Electronic Supplementary Material (ESI) for Nanoscale.

Probing T_1-T_2 interactions and their imaging implications through a thermally responsive nanoprobe

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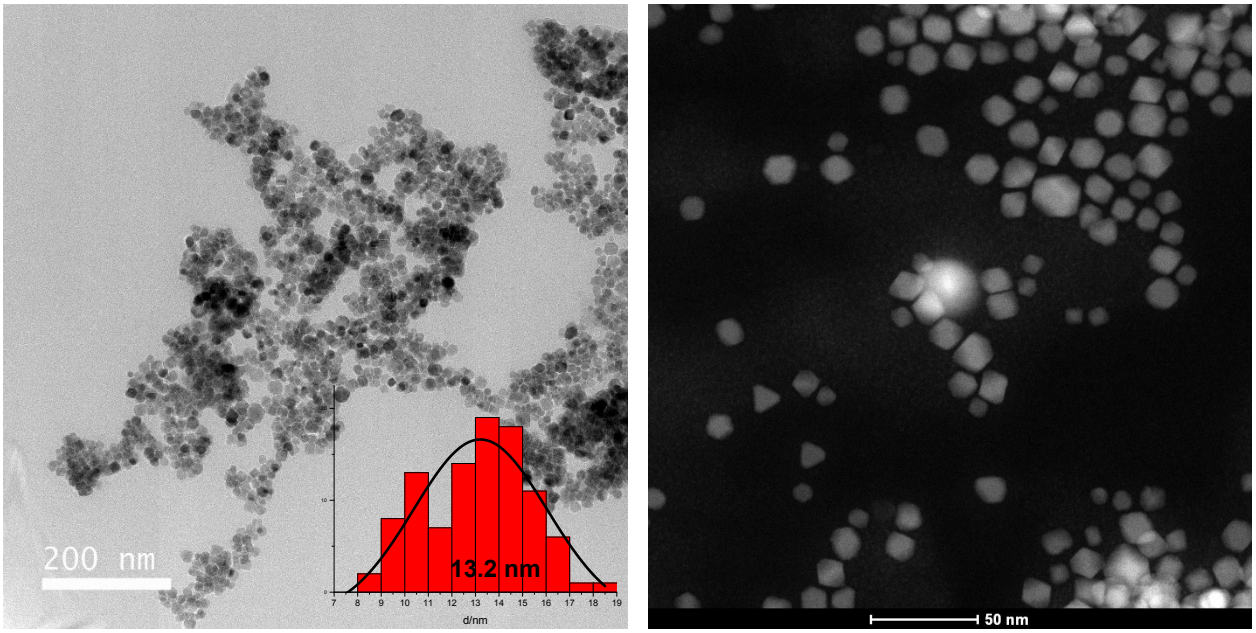


Figure S1. TEM (left) and scanning transmission electron microscopy (STEM) (right) images of $Mn_xFe_{3-x}O_4$ nanoparticles. Inset, size distribution of the nanoparticles

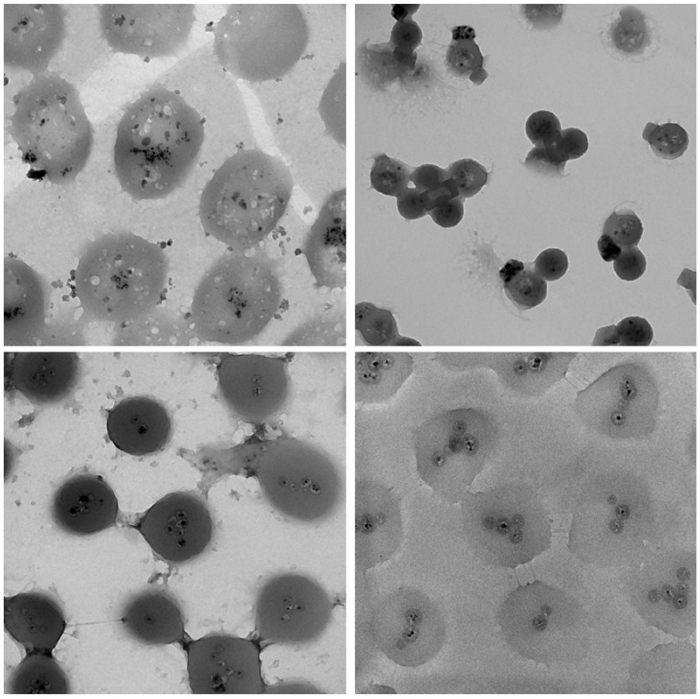
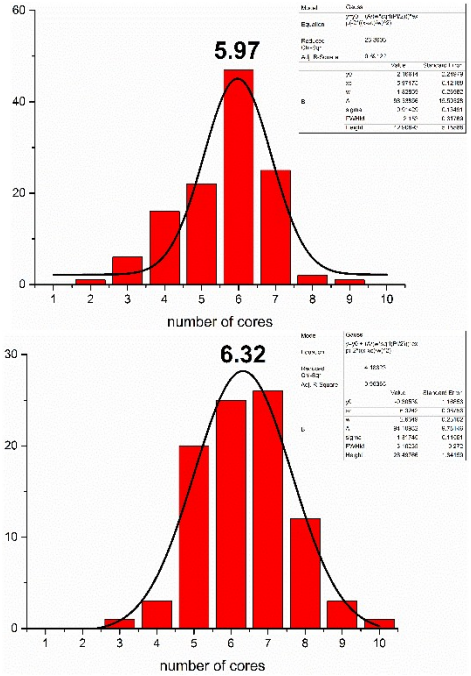


Figure S2. Overview TEM micrographs of the final temperature responsive nanoparticles.

Table 1. Hydrodynamic diameters of the samples measured in water at 20 and 40 ºC.

Sample	D_h (T = 20 ºC)	D_h (T = 40 ºC)
1	39 ± 6	37 ± 8
2	40 ± 9	42 ± 7
3	51 ± 9	49 ± 8
4	65 ± 11	67 ± 9
5	482 ± 8	235 ± 11
6	416 ± 15	246 ± 18
7	502 ± 26	222 ± 7
8	497 ± 10	243 ± 2
9	502 ± 19	250 ± 16
10	456 ± 9	212 ± 13
11	478 ± 7	235 ± 7
12	485 ± 5	225 ±23



Sample	Average number of cores
1	n/a
2	n/a
3	n/a
4	n/a
5	4.56
6	4.35
7	4.89
8	5.97
9	4.48
10	4.67
11	4.77
12	6.32

Figure S3. Left, representative histograms of samples **8** and **12** of the number of magnetic cores per pNIPAM particle. Right, table summarising the average number of magnetic cores per particle for the different samples prepared.

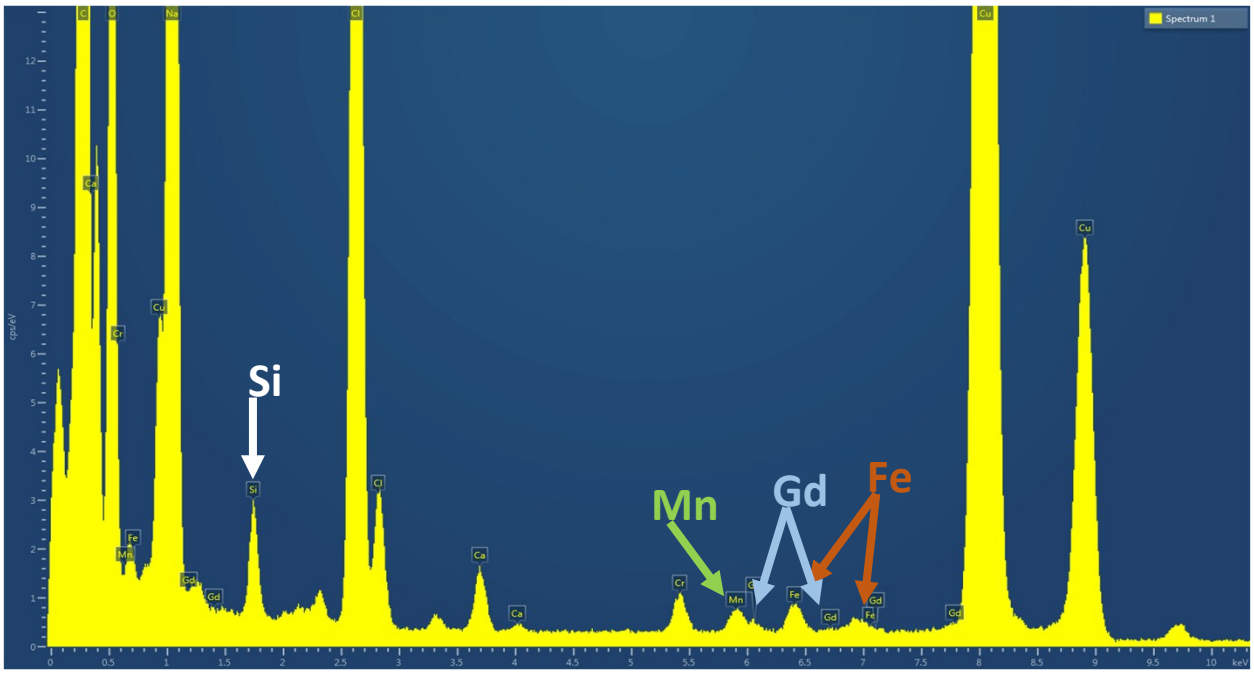


Figure S4. EDX spectra of $Mn_xFe_{3-x}O_4$@SiO_2@pNIPAM-Gd^{3+} nanoparticles showing peaks from Si (white arrow), Mn (green arrow), Gd (blue arrow) and Fe (brown arrow).

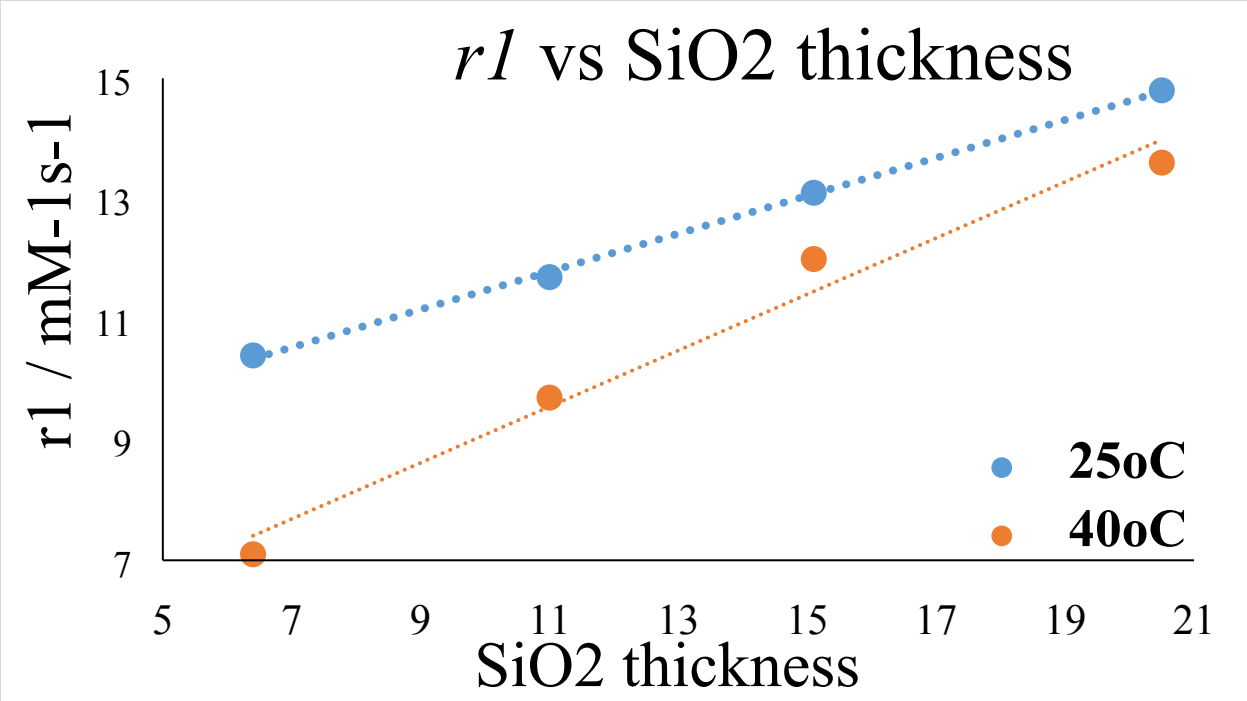


Figure S5. Plot showing the linear relationship between the longitudinal relaxivity of samples **5** to **8** and the thickness of the silica layer, both at 25 and 40 ^{o}C.

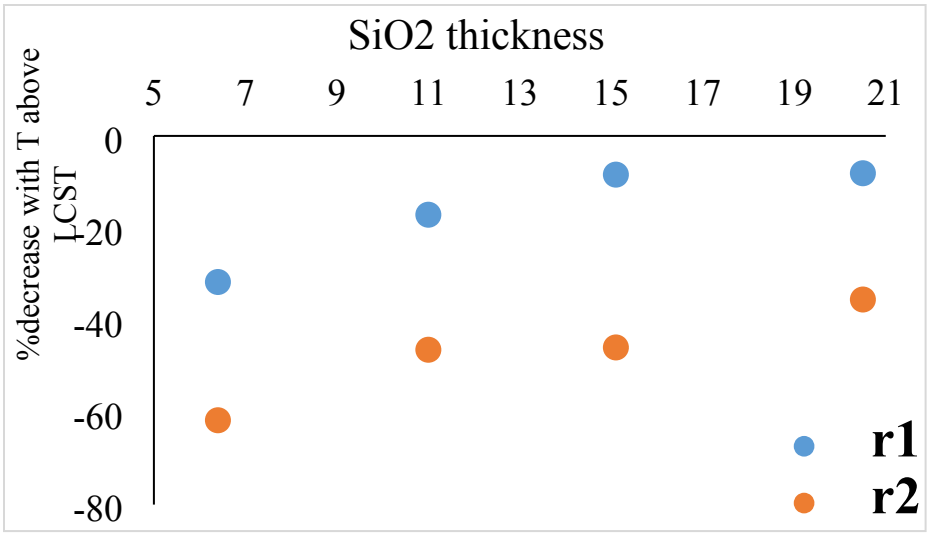


Figure S6. Plot showing the decrease of longitudinal and transverse relaxivity of samples **5** to **8** versus the thickness of the silica layer.

$$R_2 = \frac{16}{45}\nu\tau_D(\gamma B_{eq})^2 \qquad \text{(eq 1)}^{[1]}$$

Equation S1. Transversal relaxation rate in the motion average regime. ν, magnetic volume fraction; τ_D, diffusion time; γ, proton gyromagnetic factor; B_{eq}, nanoparticle equatorial field.

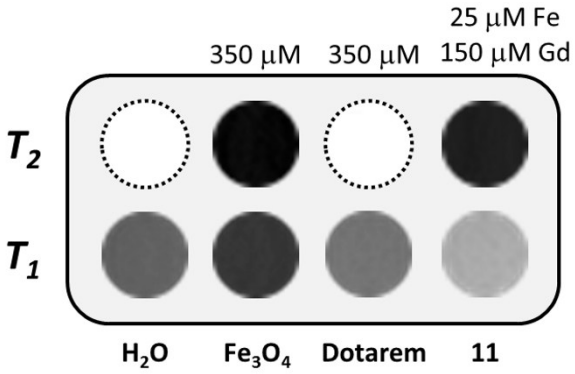


Figure S7. T_2 and T_1-weighted MRI phantoms of sample **11** compared to water, 8 nm Fe_3O_4 nanoparticles and Dotarem. Magnetite nanoparticles at a concentration of 350 μM of Fe, Dotarem at a concentration of 350 μM Gd3+.

[1] M. R. J. Carroll, R. C. Woodward, M. J. House, W. Y. Teoh, R. Amal, T. L. Hanley, T. G. St Pierre, *Nanotechnology* **2010**, *21*, 35103.