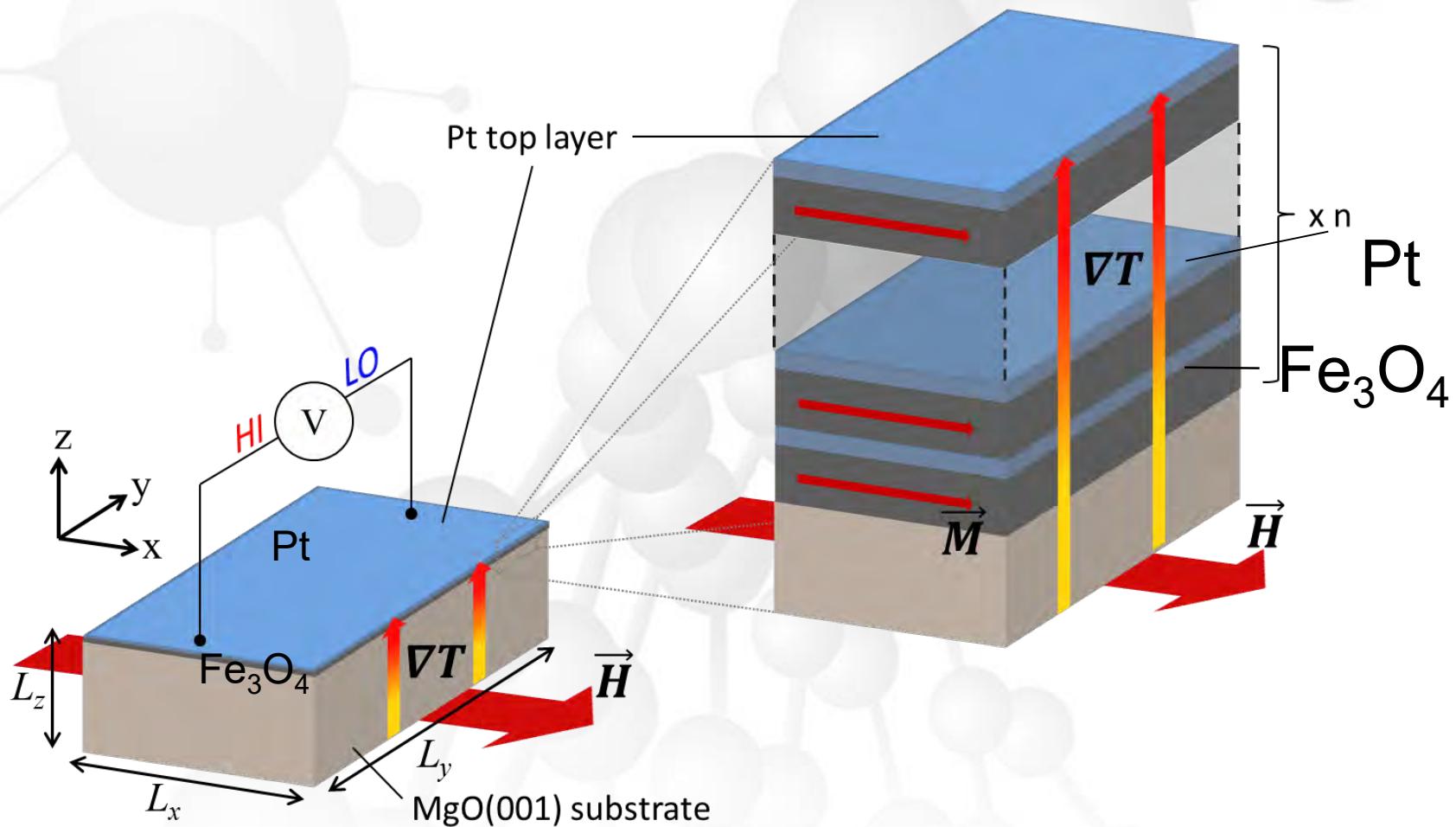
Interconversion of spin currents – charge currents in non-magnetic metals with high spin orbit coupling (high Z)



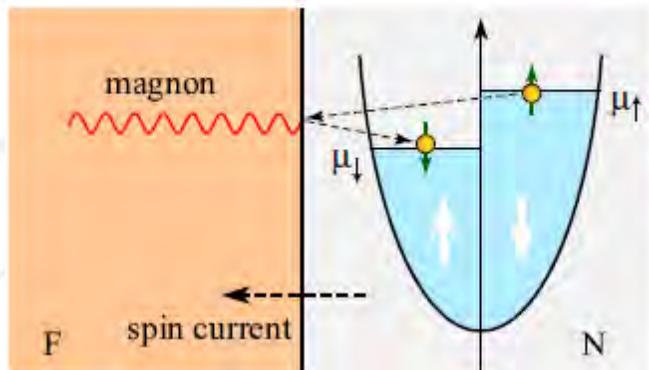
( $J_s$ ) Spin  $\longrightarrow$  ( $J_c$ ) Charge

E. Saitoh et al. Appl. Phys. Lett. 88, 182509 (2006)

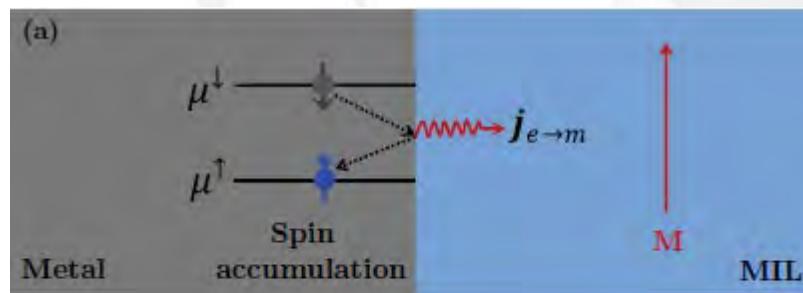
# SSE in $[F/N]_n$ multilayers



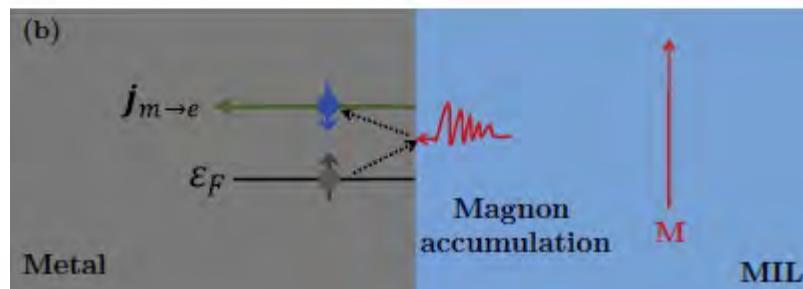
# SPIN CURRENT AT THE INTERFACES



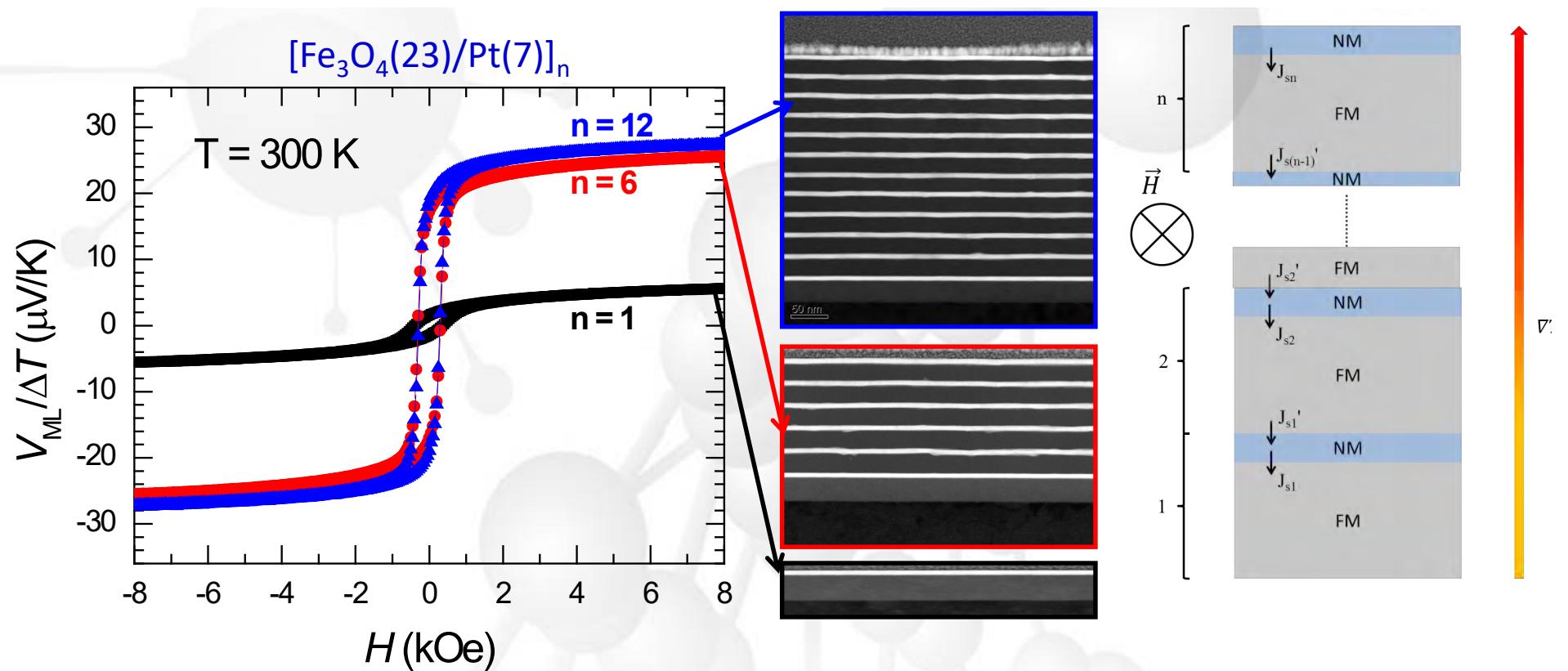
Magnon emission associated with spin accumulation at the metal-ferromagnet interface  
(Takahasi et al ICM 2009)



Spin angular momentum transfer at the interface:  
Magnon and electron spin current interconversion  
(Steven et al. PRB 86 (2012) 214424)



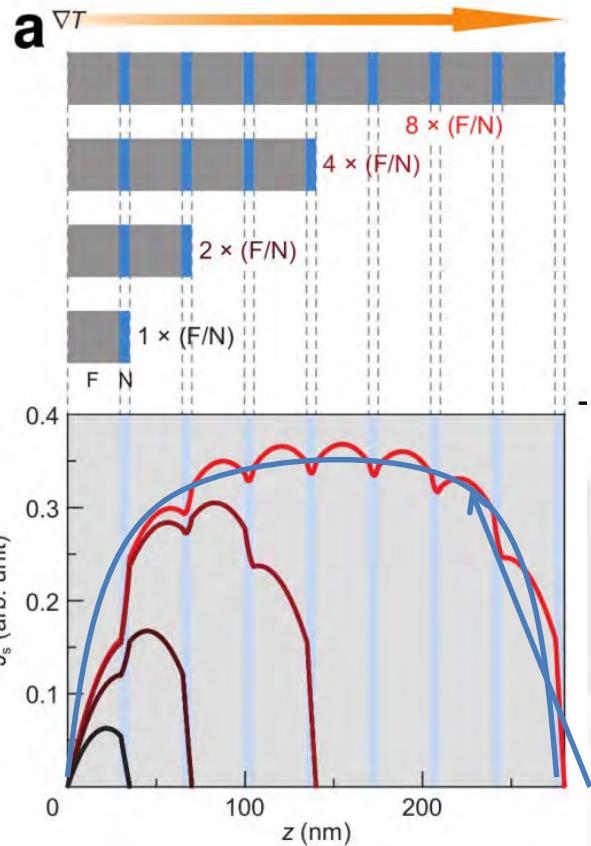
# Optimized configuration



Largest SSE voltage measured in a thin film based structure!!

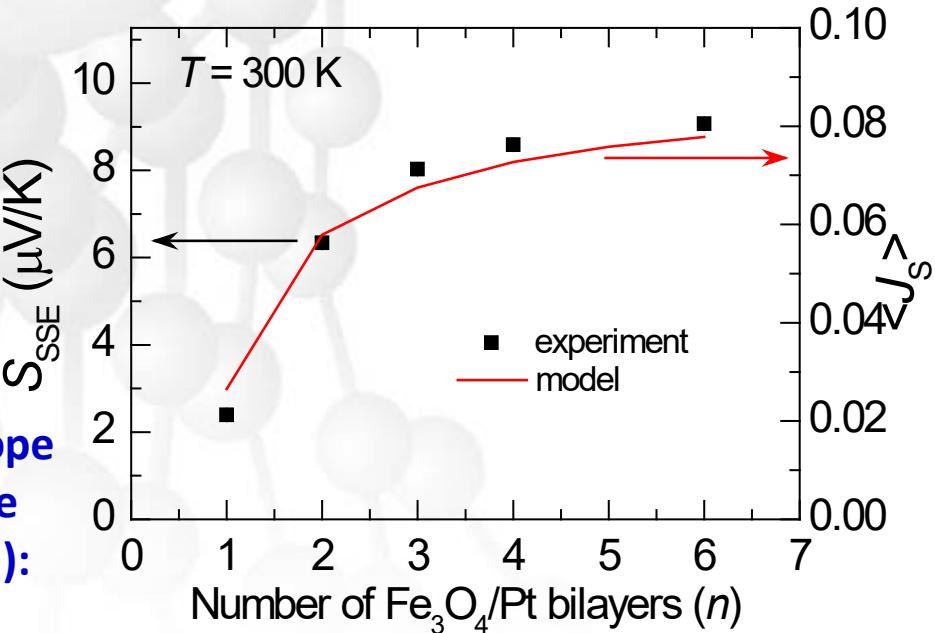
$$V_{ML} \approx 28 \mu\text{V}/\text{K} !!$$

# Qualitative agreement with experimental results



$$\langle J_S \rangle = \frac{1}{t_N n} \sum_{i=1}^n \int_{z_i=0}^{t_N} dz J_s^{(i)}(z)$$

Average SSE voltage measured:



Maximum spin current at central interlayers

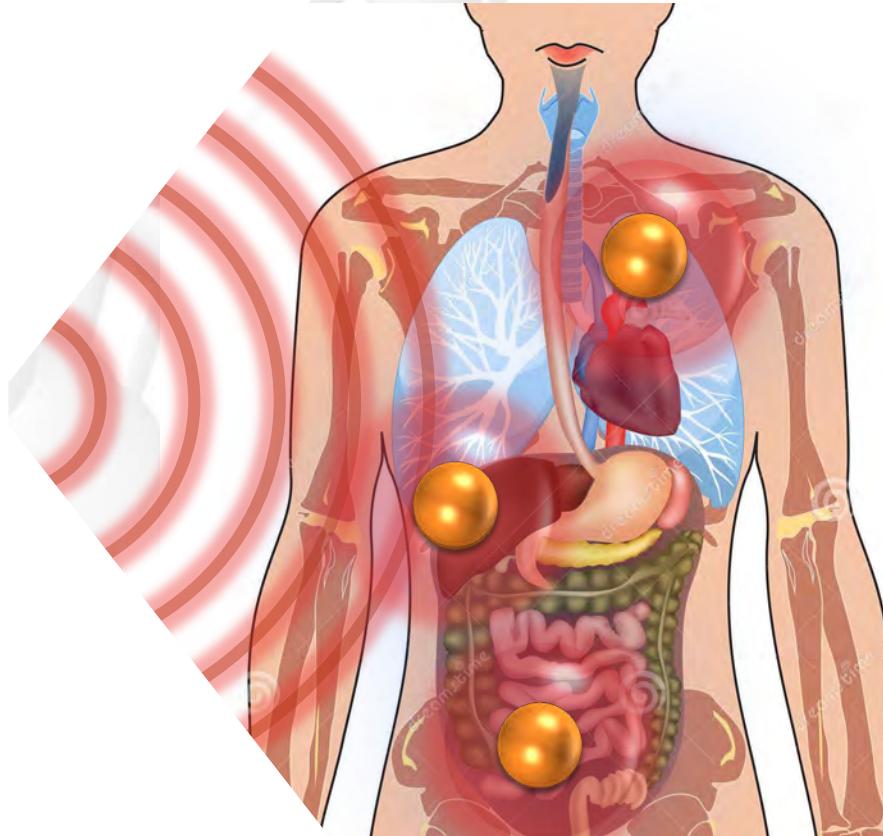
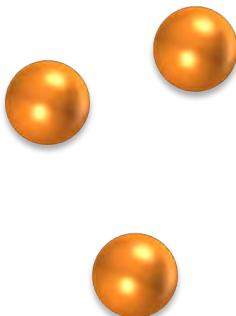
Ramos et al. Phys. Rev. B **92**, 220407(Rap. Comm.) (2015)

# Magnetic nanoparticles, due to the electromagnetic radiation adsorption in the radiofrequency range, operate as nanoheaters



*Nanoscale*, 2019, 11, 3164-3172

- **Magnetic hyperthermia** is an experimental treatment for cancer.
- Based on the fact that magnetic nanoparticles can transform electromagnetic energy from an external a.c. field to heat.
- If magnetic nanoparticles are put inside a tumor and the whole patient is placed in an a.c. magnetic field, the tumor temperature will rise.
- The elevation of temperature may enhance radio- and chemo-sensitivity, hopefully shrinking tumors.



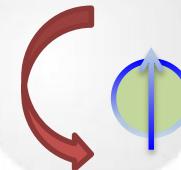
INMA

# Losses in magnetic colloids

1. In NPs suspensions (@ RT), the Brownian relaxation in viscous media is

$$\tau_B = \frac{3 \eta V_H}{k_B T}$$

Brownian rotation

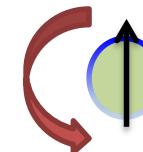


Physical movement of the MNPs

2. Néel relaxation is

$$\tau_N = \tau_0 \exp\left(-\frac{K V_M}{k_B T}\right)$$

Néel relaxation



Rotation of the magnetic moment of the MNPs

The total relaxation is

$$\frac{1}{\tau} = \frac{1}{\tau_B} + \frac{1}{\tau_N}$$

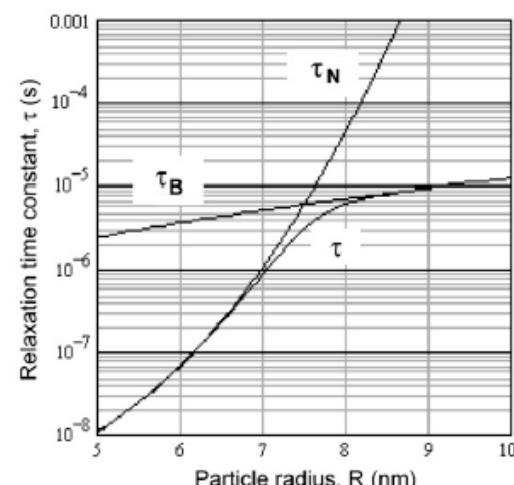
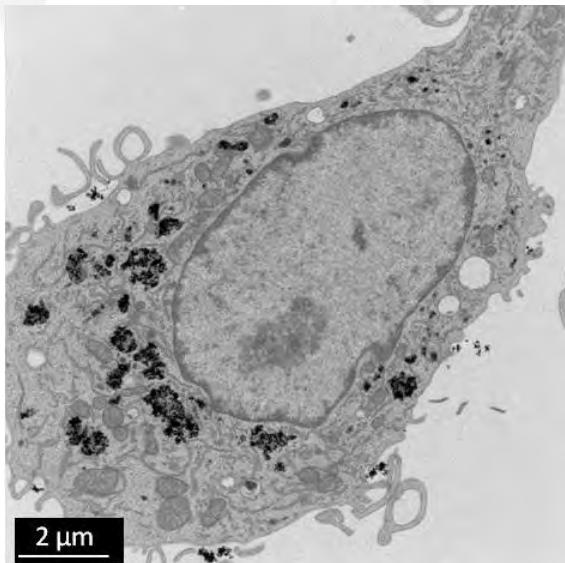
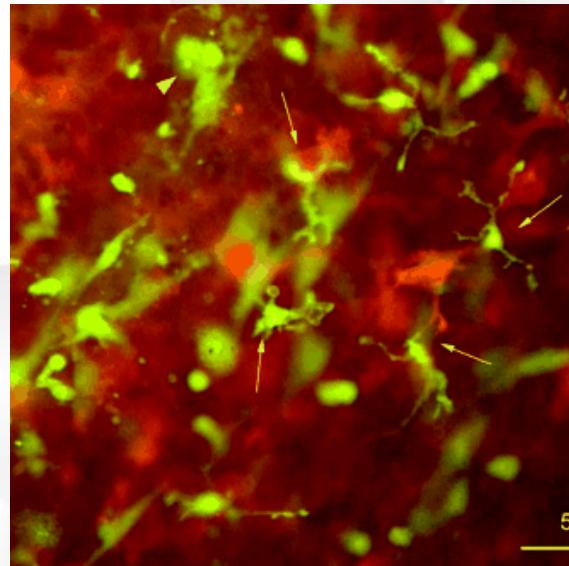


Fig. 2. Time constants vs. particle size for magnetite particles.

# Dendritic cells targeting carrying MNPs: magnetic cells



Dendritic cells +  
NPs



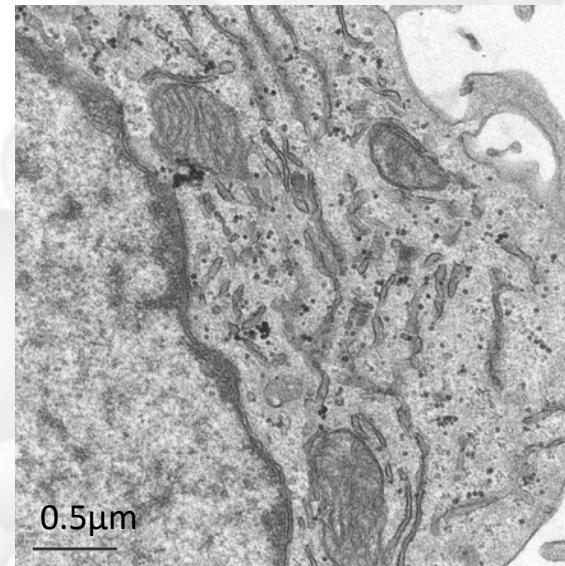
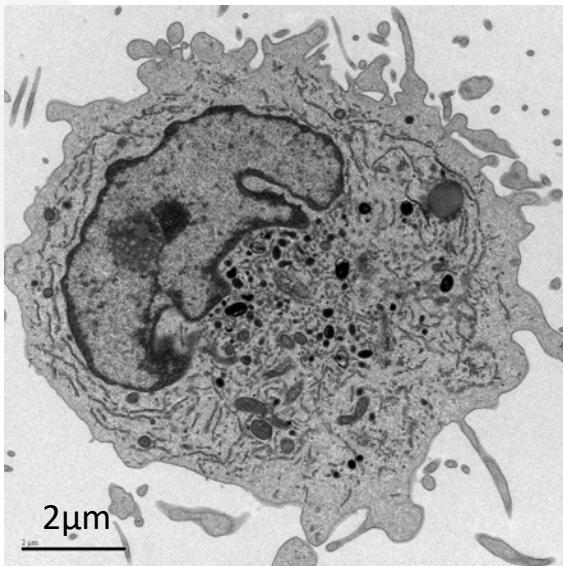
Dendritic cells  
targeted on tumor



Trojan horse

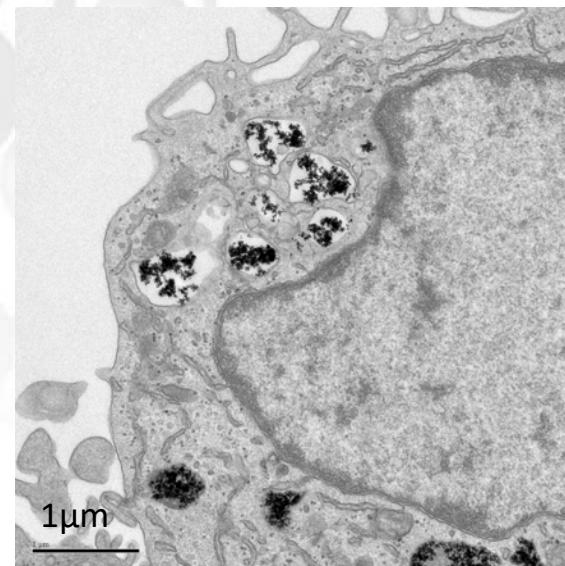
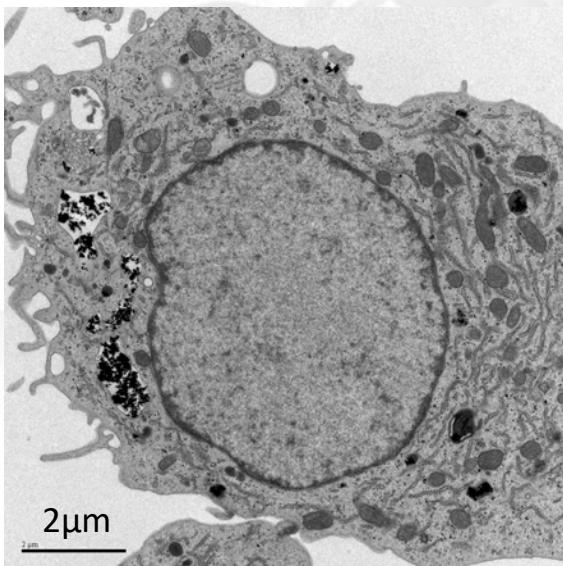
# DCs INTERNALIZATION-TEM

DCs

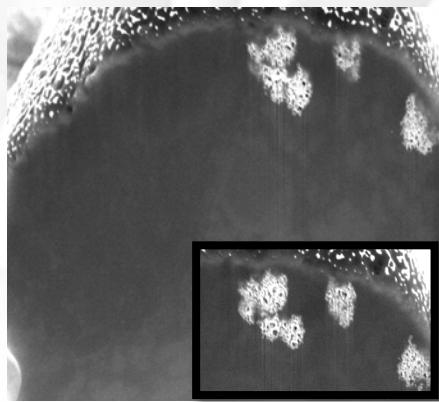
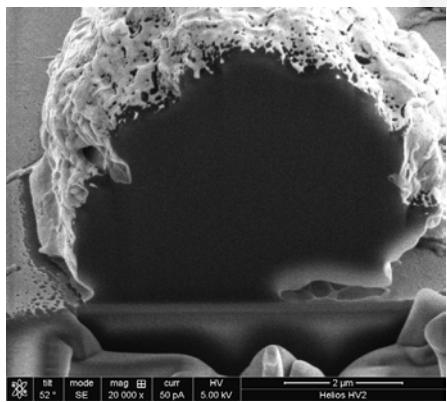


DCs+  
MNPs

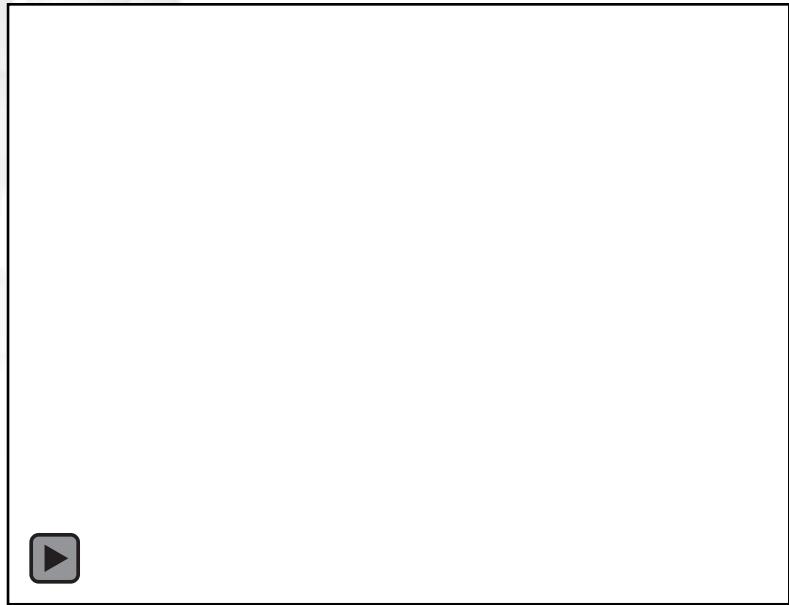
50 ug $\text{Fe}_3\text{O}_4$ /ml



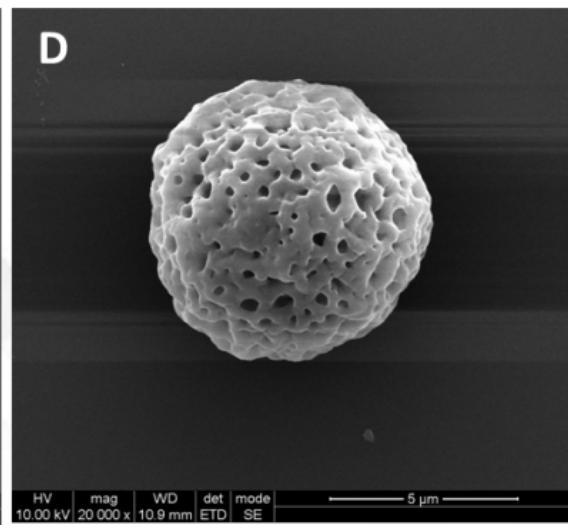
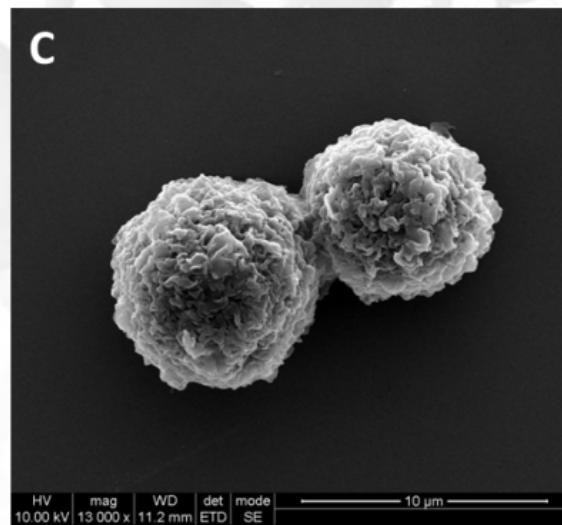
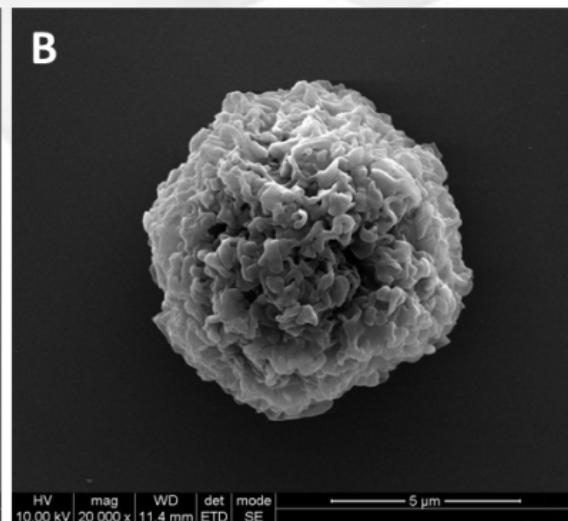
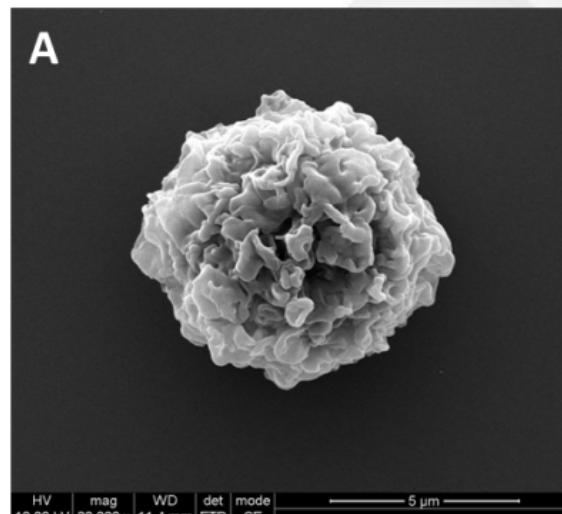
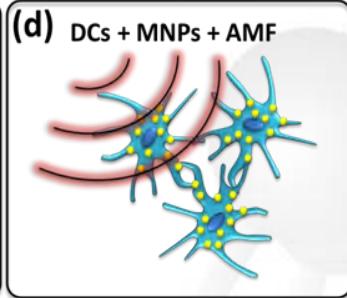
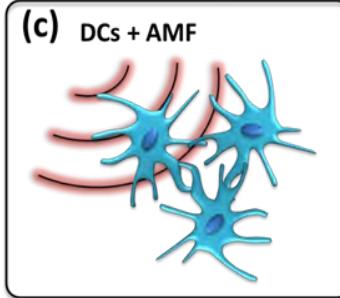
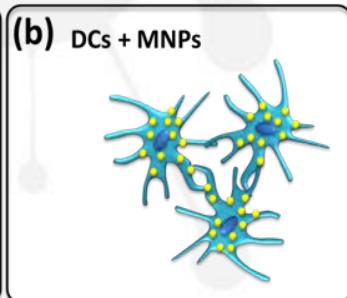
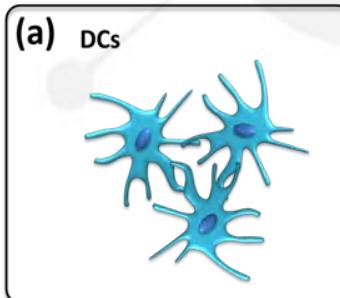
# Focused Ion Beam FIB - Dual Beam



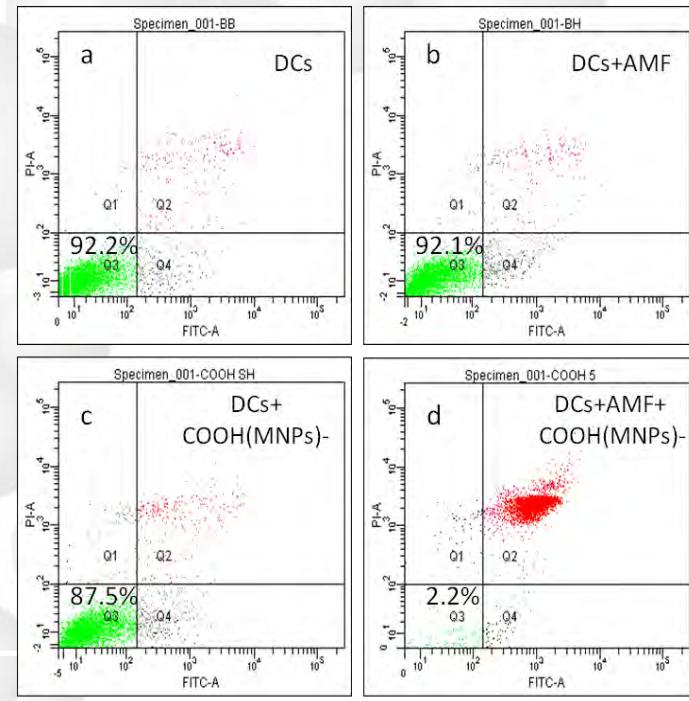
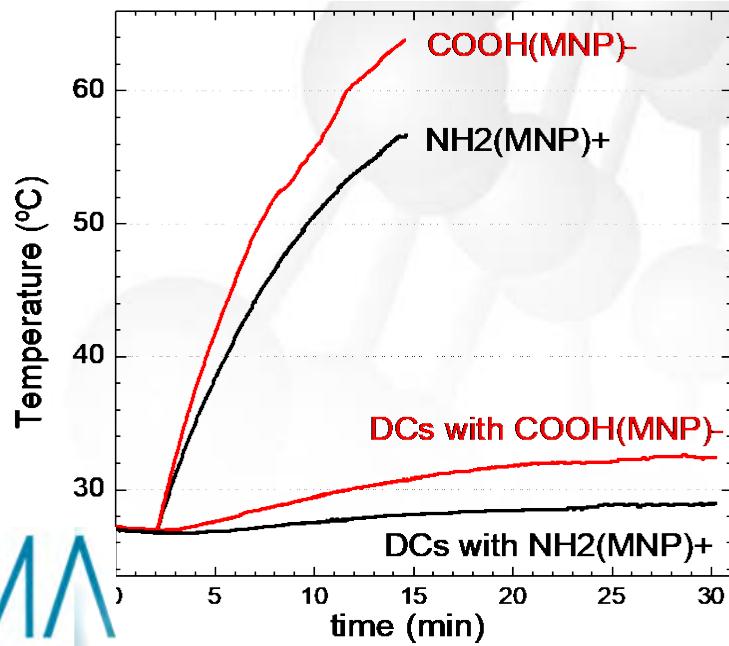
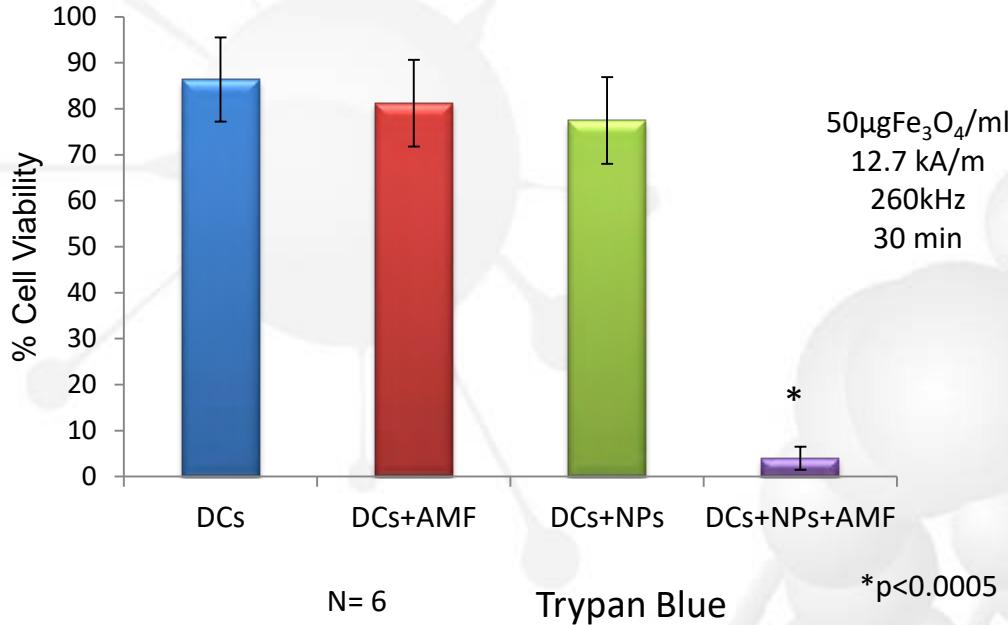
MNPs uploaded DCs



# EFFECT OF THE ELECTROMAGNETIC FIELD ON CELL VIABILITY



# DENDRITIC CELL VIABILITY

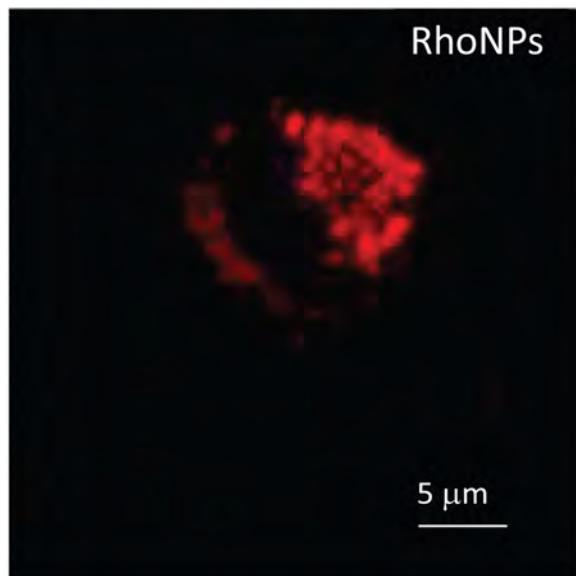


*Cell death induced by the application of alternating magnetic fields to nanoparticle-loaded dendritic cells.*  
 Marcos-Campos I, Asín L, Torres TE, Marquina C, Tres A, Ibarra MR, Goya GF. Nanotechnology 22 (2011) 205101

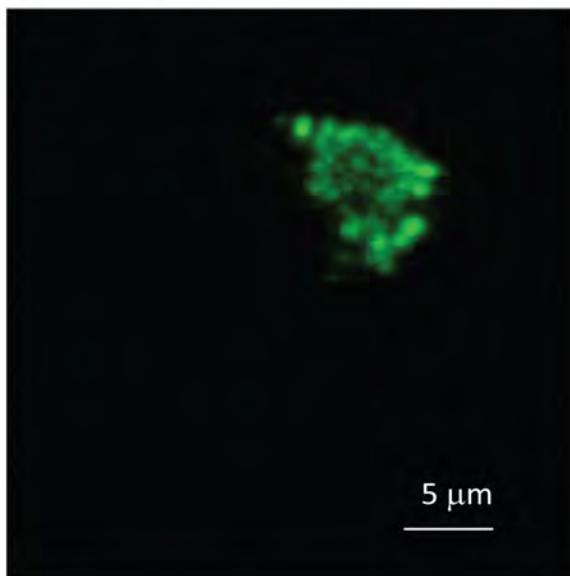
No temperature increase!!!

# Colocalization of MNP in DCs

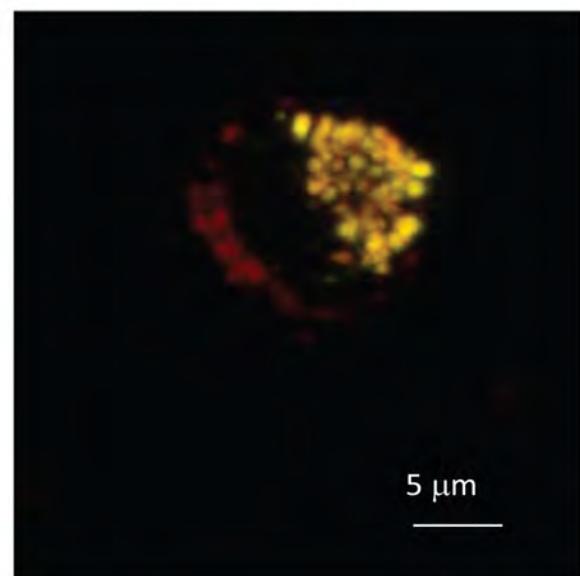
Fluorescence NPs



Lysotracker

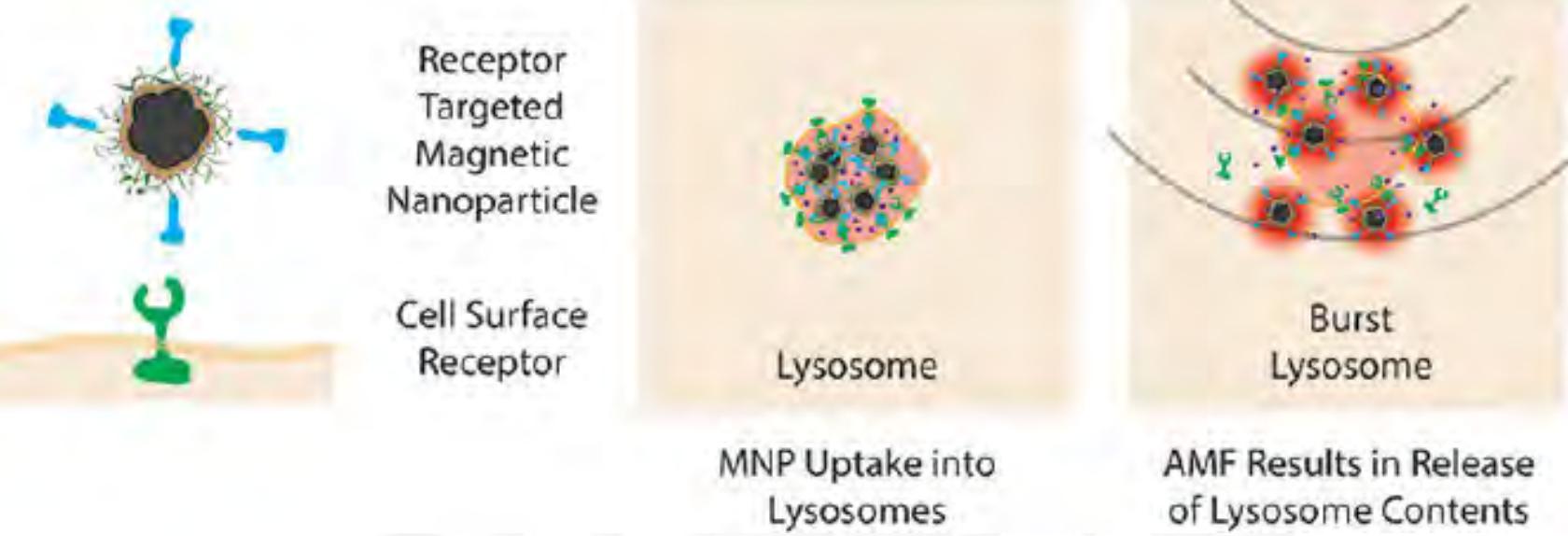


Overlay



Goya G.F. et al. Current Nanoscience **12** (2016).

# Lysosomal Membrane Permeabilization by Targeted Magnetic Nanoparticles in Alternating Magnetic Fields



Domenech et al. ACS nano (2015)

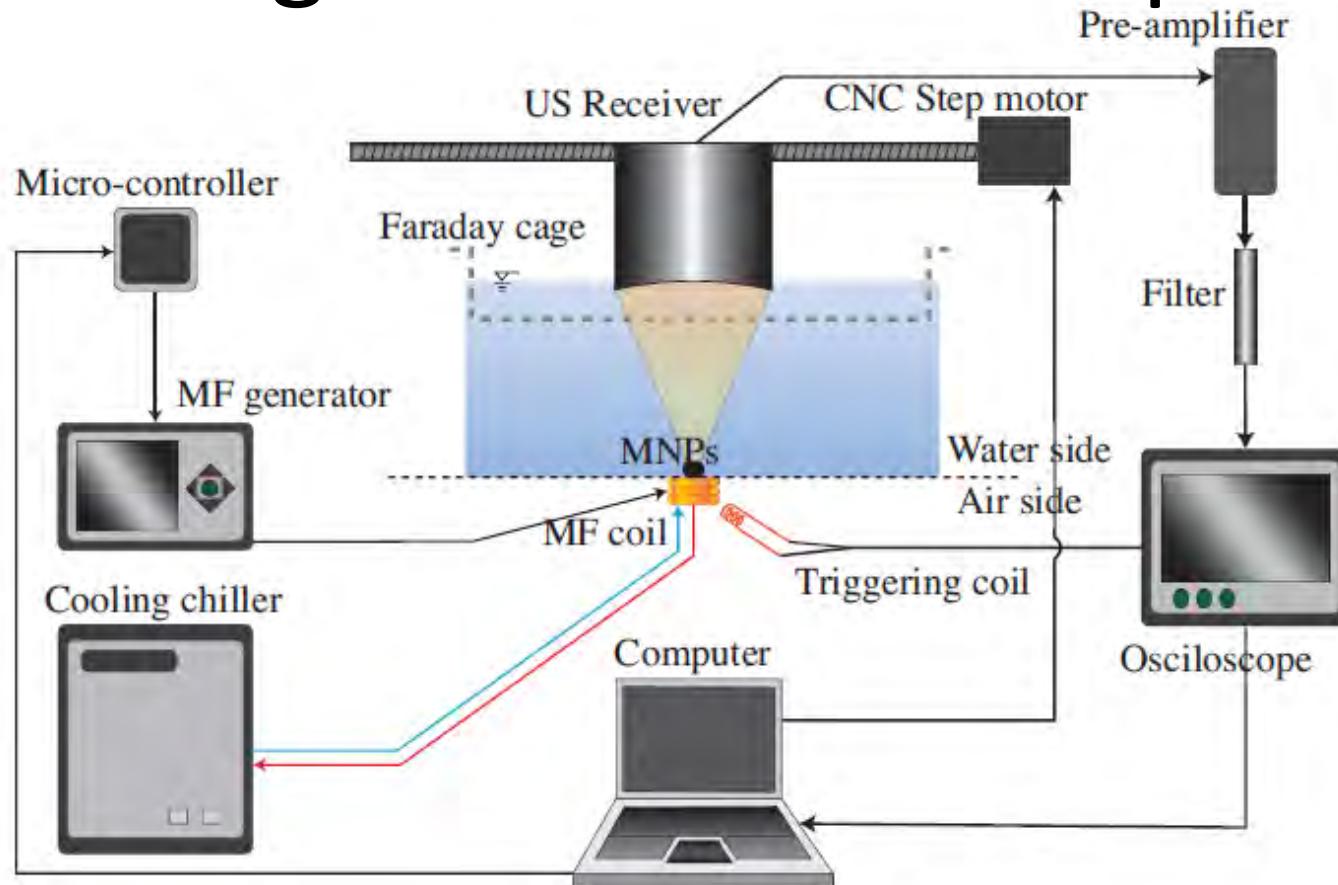
# **Induced ultrasound generation**

Mechanism for membrane disruption?

Mechanical waves, Ultrasound?

(In collaboratio with Prof. Gullermo Rus, UGR)

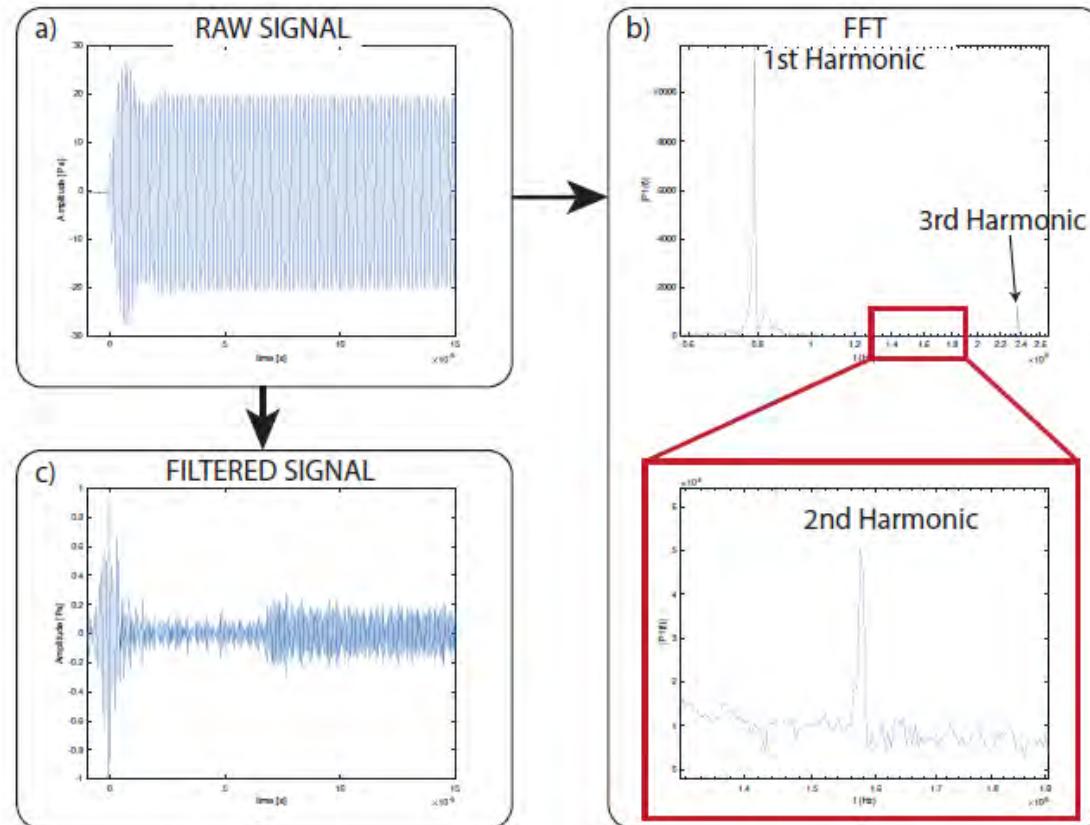
# Magneto-acoustic setup



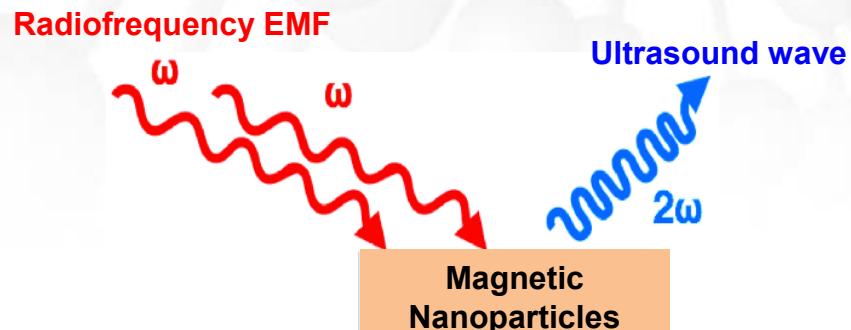
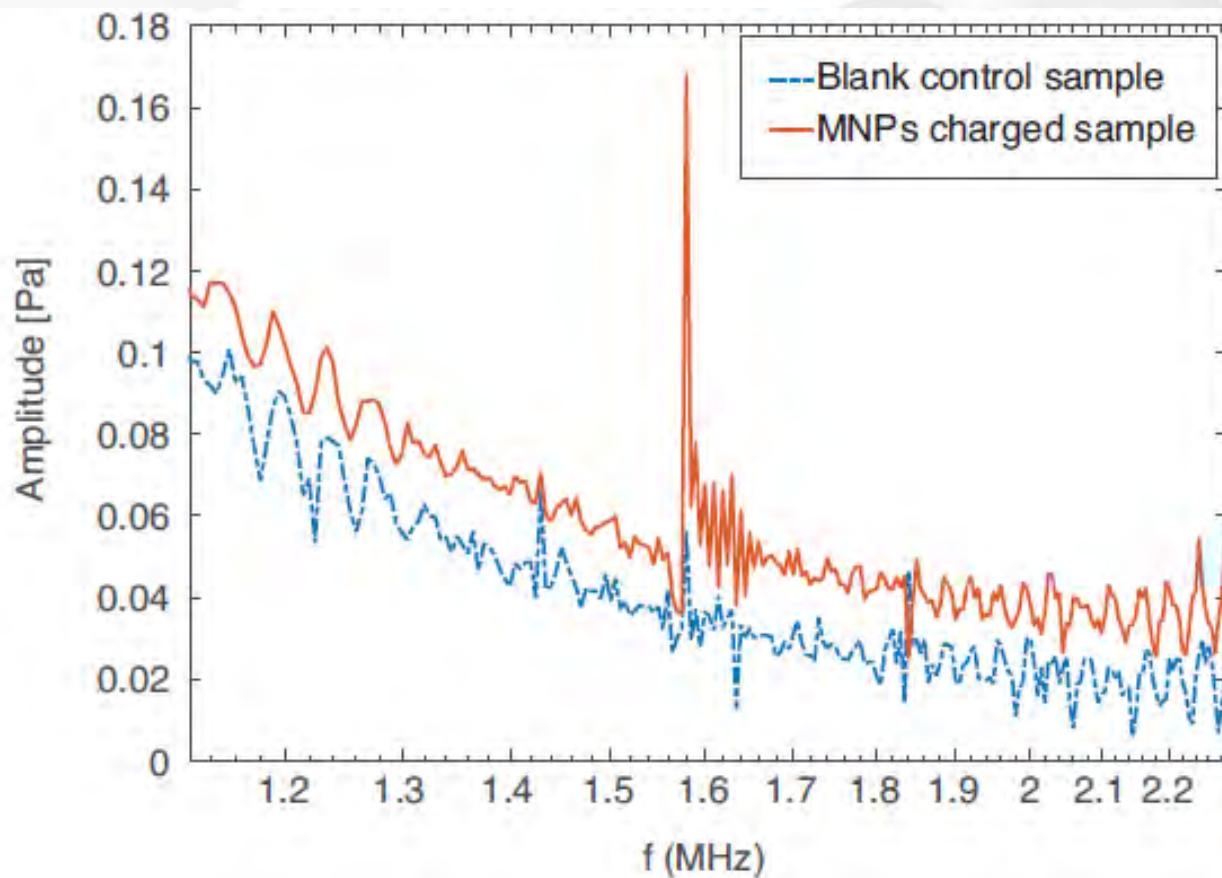
Experimental goals:

- Thermal stability (Short EMF burst)
- Lack of interferences and high sensitivity
- EMF gradients

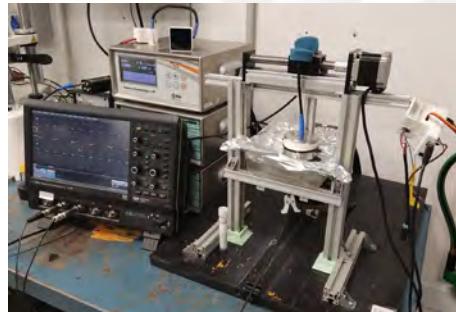
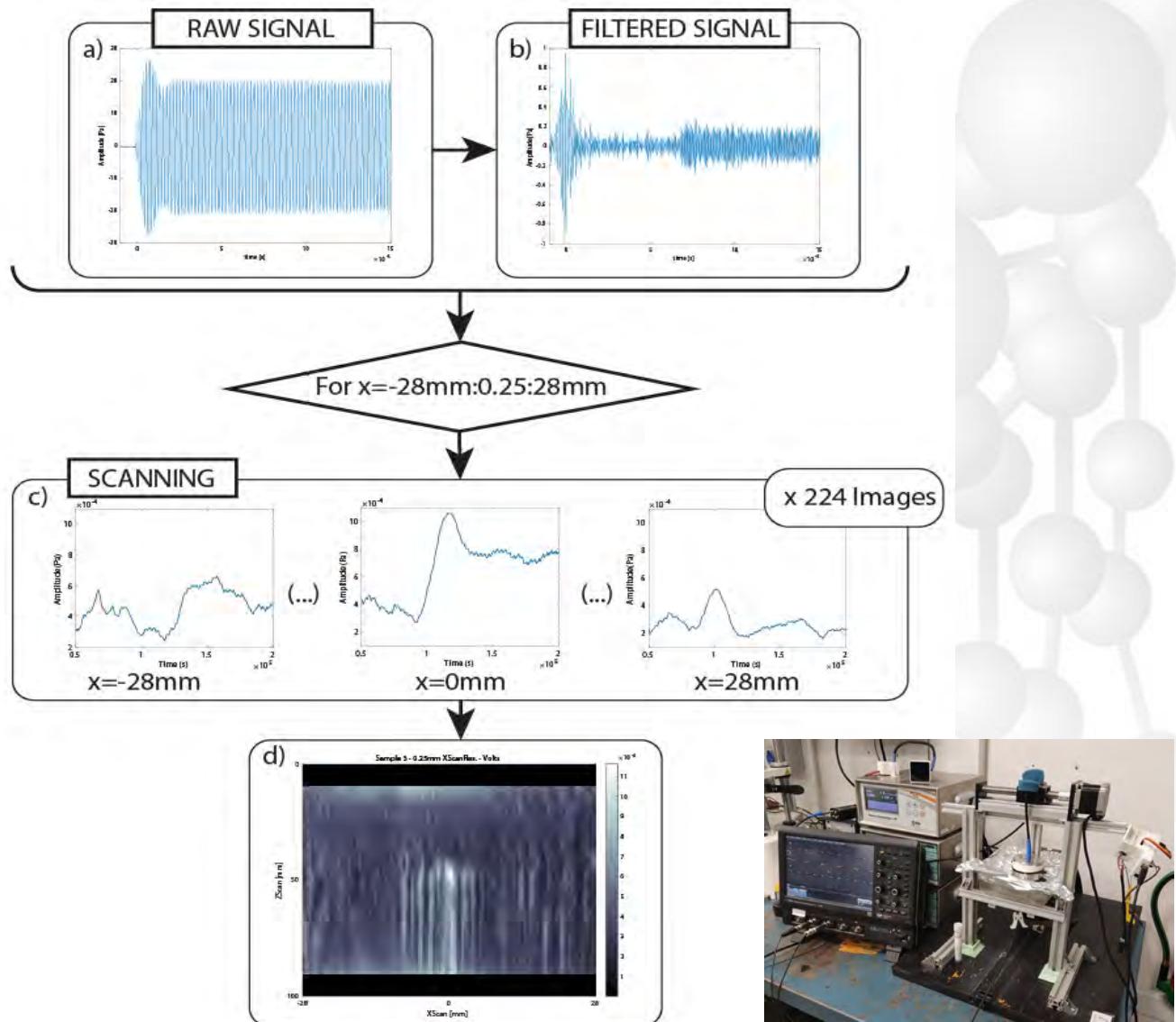
# Ultrasound response to the EMF



## US signal due to the MNP



# Prototype for scanner



A faint, light-gray molecular structure diagram consisting of numerous small circles connected by thin lines, forming a complex network of nodes and edges.

**Nanomagnetism provides new tools  
that allows a deep understanding of  
the phenomena that occurs at the  
nanoscale even at atomic level**

**This will allows to design new  
functional materials.**



# LMA

# LABORATORIO DE MICROSCOPIAS AVANZADAS

**THANK YOU FOR YOUR ATTENTION**

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