Magnetosomes as a pure system for the insight of magnetic features in magnetite nanoparticles

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ABSTRACT

Thanks to their high magnetic moment and biocompatibility, magnetite nanoparticles awaken a growing interest in different areas of science and especially in biomedical applications[1,2]. Aiming to employ these particles in any future application is crucial to understand the main features of nanoshaped magnetite. Even when bulk magnetite is a well-studied material[3], at nanoscale, magnetite presents some features still under debate. However, the critical issue to perform fundamental studies in nanoparticles arises from the difficulties in the reproducibility and control of the main properties of the synthesized particles by means of chemical and physical routes.

In this regard, magnetotactic bacteria of the strain *Magnetospirillum gryphiswaldense*, able to orient in and navigate along geomagnetic fields thanks to the presence of one or more chains of magnetite nanoparticles[4], offer a natural route to obtain magnetite nanoparticles with exceptional properties. The high biological control imposed in the synthesis of magnetosomes leads to well-defined properties with a high structural and chemical purity. Particularly, this bacterial strain synthesizes cubo-octahedral shape magnetite nanoparticles with a mean size of 45 nm-diameter. In the present study, we benefit from the advantages of magnetosomes to understand the observed magnetic features in magnetite nanoparticles by means of a combination of DC and AC magnetometry.

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EFFECT OF THE 1,2-HEXADECANEDIOL CONTENT ON THE MAGNETIC PROPERTIES OF Co-FERRITE NANOPARTICLES

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ABSTRACT

In recent years, ferrite nanoparticles with composition MFe_2O_4 (M=Fe,Co) have attracted much interest due to their potential applications in fundamental magnetism, biomedical applications and ultra-high density magnetic recording. However, ferrite NPs show magnetic properties with high variability depending on the preparation method, which limits the reproducibility and crystal quality of the samples. For this reason, a careful control of the synthesis conditions is required. This work reports on the effect of the 1,2-hexadecanediol content on the structural and magnetic properties of Co-ferrite nanoparticles synthesized by thermal decomposition of metal-organic precursors in a highly-boiling point organic solvent, using oleic acid as surfactant and stabilizer.

MAGNETOCALORIC EFFECT OF PARTIALLY AMORPHOUS FE70ZR30 POWDERS PRODUCED BY BALL MILLING

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ABSTRACT

The ability of mechanical alloying to produce metastable phases makes it widely used to produce materials with interesting magnetocaloric effect (MCE). Although this technique presents various advantages (e.g. prevent the loss of volatile elements), several aspects inherent to powder samples, such as the demagnetizing field, the possible multiphase character (due to remnant impurities or contamination) and a probable distribution of Curie temperatures, affect MCE and particularly the critical exponents derived from MCE.
The present work studies the exponent n describing the magnetic entropy change of several partially amorphous Fe70Zr30 alloys produced by ball milling with remnant α-Fe nanocrystals. The magnetization was measured using a LakeShore 7407 vibrating sample magnetometer and the MCE was calculated from Maxwell relation. An increase of magnetic entropy change at the peak and n also increases above Curie temperature of the amorphous phase being bellow 2 due to the remnant nanocrystalline phase.

ADDRESSING THE MAGNETIC HEAT RELEASE OF MAGNETIC NANOPARTICLES IN LIVE CELLS

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ABSTRACT

In the last decades, magnetic nanoparticles (MNPs) have found an increasing number of biomedical uses, such as drug delivery or gene transfection nanocarriers, magnetic separators, magnetic transducers or imaging tracers. Among them, magnetic hyperthermia is emerging as a cancer treatment based on the MNPs heat losses when subjected to alternating magnetic field (Hac). The magnetic heat release of MNPs is exchanged to their surrounding, applying thermal stress to tumour tissues in order to activate cell death mechanism (apoptosis/necrosis) while minimizing side effects. This local, minimally-invasive and remotely activated therapy, together with negligible toxicity, provide a promising potential against cancer. For clinical purpose, the magnetic heat of MNPs has to remain invariable in any biological environment. However, recent studies evidence that the magnetic heating of MNPs is significantly altered in biological matrices, resulting in strong variations inside cells or tissues respect to colloidal dispersion. The enhancement of MNPs clustering and viscosity inside cells or at the extracellular environment has been pointed as the causes of these effects.

Here, we report on a systematic study of the dynamical magnetic response of MNPs with different sizes. The magnetic experiments were performed by measuring AC susceptibility and magnetometry of MNPs inside biological matrix (biological fluids, live cells) and different aggregation and viscosity conditions. Our findings provide new guidelines in order to design suitable MNPs nanostructures for magnetic hyperthermia applications. Additionally, our methodology for a right characterization of MNPs inside biological matrices and fluids avoids inaccuracies and provide magnetic data for a quantitative modelling of the dynamical magnetic response of MNPs located in biological environments. We observe that the dynamical magnetic response of MNPs of small size is less sensitive to viscosity and aggregation, resulting in the preservation of their magnetic heating with respect to aqueous dispersion. However, larger sizes show higher heating losses but highly sensitive to viscosity and aggregation. Our experimental evidences point out that MNPs characterized by Néel magnetic relaxation and non-influenced by dipolar magnetic interactions are optimal candidates for acting as hyperthermia mediators with invariable heat release into biological environments.

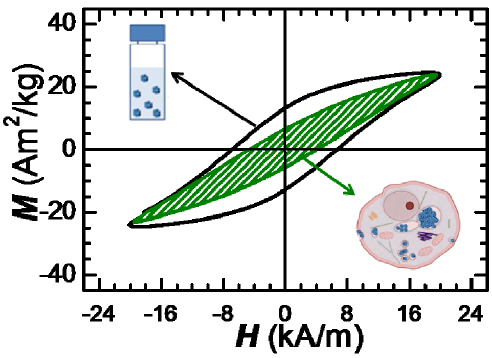

Figure 1. AC hysteresis loops of Iron Oxide Nanoparticles of 21 nm core size in colloidal dispersion (empty line) and inside MCF-7 cells (patterned line) at f = 100 kHz.

STRESS RELAXATION AND NANOCRYSTALLIZATION INFLUENCE ON HIGH FREQUENCY GIANT MAGNETOIMPEDANCE EFFECT OF MAGNETIC MICROWIRES

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ABSTRACT

Due to the extraordinary magnetic properties lend by the absence of long-range order, the amorphous magnetic microwires are interesting for the investigation and development of devices and sensors. That is the reason why an interest in changing and controlling the magnetic properties by thermal treatment at different temperatures is arising, which allows us to adapt them to our requirements. In this work, we have studied the changes in magnetic properties brought by the induced nanocrystallization by thermal treatment in microwires with composition $Fe_{2.25}Co_{72.75}Si_{10}B_{15}$, getting as a result appreciable changes in the high frequency reflectivity of magnetic microwires, with application in the development of electromagnetic absorbers.

MULTISEGMENTED NANOWIRES: A STEP TOWARDS THE CONTROL OF THE DOMAIN WALL CONFIGURATION

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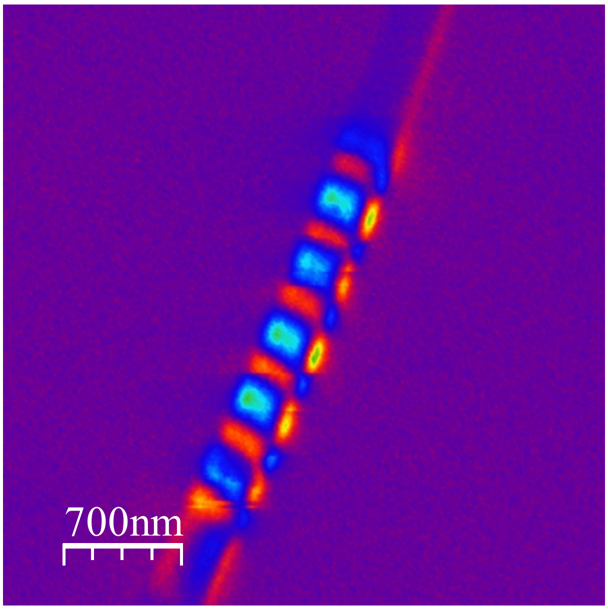
ABSTRACT

Cylindrical nanowires synthesized by controlled electrodeposition constitute excellent strategic candidates to engineer magnetic domain configurations. In this work, multisegmented CoNi/Ni nanowires are synthesized for tailoring a periodic magnetic structure determined by the balance between magnetocrystalline and magnetostatic energies. High-resolution Transmission Electron Microscopy confirms the segmented growth and the sharp transition between layers. Although both CoNi and Ni segments have similar fcc cubic crystal symmetry, their magnetic configuration is quite different as experimentally revealed by Magnetic Force Microscopy (MFM) imaging. While the Ni segments are single domain with axial magnetization direction, the CoNi segments present two main configurations: a single vortex state or a complex multivortex magnetic configuration, which is further interpreted with the help of micromagnetic simulations. This original outcome is ascribed to the tight competition between anisotropies. The almost monocrystalline fcc structure of the CoNi segments, as revealed by the electron diffraction patterns, which is atypical for its composition, contributes to balance the magnetocrystalline and shape anisotropies. The results of MFM measurements performed under in-plane magnetic field demonstrate that it is possible to switch from the multivortex configuration to a single vortex configuration with low magnetic fields.



DEVELOPTMENT OF GMI MONITORING SYSTEM BASED ON MAGNETIC AMORPHOUS MATERIALS

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ABSTRACT

Sensors and monitoring systems are more and more widely used in industry and in general in all human activities. Among then, magnetic sensors play a relevant role, being used in many different applications, such us displacement sensors, magnetic recording, non-contact position sensors, biomagnetism, non-destructive testing [1], [2]. Among magnetic sensors, those based on the Giant Magneto-Impedance (GMI) effect have attracted much attention because they exhibit an ultra-high sensitivity and a high versatility (possibility of automatization and monitoring) at a low cost enabling them for a wide range of applications [3]–[5]. In this work, the development of several GMI based sensors for different technological field of interest is addressed:

A) Cross section of cylindrical pieces.

The determination of cross section variations in cylindrical pieces is relevant for example in biomedicine, agronomics and in tubes and pipes with changing internal pressure. The used devices do not allow cheap automatization and monitoring processes (micrometers or calipers) or show lower resolution (strain gauges). For that reason, we propose the measurement of the relative variations of diameters from a double point of view:

- Magnetoelastic sensors: Melt spun ribbons (66 µm thickness and 530 µm width) with nominal composition $(Co_{0,93}Fe_{0,07})_{75}Si_{12,5}B_{12}$, were employed as magnetoelastic sensor nucleus, by gluing them on the cylindrical surfaces. Relative diameter variations will induce stresses causing measurable changes in impedance, Z. A practical use in in agronomics will be shown [6].

- Magnetoinductive sensors: In this case a wire ($Co_{66}Fe_2Si_{13}B_{15}Cr_4$, diameter 90 µm) was proposed as a non contact sensor for the measurement of the position in

the micrometric range of mobile pieces with a joined magnet. As the piece moves, the magnetic field in the sensor changes, leading to variations in Z. Their performance in a particular agronomical application was also checked.

B) Magnetic nanoparticles detection.

Biosensors based on Giant magnetoimpedance (GMI) effect have been proposed as an alternative procedure for the detection of magnetic nanoparticles. The interaction between stray fields of the deposited nanoparticles and the GMI sensor produces measurable changes in the Z modulus but with low detection rates 5-15%. Nevertheless, the sensitivity can be largely enhanced by the analysis of the magneto-reactance, that is, the imaginary component, X, of the electrical impedance, Z, in the lower frequency range (f = 100 kHz) [7,8]. In turn, non-linear magnetoimpedance, displays a very high sensitivity to both stresses and magnetic field, therefore the design of a GMI nanoparticle sensor based on non- linear terms is shown in this work. Melt spun ribbons $(Co_{0,94}Fe_{0.06})_{72.5}Si_{12.5}B_{15}$, displaying a meander structure, show variations in Z before and after adding magnetic nanoparticles. Both, the first and the second armonics were analyzed [9].

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Magnetization Reversal Processes in Homogeneous geometry and Diameter Modulated Ni and Co Nanowires. A micromagnetic simulation approach.

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ABSTRACT

The usage of magnetic nanowires (NWs) for ultra-high density magnetic data storage applications has gained increased attention in the last years due to the growing need for the miniaturization of devices[1]. In arrays of high aspect ratio ferromagnetic NWs, the information is stored along the wire, as a sequence of magnetic domains (bits of information), where magnetization pointing upward or downward represent a different state of a magnetic bit of information. The precise control of the NWs geometries and/or compositions[1,2], allows for the tuning of the magnetization reversal mode by external parameters, such as the direction of the applied magnetic field or the spin-polarized current, and the pinning of magnetic domain walls (DWs)[3]. This fact is of extreme importance for the implementation of these magnetic memories in new devices.
For a better understanding of the magnetization reversal processes in NWs, we propose in this work to study the influence of geometrically modulated single isolated Ni and Co NWs, with a diameter-modulated ratio 1:2, by micromagnetic simulations using the open source software called Mumax3[4]. The narrow and wide diameter segments of the NWs are 40 nm and 80 nm, respectively, and the total NW length is 4 µm. The micromagnetic simulations were done for different length segments that are ranging from 2 to 3.6 µm for the narrow segment and 0.4 to 2 µm for the widest one segment. For comparison purposes, we have also simulated non-modulated magnetic NWs having uniform diameter with the above-mentioned diameters lengths to highlight the differences between non-modulated and modulated NWs.

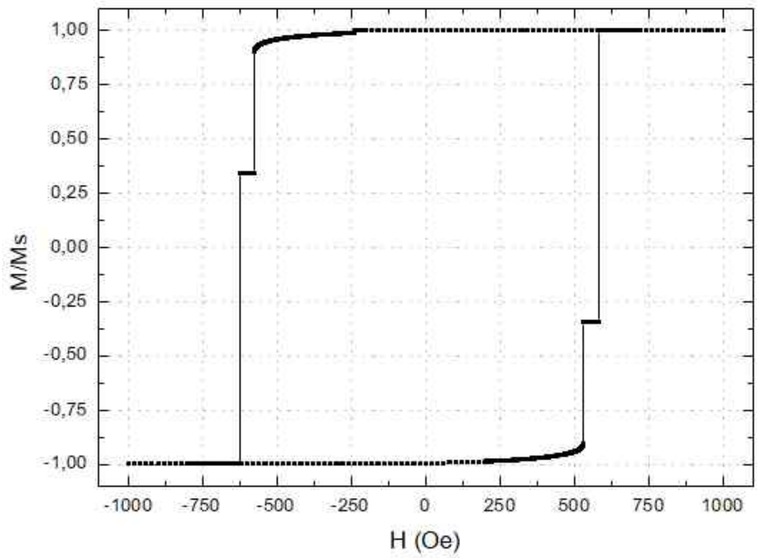

Fig. 1 Selected hysteresis loop simulated for a modulated Ni NW with the following geometry: 40 nm of diameter and 3.6 um length for the narrow segment, and 80 nm of diameter and 0.4 um length for the wide segment.

The simulated hysteresis loops (Fig. 1), shows a rich variety of magnetization reversal processes that ranges from magnetization rotation for almost any NW length to several kinds of domain wall displacements that depend on the NW diameter, length segments and magnetic material. For example, vortex domain walls can be nucleated at the edge of the wider segment and propagated to the interface, while the magnetization in the shorter segment remains unmodified, producing a fast two-step magnetization reversal.
Magnetization reversal processes have been studied in Ni and Co NWs with different length segments for those modulated in diameter by micromagnetic simulations, which can give a better understanding of the fundamental physics behind the magnetic behavior observed for this kind of nanostructures. We expect in the near future to extend this work to studies related with the propagation velocity of the domain walls under the action of either an applied magnetic field or a spin polarized current.

INFLUENCE OF CALORIMETRIC LOW TEMPERATURE LIMIT ON MAGNETOCALORIC MEASUREMENTS

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ABSTRACT

Nowadays, the Magnetocaloric Effect (MCE) has gained a growing interest due to its application in solid state magnetic refrigerators at room temperature. This technology has been shown as an energy efficient and environmental friendly alternative to the conventional systems. A crucial step is an accurate characterization of the magnetocaloric magnitudes (magnetic entropy change, ΔS_M, and adiabatic temperature change, ΔT_{ad}). Using heat capacity measurements (C), the total entropy is calculated as:

$$S_H(T) = S_{0,H} + \int_{0\text{ K}}^{T} \frac{C_H(T)}{T} \mathrm{d}T, \quad (1)$$

where $S_{0,H}$ is the zero entropy term, the magnetocaloric magnitudes are obtained as $\Delta S_M = [S_{H_F} - S_{H_I}]_T$ and $\Delta T_{ad} = [T_{H_F} - T_{H_I}]_S$, and H_I and H_F are the initial and final magnetic fields, respectively. To obtain these data, it is necessary to calculate the zero entropy term and to experimentally reach temperatures close to 0 K. Regarding the first condition, $S_{0,H}$ is usually neglected because its effect is assumed small and field independent. With respect to the second condition, it is difficult to achieve such a broad temperature span for materials with a transition close to room temperature due to experimental limitations. In that case, some assumptions have to be considered for the missing data range. Assuming a linear behavior of C/T down to zero kelvin (with $C/T(0\text{ K}) = 0$), total entropy (under the used approximation) can be obtained as [1]:

$$S_H^{ap}(T) = \frac{1}{2} C_H(T_{ini}) + \int_{T_{ini}}^{T} \frac{C_H(T)}{T} \mathrm{d}T, \quad (2)$$

where T_{ini} is the lower measured temperature. The influence of this approximation on the determination of the magnetocaloric effect has been analyzed by using numerical simulations and experimental measurements. The Brillouin equation of state has been used to describe the magnetic behavior of materials exhibiting a second order phase transition. Simulations show that the approximated magnetocaloric curves deviate with respect to those curves integrated from zero kelvin in a non-monotonic way. However there is a certain temperature (T_{ini}) for which both resulting curves from (1) and (2) coincide (the initial temperature that reduces the deviations is the same for ΔS_M and ΔT_{ad} curves). Hence a procedure is proposed to obtain the experimental magnitudes of ΔS_M and ΔT_{ad} with a minimum error using heat capacity data in a limited temperature span. It has been successfully applied to a GdZn alloy and its results are comparable to those derived from magnetization measurements [2].

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NON-STANDARD MFM IMAGING

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Despite decades of advances in magnetic imaging, obtaining direct, quantitative information with high spatial resolution remains an outstanding challenge. The imaging technique most widely used for local characterization of magnetic nanostructures is the Magnetic Force Microscope (MFM), which is indeed a very active topic of investigation [1]. Advantages of MFM include relatively high spatial resolution, simplicity in operation as well as sample preparation, and the capability to applied in situ magnetic fields to study magnetization process [2,3]. Recently we have also demonstrate the possibility of operate in different environments including liquid media that allow us to investigate biological samples [4]. In the present work we try to approach some of the challenges of MFM, spatial resolution, sensitivity and quantitative measurements, by following different routes. One route is the development of high-performance MFM probes with sub-10 nm (sub-25 nm) topographic (magnetic) lateral resolution by following different easy and quick low-cost approaches. This allows one to not only customize the tip stray field, avoiding tip-induced changes in the sample magnetization, but also to optimize MFM imaging in vacuum (or liquid media) by choosing tips mounted on hard (or soft) cantilevers, a technology that is currently not available on the market [5].
Furthermore, the idea of explore new MFM probe architectures [6] allow us to focus some of the challenges of the technique as the lack of quantitative information. In that sense, alternative advanced methods as measuring energy dissipation with MFM (see Figure 1) is of great interest not only for nanomechanics but also to understand important energy transformation and loss mechanisms that determine the efficiency of energy of data storage device [7].

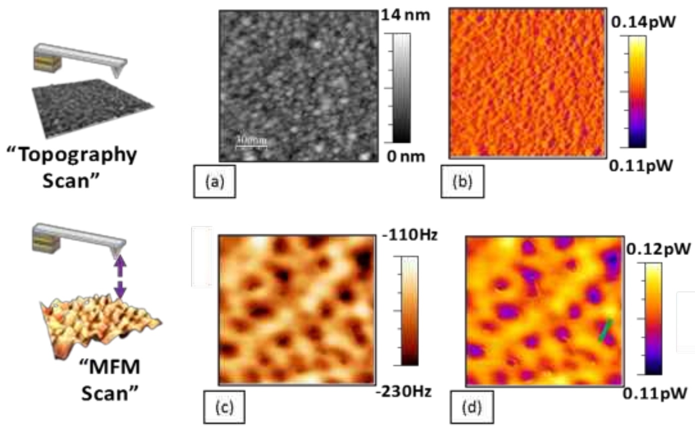

Figure 1. Dissipation maps obtained by MFM. (b) has been measured simultaneously to the topography (a) and (d) is recorded simultaneously to the magnetic domains (c)

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MAGNETIC NANOWIRES WITH CHEMICAL NOTCHES FOR SPINTRONICS APPLICATIONS

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ABSTRACT

Future magnetic storage technology might rely on the movement of magnetic domain walls using spin polarized currents[1]. When not in motion, the magnetic domain walls which define the bits should be pinned in artificially created notches along magnetic wires[2]. It has been shown recently that the depinning of a domain wall from these notches has an intrinsic stochastic component which complicates the application of the proposed scheme in real devices[3]. New types of notches are needed to overcome those limits. In this work we have introduced local changes in composition along permalloy nanowires. These chemical boundaries may act as local pinning sites (chemical notches).

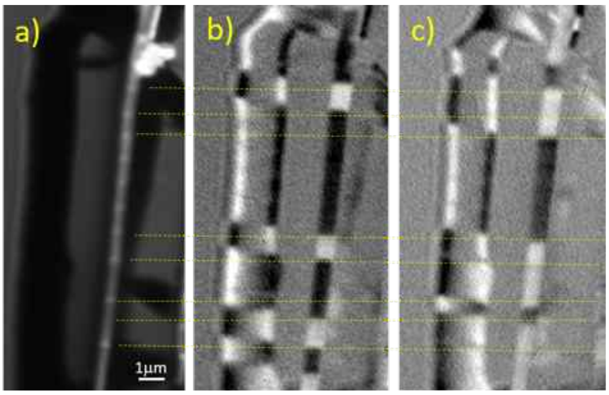

Image 1. (a) Image with chemical contrast (b) XMCD image at Fe K-edge. (c) XMCD image at Fe k- edge after apply magnetic field.

To study the behaviour of magnetic domain walls, it is mandatory to use a technique that combines magnetic and chemical information at submicrometer spatial resolution. The LEEM-PEEM microscope in the CIRCE beamline at ALBA synchrotron fulfils all these requirements. The LEEM microscope has enough resolution to search for the individual nanowires in the substrate. We have used XAS-PEEM to obtain images with chemical contrast (see Fig. 1a) and XMCD-PEEM to get magnetic contrast (Fig 1b). From these figures we correlate composition and magnetic properties of the nanowires. We have proven that chemical notches act as pinning sites for the domain walls. After applying a magnetic field, the domain walls move and pin to a different notch (see Fig 1c). These results open the possibility of using these chemically modulated nanowires as storage elements in race-track memories.

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Systematic study of Fe3O4 and γ-Fe2O3 nanoparticles for hyperthermia

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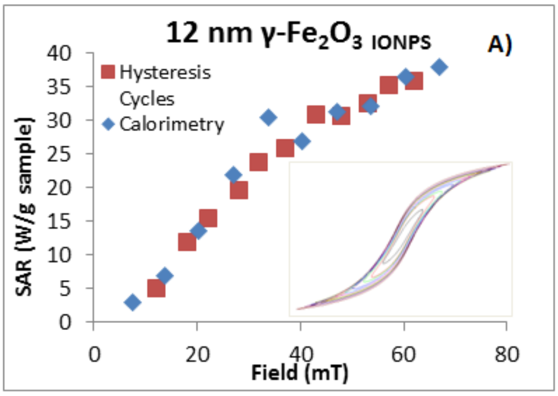
ABSTRACT

In order to improve the heating performance of magnetic nanoparticles during hyperthermia treatments, a systematic study of different Fe_3O_4 and γ-Fe_2O_3 nanoparticles has been done by means of a hand-made high frequency hysteresis loop meter and a calorimeter. This study goes deep into the knowledge of the behavior and properties of magnetic nanoparticles subjected to high frequency alternating fields and the effect of nanoparticle properties like size, saturation magnetization, magnetic anisotropy, etc. on the heating efficiency.

Iron oxide nanoparticles suspended in water with sizes between 6 and 300 nm with different coatings and synthesis routes have been analized. All the colloids were previously characterized by TEM, XRD, DLS and SQUID magnetometry. For the calorimetric and magnetometric measurements, fields between 10-60 mT have been used, and a frequency of 100 kHz and 50 kHz respectively. We have seen that 35 nm Fe_3O_4 nanoparticles reach the highest heating efficiency of all samples, having a size close to the monodomain-multidomain limit. Also, no differences on the heating efficiency were found between uncoated and APS (aminopropyltriethoxisilane) coated 12 and 14 nm γ-Fe_2O_3 nanoparticles, even at high fields. On the other hand, 14 nm uncoated γ-Fe_2O_3 nanoparticles have a 35% higher heating efficiency than the 12 nm nanoparticles. The results obtained for the calorimetric and magnetometric measurements are in good agreement.

The main goal of this work is to be able to measure the heating efficiency of the nanoparticles at the same time as obtaining information regarding the dynamic magnetic properties of the sample, which will help to achieve an optimized hyperthermia treatment and also to understand the physics that underlies the heating mechanisms.



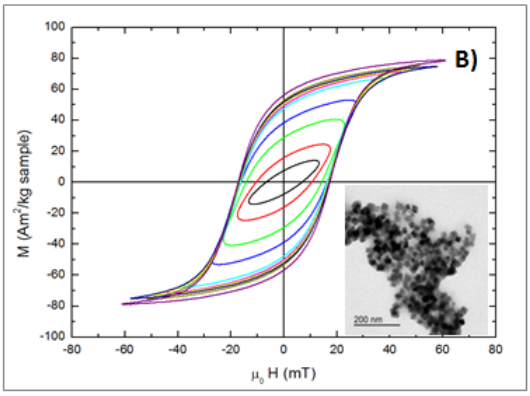


Figure: A) SAR as a function of the applied field obtained for the 12 nm γ-Fe_2O_3 sample; magnetometric and calorimetric results. *Inset:* shape of the high frequency hysteresis cycles measured from 10 – 60 mT and 50 kHz. B) High frequency hysteresis cycles measured from 10 – 60 mT and 50 kHz for the 35 nm Fe_3O_4 colloid. Inset: TEM image of the sample.

EFFECT OF THE ORTHORHOMBIC PHASE ON THE STRUCTURAL AND PHYSICAL PROPERTIES OF MULTIFERROIC $YMnO_3$

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ABSTRACT

Multiferroic materials have a great technological interest due to the possibility of electrical field control of a magnetic state. This is possible through the coupling between ferroelectricity and magnetism of these materials. The coupling can be due to different effects: strain or piezoelectric coupling, direct spin exchange, or by charge transfer. Several aspects of this field are still unclear and much experimental and theoretical work remains to be performed.

In this work, we present a physical characterization of the multiferroic $YMnO_3$. This compound can be in two possible phases: a stable hexagonal phase and a metastable orthorhombic phase. Herein we discuss the effect of the metastable orthorhombic phase on the physical properties of $YMnO_3$. For this purpose, a set of samples with different structure was obtained. The starting parent compound $YMnO_3$ was synthetized via solid-state reaction. A family of samples was obtained by means of mechanical milling combined with subsequent heat treatments. By these processes, we are able to tune the structure and the metastable phase. Structural, magnetic and transport properties of these samples are analyzed. [1] [2]

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2. Kristin Bergum et al., *Dalton Trans* **40**, 7583-7589 (2011)

FERROMAGNETIC MICROWIRE TO BE USED IN TUNABLE REFLECTARRAYS/TRANSMITARRAYS ANTENNAS BY MEANS OF MAGNETIC FIELD

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ABSTRACT

In this work, we report a research work related to the interaction between microwaves and finite length FeCoSiB ferromagnetic microwires as tunable reflectarrays/transmitarrays antennas. The scattering coefficient of Cu and amorphous ferromagnetic microwires without magnetic field has been measured for frequency range between 0.5 to 10 GHz. The electric current induced along the microwire by the electromagnetic wave produce the emission of other electromagnetic wave. The interference between waves is measured and compared with different distances of broadcasting antenna. The electric current depends on the microwire impedance as well as on the relationship between its length and the wavelength of the microwave. In the case of ferromagnetic microwire the impedance is tuned with a bias magnetic field. The tune of the impedance let us change the operating frequency of the array.

THE RELIABILITY OF POWER LAWS AND UNIVERSAL SCALING OF THE MAGNETOCALORIC EFFECT: RANGE OF APPLICABILITY AND UNIVERSALITY CLASSES

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ABSTRACT

The magnetocaloric effect (MCE) has attracted a lot of attention among the scientific and engineering communities due to its promising applications in near room temperature refrigeration and other energy conversion matters. With the aim of comparing the performance of different materials regardless their nature, processing or experimental conditions during measurements, the use of power laws or universal scaling was proposed [1]. However, these approaches are based on critical phenomena and are restricted to a certain region near a phase transition. Therefore, it is necessary to quantify the limits in which these approaches remain applicable for MCE research purposes and if they are valid for all universality classes.
The aim of this work is twofold. On the one hand, by means of two independent models, as well as experimental data, we determine the range of applicability of these analysis methods, in terms of temperature and applied field. In practical terms, our study confirms the proper applicability of the mentioned procedures in materials whose Curie temperature is close to room temperature until applied fields as large as 10 T, covering the usual field range available at magnetism laboratories and used in most applications [2]. On the other hand, we analyze in detail the behavior of the critical scaling not only under the frame of a second-order phase transition in a mean-field model but also in the environment of a tricritical point, comparing with a broad data set from the literature. Finally, we confirm the feasibility of these approaches also in this distinct regime with different critical exponents [3].

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