Experimental determination of the frequency and field dependence of Specific Loss Power in Magnetic Fluid Hyperthermia

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INTRODUCTION

We present an experimental study of the **Specific Loss Power** (**S**LP) of three maghemite-based ferrofluid samples with different core diameter, as a function of frequency and intensity of the applied alternating magnetic field H. The results allowed us to highlight the size-dependence of the physical mechanism responsible for the heating and to establish the phenomenological functional relationship SLP = c·H^x with 2 ≤ x < 3, the x-value depending on sample size and field frequency, here chosen in the typical range of operating magnetic hyperthermia devices.

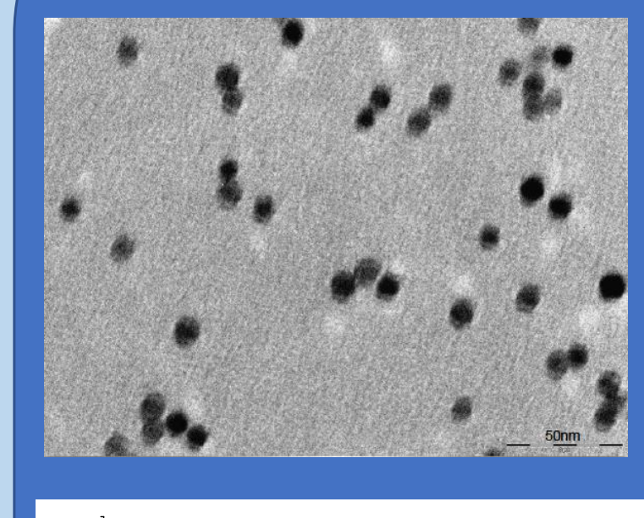
SAMPLES

We studied three novel maghemite (γ-Fe$_2$O$_3$) nanoparticles samples with different magnetic core diameters coated with PolyAcrylic Acid (PAA), in water.
The compositional, structural and magnetic properties of the samples were investigated with

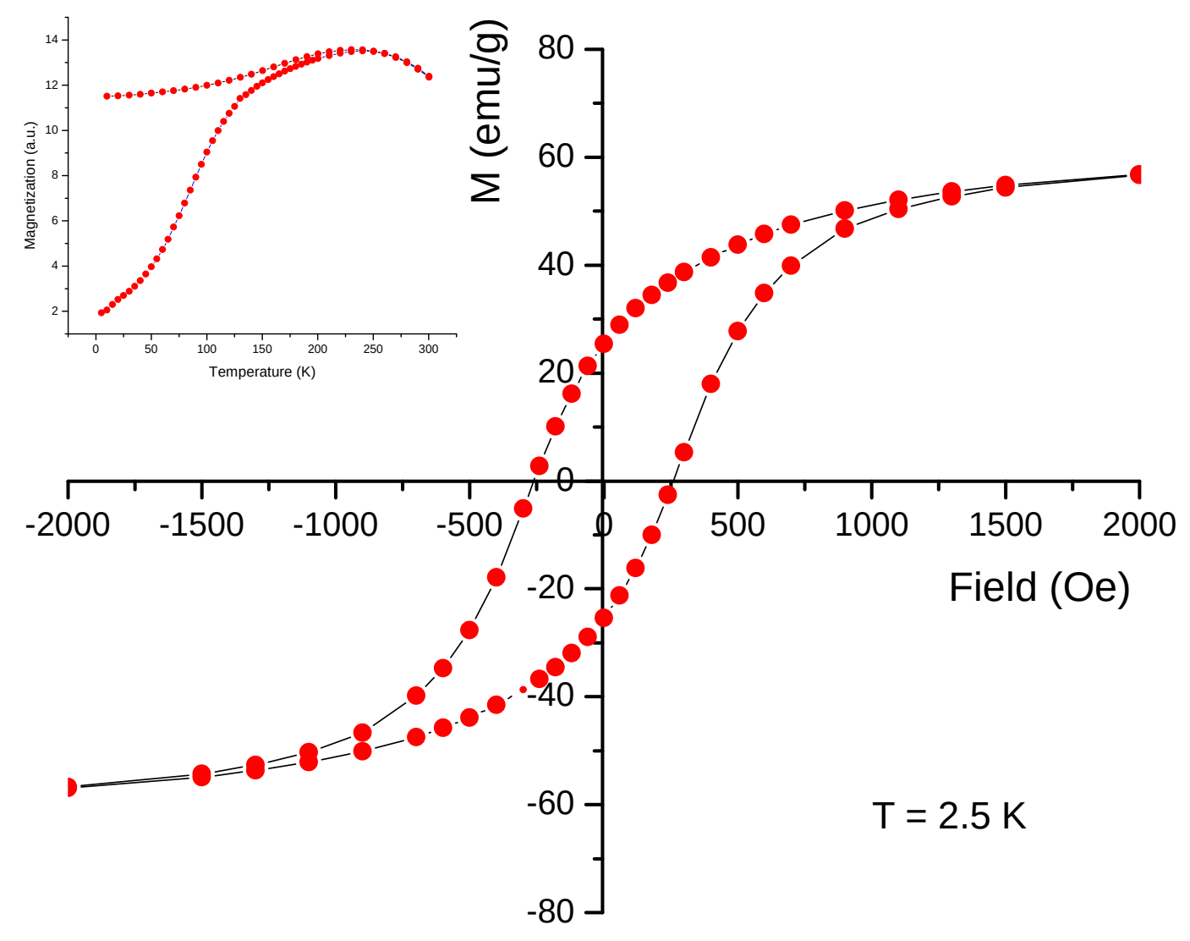
- TEM
- XRD
- AFM
- SQUID (ZFC-FC at 5 mT, M(H) at -5 ÷ 5 T and T = 2.5 K/300 K)

Sample	d$_{TEM}$ (nm)	d$_{AFM}$ (nm)	M$_s$ at 2.5K (emu/g)	H$_c$ (Oe)	M$_s$ at 300K (emu/g)
A	10.2 ± 1.1	11.4 ± 0.9	62.4 ± 3.4	265 ± 13	54.6 ± 3.0
B	14.6 ± 1.8	15.6 ± 0.8	67.2 ± 3.7	239 ± 15	58.3 ± 3.2
C	19.7 ± 1.7	20.5 ± 0.8	69.3 ± 3.8	360 ± 12	60.9 ± 3.3

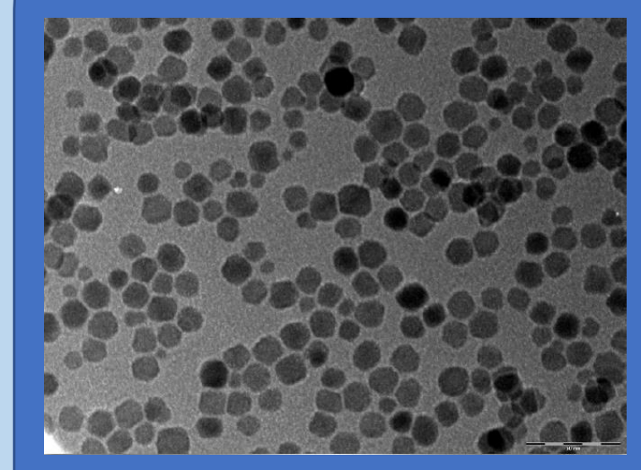
Sample A



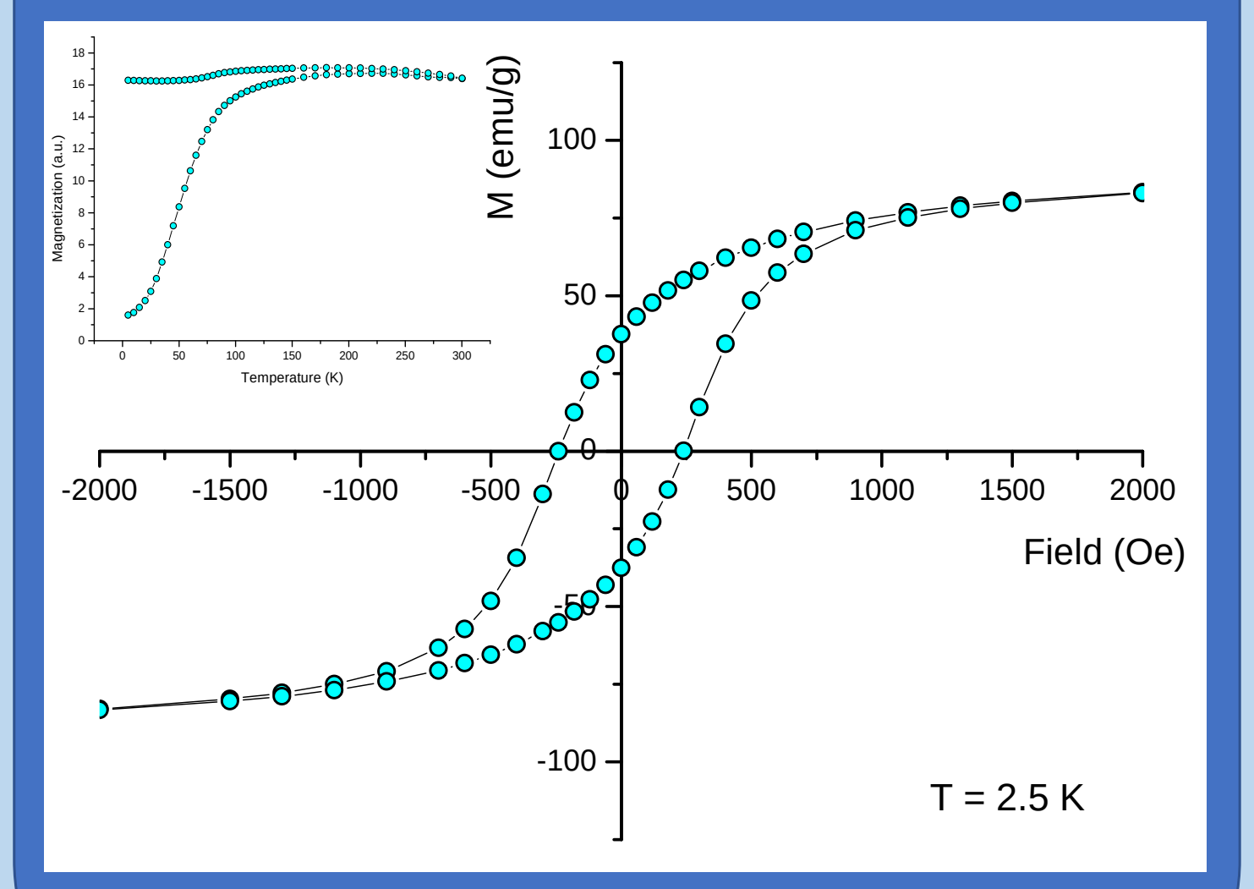
d = 10.2 nm
σ_d = 1.1 nm



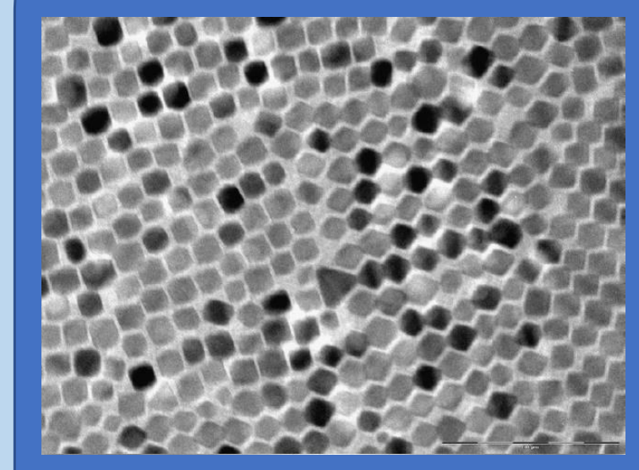
Sample B



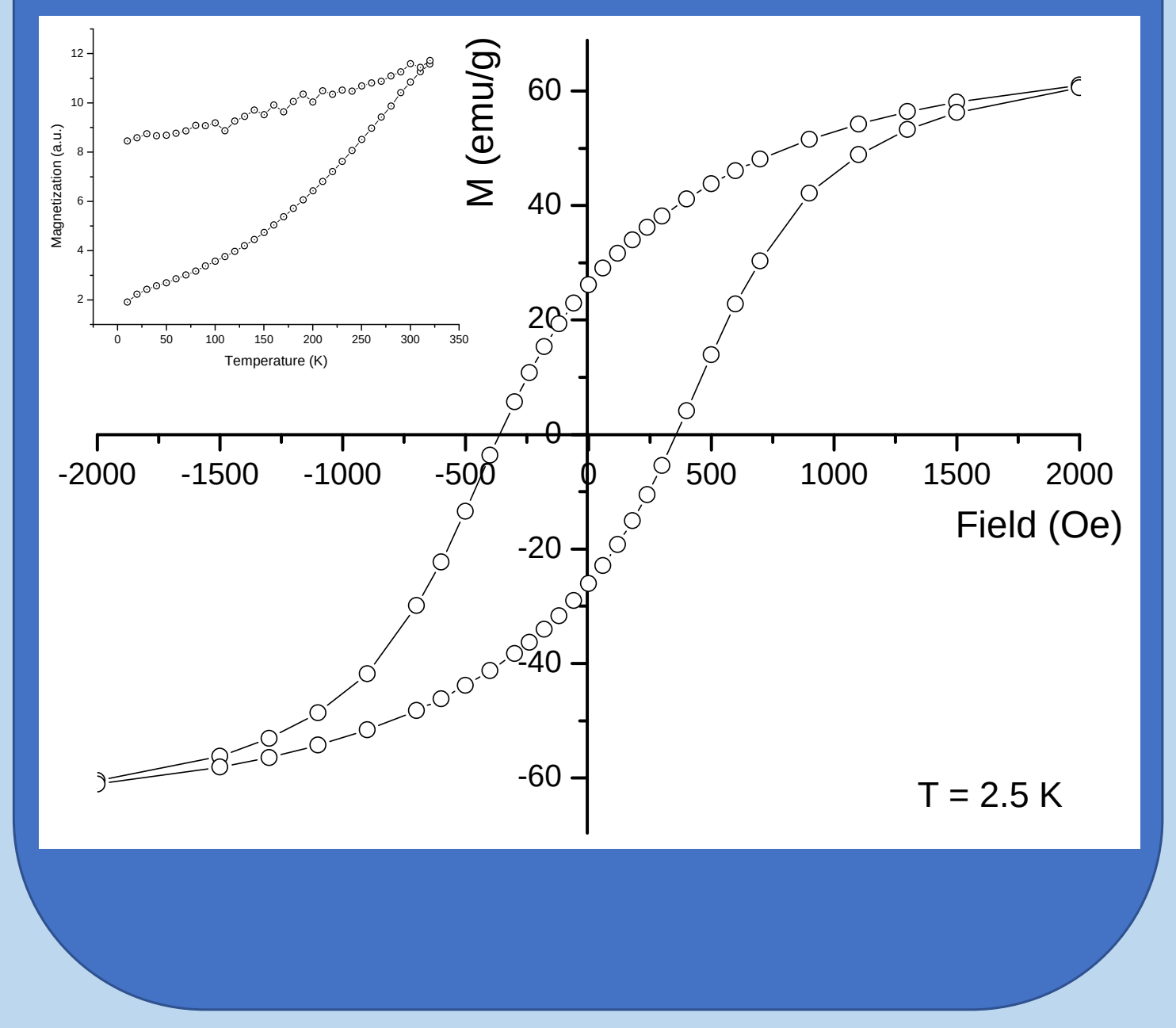
d = 14.6 nm
σ_d = 1.8 nm



Sample C

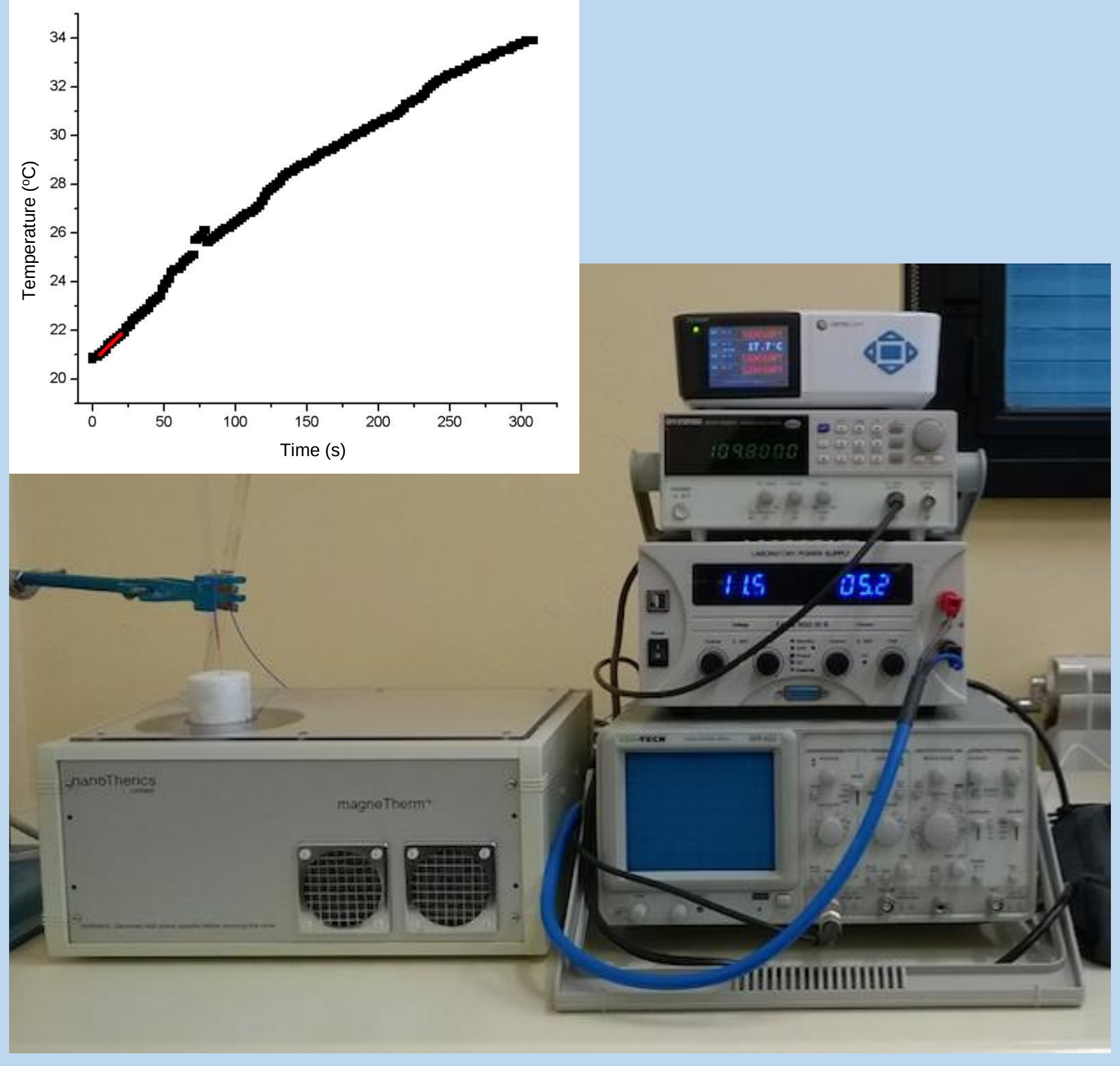


d = 19.7 nm
σ_d = 1.7 nm



EXPERIMENTAL MEASUREMENTS

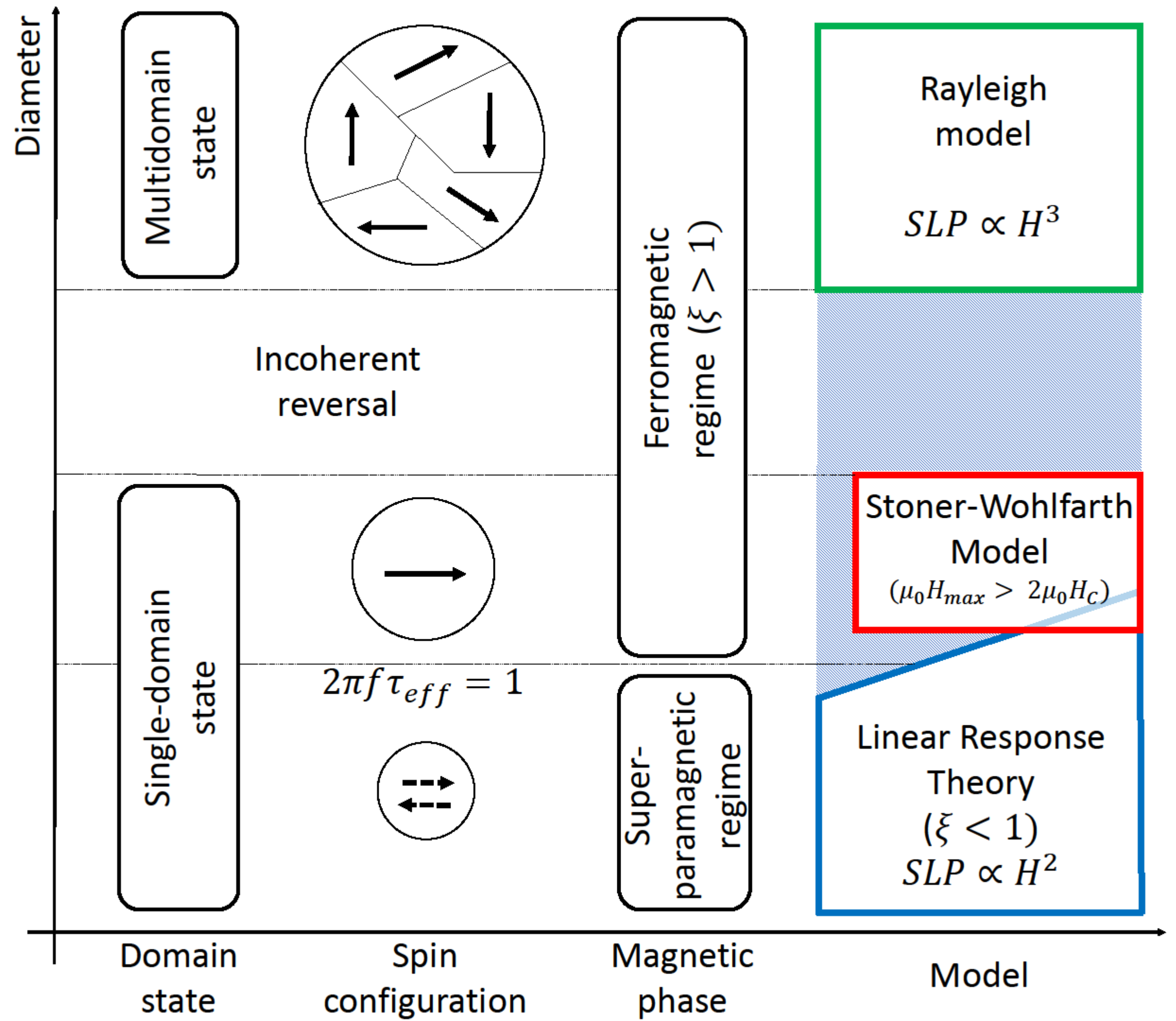
Magnetic heating experiments were performed on 1 mL of stable aqueous solutions at room temperature by a Magnetherm (nanoTherics^{TM}) set-up, varying the **frequency** f (**100 kHz ÷ 1 MHz**) and the **amplitude** μ_0H (**3 ÷ 17 kA/m**). The temperature was measured using an optical fiber thermometer (Opticon^{TM}) and the temperature vs. time curve, T(t), was acquired (5÷15 minutes).



To estimate the temperature increment rate *ΔT/Δt*, we used the initial slope method or the Box-Lucas model [1] fitting the curve T(t) = $\alpha(1 - e^{-\beta t})$. The SLP values were evaluated using the following equation:

$$SLP = \frac{m_{H_2O}c_{H_2O} + m_{\gamma-Fe_2O_3}c_{\gamma-Fe_2O_3}}{m_{\gamma-Fe_2O_3}}\frac{\Delta T}{\Delta t}$$

where c$_{H2O}$ and c$_{\gamma\text{-Fe2O3}}$, m$_{H2O}$ and m$_{\gamma\text{-Fe2O3}}$ are the specific heat and the mass of the main constituents of the solution [2].



Diameter
Multidomain state
Incoherent reversal
Single-domain state
$2\pi f\tau_{eff} = 1$
Ferromagnetic regime ($\xi > 1$)
Super-paramagnetic regime
Rayleigh model
$SLP \propto H^3$
Stoner-Wohlfarth Model
($\mu_0 H_{max} > 2\mu_0 H_C$)
Linear Response Theory
($\xi < 1$)
$SLP \propto H^2$
Domain state
Spin configuration
Magnetic phase
Model

MODELS FOR SLP

From the **Linear Response Theory (LRT)**, when the magneto-thermal quantity ξ<1, where $\xi = \mu_0 M_s V H_{max}/k_B T$ and H$_{max}$ is the maximum applied field [3]:

$$SLP = \frac{\mu_0 \pi \chi''(f) f H_{max}^2}{\rho}$$

χ'' is the out-of-phase component of the magnetic susceptibility:

$$\chi''(f) = \frac{\mu_0 M_s^2 V}{3k_B T}\frac{2\pi f \tau_{eff}}{\left[1+(2\pi f \tau_{eff})^2\right]}$$

and τ_{eff} is the effective relaxation time $1/\tau_{eff} = 1/\tau_N + 1/\tau_B$ being τ_N and τ_B the Néel and Brown relaxation times.

From the **Rayleigh model**, when $\xi > 1$: $SLP \propto H^3$

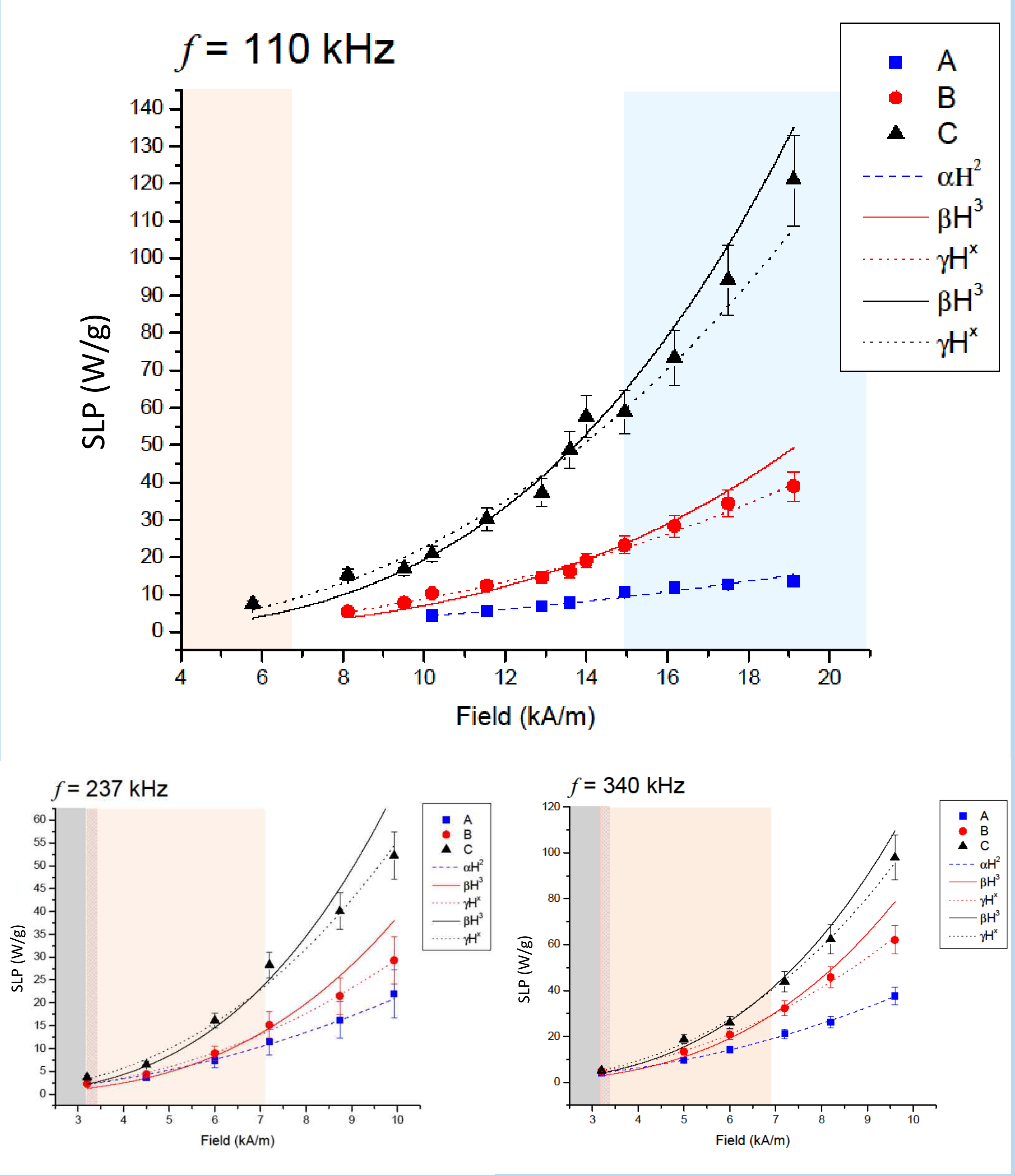
Stoner-Wohlfarth model is valid if the condition μ_0H$_{max}$ > 2μ_0H$_c$ is satisfied.

RESULTS

The SLP of sample C is the highest at all measured frequencies and amplitudes, while samples A and B show lower SLP values with slight differences.
In order to evaluate the best theoretical model to describe the data we calculated for each sample the maximum value of the field H that satisfies the condition $\xi < 1$ for the applicability of the LRT model (coloured zones), showing that this model applies only for sample A. These observations have been confirmed by fitting the SLP data to a power law with a free exponent, SLP ∝ H^x.

x values from the fit with SLP ∝ H^x are reported in the table.

Sample	x at 110 kHz	x at 237 kHz	x at 340 kHz
A	2.0 ± 0.1	2.03 ± 0.01	2.09 ± 0.09
B	2.23 ± 0.08	2.30 ± 0.02	2.34 ± 0.03
C	2.3 ± 0.1	2.47 ± 0.09	2.64 ± 0.06



CONCLUSIONS

The LRT model explains the data for small MNPs (sample A) but fails when the nanoparticles size becomes larger than the critical diameter corresponding to the onset of the transition from the Superparamagnetic to the Ferromagnetic regime: for samples B and C the SLP follows the power law SLP = c·H^x with 2 < x < 3, where x increases with increasing frequency. These x values are intermediate between the one predicted by the LRT (x = 2) and the Rayleigh (x = 3) models.

REFERENCES

[1] A. G. E. P. Box, and H. L. Lucas, *Biometrika* 46, 1 (2016); [2] E. A. Périgo, G. Hemery, O. Sandre, D. Ortega, E. Garaio, F. Plazaola, and F. J. Teran, *Appl. Phys. Rev.* 2, 041302 (2015); [3] J. Carrey, B. Mehdaoui, and M. Respaud, *J. Appl. Phys.* 109, 8 (2011)