1.0- Synthesis of Iron Oxide Nanocubes

Iron Oxide Nanocubes (IONCs) with edge sizes of 14 ± 2 and 24 ± 4 nm were synthesized by thermal decomposition method as described elsewhere. Briefly, to obtain Fe$_3$O$_4$ nanocubes of 14 nm size, 0.353 g (1 mmol) of iron(III) acetylacetonate with 1.04 g (6 mmol) decanoic acid and 15 mL dibenzyl ether were dissolved in 10 mL squalane in a 50 mL three-neck flask. After degassing for 120 min at 65 °C, the mixture was heated up to 200 °C (3 °C/min) and kept at this value for 2.5 h. Finally, the temperature was increased at a heating rate of 7 °C/min up to 310 °C or reflux temperature and maintained at this value for 1 h. After cooling down to room temperature, 60 mL acetone was added, and the whole solution was centrifuged at 8500 rpm. After removing the supernatant, the black precipitate was dispersed in 2–3 mL chloroform, and the washing procedure was repeated at least two more times. Finally, the collected particles were dispersed in 15 mL chloroform. Fe$_3$O$_4$ nanocubes with 24 nm edge size were obtained following the same protocol but using 4 instead of 6 mmol decanoic acid.

1.1- Transfer of IONCs to water

IONCs were transferred into water by exchanging their surfactant molecules with gallic acid PEG ligand (GA-PEG) as described elsewhere. Briefly, at the end of the IONCs synthesis after reducing the temperature of the reaction mixture from 310 to 70 °C, 15 mL GA-PEG solution (0.1 M in chloroform containing 1 mL triethylamine) was injected and the resulting mixture was stirred overnight at constant temperature. Then, the mixture was allowed to cool to room temperature and was transferred to a separating funnel. 10 mL de-ionized water was added, resulting in the formation of two phases. After emulsification by means of shaking, the phases were allowed to separate and the aqueous phase containing the IONCs bearing GA-PEG was collected. This step was repeated until all nanocubes were transferred into water. Then, the solution was concentrated under reduced pressure at 40 °C to a final volume of about 50 mL and the excess of GA-PEG was removed by dialysis versus 5L de-ionized water, using cellulose membrane tubing with a pore size of 50 kDa. The sample was
dialyzed overnight at room temperature. This step was repeated five times. Finally, the IONCs solution was concentrated by centrifugation with a centrifuge filter (molecular cut-off point 100 kDa). The IONCs dispersed in aqueous media were employed for structural, colloidal and magnetic studies.

2.0- Synthesis of Co$_x$Fe$_{3-x}$O$_4$ nanocubes

Co$_x$Fe$_{3-x}$O$_4$ nanocubes (CoFeNCs) with edge sizes of 21 ± 2 nm and cobalt fraction of x=0.7 were synthesized by thermal decomposition following a procedure described elsewhere.$^3$ In a three-neck flask connected to a standard Schlenk line, 0.71 mmol (183 mg) Co(acac)$_2$, 0.84 mmol (297 mg)Fe(acac)$_3$, and 6 mmol (1.04 g) dodecanoic acid were dissolved in a mixture of 7 mL squalane and 18 mL benzyl ether. The resulting deep red solution was degassed at 65 °C for 2 h under reduced pressure of 50 mTorr. Under N$_2$ flow, the mixture temperature was increased to 200 °C at the rate of 5 °C/min and kept at 200 °C for 2 h. The reaction temperature was increased to 305 °C at the rate of 7.5 °C/min, and then the mixture was kept at reflux for 1 h. The flask was cooled down to room temperature under inert atmosphere. The black colloidal solution was washed three times with excess amount of isopropanol (10 mL) and acetone (30 mL) mixture and centrifuged at 4500 rpm for 10 min. The final particles were dispersed in chloroform (8 mL) for further measurements.

2.1- Transfer of CoFeNCs to water

The as-synthesized, hydrophobic CoFeNCs were transferred into water by using poly(maleic anhydride-alt-1-octadecene) following a protocol described previously.$^{3,5}$ Initially, 21 nm size nanocubes (3 mL with Co and Fe concentrations of 4.3 and 8.3 g/L) were diluted with excess amount of chloroform (200 mL) and sonicated for 10 min. A specific amount of poly(maleic anhydride-alt-1-octadecene) solution in chloroform (137 mM, concentration referred to the monomer unit) was added by fixing 500 molecules of monomer unit per each nm$^2$ of nanocube surface, and the solvent was evaporated under reduced pressure (fixed at 460 mbar) until complete solvent evaporation. The sample was then resuspended in 20 mL of borate buffer and was shaken overnight at 65 °C. The well-dispersed sample was concentrated to nearly 2 mL with centrifuge filters (Amicon ultra, with 100 kDa in molecular weight cut off) by sequential centrifugation steps at room temperature (RT) and 1500 rpm
for 10 min per each cycle. Subsequently, the nanocubes were separated from the free polymer by ultracentrifugation at 30000 rpm for 60 min on sucrose gradient (2 mL of 20%, 4 mL of 40%, and 3 mL of 60% in a ultracentrifuge tube, from top to bottom). Excessive polymer (visible under ultraviolet (UV) lamp) at the 20% sucrose was removed by syringe aspiration, and the nanocubes were usually found in the fraction at 40–60% of the sucrose gradient. The collected sample was washed further with a borate buffer solution (pH 9) several times to remove the excess of sucrose, and finally it was dispersed in buffer solution. The CoFeNCs dispersed in aqueous media were employed for structural, colloidal and magnetic studies.
Figure S1: Representative TEM micrographs of MNPs: A) 14 nm IONCs, B) 24 nm IONCs, C) 21 nm CoFeNCs. Size histograms of D) 14 nm IONCs, E) 24 nm IONCs, F) 21 nm CoFeNCs. Number weighted hydrodynamic size histograms of G) 14 nm IONCs, H) 24 nm IONCs, I) 21 nm CoFeNCs measured in water (black curves) and in 36% glycerol solution (blue curves). Mean values, standard deviations and polydispersity index are in Tables S1 and S2.
3.- TEM characterization

Transmission electron microscopy (TEM) was carried out on a JEOL JEM-1011 with an acceleration voltage of 100 kV. The sample preparation was conducted by drop-casting a droplet of the sample solution onto a carbon-coated copper grid 300 mesh with subsequent removal of the solvent by evaporation at room temperature. Figures S1 A-C show the representative TEM images of the studied nanoparticles. The MNP size distributions shown in Fig.S1.D-F were determined through manual size analysis of ensembles over 100-150 particles found in randomly selected areas of the enlarged micrographs, with a script of DigitalMicrograph software and the data plotted in a spreadsheet to obtain the mean size and standard deviation. Mean values and standard deviations of the studied MNPs are gathered in Table S1.

Table S1: Average MNP sizes ($l_c$) and standard deviations of the studied MNPs extracted from Fig. S1 D-F.

<table>
<thead>
<tr>
<th>MNP</th>
<th>$l_c$ (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>14 nm IONCs</td>
<td>14 ± 2</td>
</tr>
<tr>
<td>24 nm IONCs</td>
<td>24 ± 4</td>
</tr>
<tr>
<td>21 nm CoFeNCs</td>
<td>21 ± 2</td>
</tr>
</tbody>
</table>

4.- Dynamic Light Scattering measurements

In order to determine the hydrodynamic size ($D_H$) of magnetic colloids, dynamic light scattering (DLS) measurements were performed by using a Zetasizer Nano ZS90 (Malvern Instruments) equipped with a 4.0 mW He–Ne laser operating at 633 nm as energy source with an angle of 173˚ between the incident beam and the avalanche photodiode detector. The measurements were performed with an automatic scan time, and three scans per measurement. Figures S1 G-I show number weighted size distributions performed in a ZEN0112-low volume disposable sizing cuvette with nanoparticles dispersed in water and in aqueous solution of glycerol 36% at a final concentration of ~0.02 mg$_{Fe}$/mL. The refractive index was set at 1.330 for water and 1.378 for glycerol 36%; while viscosity values were set at 0.9 mPa·s and 3.2 mPa·s for water and 36% glycerol.
dispersions, respectively. General purpose (normal resolution) analysis model was employed to determine the hydrodynamic size of the magnetic colloids in water and solutions of glycerol 36% (see Table S2).

**Table S2**: Mean number weighted hydrodynamic sizes ($D_H$) and polydispersity index (PDI) of the studied MNPs dispersed in water and 36% glycerol, extracted from Figures S1G-I.

<table>
<thead>
<tr>
<th>MNP</th>
<th>Water dispersion</th>
<th>36% Glycerol dispersion</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$D_H$ (nm)</td>
<td>PDI</td>
</tr>
<tr>
<td>14 nm IONCs</td>
<td>28</td>
<td>0.15</td>
</tr>
<tr>
<td>24 nm IONCs</td>
<td>42</td>
<td>0.12</td>
</tr>
<tr>
<td>21 nm CoFeNCs</td>
<td>32</td>
<td>0.21</td>
</tr>
</tbody>
</table>

5.- **Elemental analysis**

An inductively coupled plasma atomic emission spectrometer (ICP-AES, iCAP 6500, Thermo) was used for the elemental analysis of IONCs and CoFeNCs. The samples were prepared by overnight digestion of 25μL nanoparticles solution in 2.5mL of aqua regia. Subsequently, the sample was diluted with deionized water to a final volume of 25mL.

6.- **Viscosity measurements**

The rheological properties of the different media (12 mL volume of 0%, 15%, 36%, 60%, 81% and 86% Glycerol in double distillated water) were characterized by means of viscosity measurements (DH-2 rheometer, TA Instruments, Delaware, USA) under both controlled shear and temperature conditions. The rheometer is equipped with a double gap cylinder chamber and cylindrical geometry rotating in between, in order to grant high precision measurement with low viscosity fluids. Table S3 shows the average viscosity values (two repetitions) for the different media studied by using a temperature ramp mode: specific shear rate (50 s$^{-1}$), the temperature range (15 and 35 °C) and rate (2 °C/min).
Table S3: Mean viscosity values and related standard deviations at 22º and 24ºC.

<table>
<thead>
<tr>
<th>Glycerol fraction (%)</th>
<th>Mean viscosity values at 297 K (mPa-s)</th>
<th>Mean viscosity values at 295 K (mPa-s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0.9 ± 0.1</td>
<td>1.0 ± 0.1</td>
</tr>
<tr>
<td>15</td>
<td>1.4 ± 0.1</td>
<td>1.5 ± 0.2</td>
</tr>
<tr>
<td>36</td>
<td>3.5 ± 0.1</td>
<td>3.8 ± 0.2</td>
</tr>
<tr>
<td>60</td>
<td>14.1 ± 0.10</td>
<td>15.4 ± 0.3</td>
</tr>
<tr>
<td>81</td>
<td>85.9 ± 0.1</td>
<td>97.3 ± 0.3</td>
</tr>
<tr>
<td>86</td>
<td>153.5 ± 0.3</td>
<td>176.2 ± 0.3</td>
</tr>
</tbody>
</table>

7.0- Magnetic characterization

Figure S2A: Mass-normalized magnetisation cycles measured at 260 K for 14 nm IONCs (black line), 24 nm IONCs (red line), and 21 nm CoFeNCs (blue line) dispersed in water.
Figure S2B: Mass-normalized AC hysteresis loops of 14 nm IONCs dispersed in distinct viscosity media (black line 0.9 mPa s, red line 3.5 mPa s, green line 85.9 mPa s) and under different AMF conditions (grouped by field intensity).
7.1- Calorimetry measurements

The volumetric specific heat constants considered for Milli-Q water \(C_{DDW}\) and different glycerol dispersions in water \(C_{\%\text{glycerol}}\) used were: \(C_{DDW}=4.20 \pm 0.02\ \text{J/g}^\circ\text{C}\), \(C_{15\%}=4.03 \pm 0.01\ \text{J/g}^\circ\text{C}\), \(C_{36\%}=3.55 \pm 0.03\ \text{J/g}^\circ\text{C}\), \(C_{60\%}=3.07 \pm 0.03\ \text{J/g}^\circ\text{C}\), \(C_{81\%}=2.66 \pm 0.01\ \text{J/g}^\circ\text{C}\), \(C_{86\%}=2.33 \pm 0.01\ \text{J/g}^\circ\text{C}\). These values were obtained by Discovery Differential Scanning Calorimeter (TA Instruments) at Servicio Interdepartamental de Investigación at the Universidad Autónoma de Madrid.

7.2.- Analysis of the imaginary part of ac-susceptibility: Debye model

The response of magnetic nanoparticles to an alternating magnetic field can be described using the complex Debye model given by the following equations (Eqs.):

\[
\chi'(\omega) = \chi'(\omega) + i\chi''(\omega) \quad \text{(Eq.S1)}
\]

\[
\chi'(\omega) = \frac{X_0}{1+(\omega \tau)^2} \quad \text{(Eq.S2)}
\]

\[
\chi''(\omega) = \frac{X_0 \omega \tau}{1+(\omega \tau)^2} \quad \text{(Eq.S3)}
\]

with \(\chi'\) and \(\chi''\) the real and imaginary part of AC susceptibility. For nanoparticles relaxing via both Néel and Brownian mechanisms, the imaginary part can be described by a double integration over particle core \(f(l_C)\) and hydrodynamic \(f(D_H)\) size distributions:

\[
\frac{\chi''(\omega)}{X_0} = \int_0^\infty f(D_H) \int_0^\infty \frac{\omega \tau}{1+(\omega \tau)^2} \cdot l_C^6 \cdot f(l_C) \cdot dl_C dD_H \quad \text{(Eq.S4)}
\]

where \(X_0 = \frac{\mu_0 n M^2}{3 k_B T}\).

The particle core and hydrodynamic size distributions were assumed to have a log-normal distribution function given by:
The effective relaxation time is given by the following expressions:

\[ \tau_{\text{eff}} = \frac{\tau_B \tau_N}{\tau_B + \tau_N} \]  

(Eq.S6)

where \( \tau_B \) and \( \tau_N \) are the Brownian and Néel relaxation times in absence of magnetic field (see Eq. 1 and 2 from manuscript).

The hydrodynamic size distribution parameters \( (D_H, \sigma_H) \) were derived from the analysis of particle water solutions. The \( K \) value for 14 nm IONCs was derived by setting \( l_c = 14 \) nm as obtained from TEM studies. For the analysis of 36%, 81% and 86% glycerol dispersions, \( D_H \) and \( \sigma_H \) were kept constant and \( \eta \) was refined. This is an attempt to reduce the number of fitting parameters and to obtain more reliable values (see Tables S3 and S4). It is worth noting that viscosity values obtained from fitting were close to the ones measured: \( \eta = 4.32 \) m·Pa·s for 36%, \( \eta = 70 \) m·Pa·s for 81% and \( \eta = 200 \) m·Pa·s for 86%.

**Table S4**: Results obtained from the best fits to the ACS imaginary part for 14nm IONCs using Eq.S4.

<table>
<thead>
<tr>
<th>14 nm IONCs</th>
<th>0% glycerol dispersion</th>
<th>36% glycerol dispersion</th>
<th>81% glycerol dispersion</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \eta ) (mPa.s)</td>
<td>1</td>
<td>4.33</td>
<td>70</td>
</tr>
<tr>
<td>( D_H ) (nm)</td>
<td>38</td>
<td>38</td>
<td>38</td>
</tr>
<tr>
<td>( \sigma_H )</td>
<td>0.3</td>
<td>0.3</td>
<td>0.3</td>
</tr>
<tr>
<td>( K ) (kJ/m³)</td>
<td>11.3</td>
<td>11.3</td>
<td>11.3</td>
</tr>
<tr>
<td>( \sigma_C )</td>
<td>0.17</td>
<td>0.17</td>
<td>0.15</td>
</tr>
</tbody>
</table>

For CoFeNCs where the Brownian relaxation dominates, the Néel contribution is neglected and thus Eq. S4 can be rewritten as:

\[ \frac{\chi''(\omega)}{\chi_0} = \int_0^\infty \frac{\omega \tau_B}{1 + (\omega \tau_B)^2} \cdot f(D_H) \cdot dD_H \]  

(Eq.S7)
Table S5: Results obtained from the best fits to the ACS imaginary part for CoFeNCs using Eq.S7.

<table>
<thead>
<tr>
<th>CoFeNCs</th>
<th>0% glycerol dispersion</th>
<th>36% glycerol dispersion</th>
<th>86% glycerol dispersion</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\eta$ (mPa.s)</td>
<td>1</td>
<td>4.16</td>
<td>200</td>
</tr>
<tr>
<td>$D_H$ (nm)</td>
<td>33.7</td>
<td>33.7</td>
<td>33.7</td>
</tr>
<tr>
<td>$\sigma_H$</td>
<td>0.17</td>
<td>0.17</td>
<td>0.17</td>
</tr>
</tbody>
</table>

The $\chi''$ peak of CoFeNCs is slightly asymmetric at the lower frequency side (Fig. 4), regardless of the viscosity, which is an indication of the presence of a small fraction of clusters. This fraction is so insignificant that it is not visible in the DLS result (Fig. S1). In order to exclude its effect on the fit results, the low frequency side of the ACS imaginary peak was not considered in the simulations. To include this portion, a bi-modal hydrodynamic size distribution should be taken into account. This implies two more fitting parameters, compromising the validity of the other parameters, and requiring longer computation times.

7.3- Numerical Simulations of hysteresis loops

7.3.1- Method

When physical rotation of MNPs is allowed, the dynamics of the magnetic moment results from the combination of Brownian and Néel processes. The dynamics of a unit vector along the easy axis $\vec{n}$ is given by the Langevin equation$^6$:

$$\frac{d\vec{n}}{dt} = \frac{\mu_0}{6\eta V_H} \vec{m} \cdot \vec{H}_{ex} \vec{n} + \sqrt{\frac{k_B T}{6\eta V_H}} \vec{f}$$  \hspace{1cm} (Eq.S8)

where $\mu_0$ is the permeability of free space, $\eta$ is the viscosity of the surrounding medium, $V_H$ is the nanoparticle hydrodynamic volume, $\vec{m}$ is the magnetic moment vector of the MNP, $\vec{H}_{ex}$ is the external AC magnetic field,
and $k_B T$ is the thermal energy. The random torque ($\vec{F}$) on the MNP caused by thermal fluctuations satisfies the following equations:

$$\langle \vec{F}_i \rangle = 0$$  \hspace{1cm} (Eq.S9)

$$\langle \vec{F}_i(t) \vec{F}_j(t') \rangle = 2 \delta_{ij} \delta(t - t')$$  \hspace{1cm} (Eq.S10)

Here, $\langle \rangle$ represents the average over an ensemble, $i$ and $j$ are Cartesian indices, $\delta_{ij}$ is the Kronecker delta function, and $\delta$ is the Dirac delta function.

On the other hand, the dynamics of the magnetic moment vector $\vec{m}$ is given by Equation 4 (see Material and Methods Section).

The fluctuating magnetic field due to thermal noise, $\vec{H}_{th}$, satisfies the following equations:

$$\langle \vec{H}_{th,i} \rangle = 0$$  \hspace{1cm} (Eq.S10)

$$\langle \vec{H}_{th,i}(t) \vec{H}_{th,j}(t') \rangle = \frac{2\gamma \delta_{ij}}{1 + \lambda^2 \gamma m \mu_0^2} \delta(t - t')$$  \hspace{1cm} (Eq.S11)

where $\gamma$ is the gyromagnetic ratio, and $m = M_s l_0^3$ is the magnitude of the magnetic moment, and $\lambda$ is a dimensionless damping coefficient. Equation 4 (see Material and Methods Section) is the stochastic LLG equation in which thermal fluctuations are taken into account. Magnetic torque due to the anisotropy field $\vec{H}_k$ is included for the dynamics of the magnetic moment vector $\vec{m}$, i.e., Néel rotational mechanism. This torque acts on $\vec{m}$ so that it aligns along the direction of the easy axis. For large $K$ or $V$ values, $\vec{m}$ attempts to stay in one of the directions of the easy axes. In other words, Néel relaxation time becomes longer when increasing $K$ and/or $V$ values. Besides, magnetic torques due to an external field $\vec{H}_{ex}$ are included in both the Brownian and Néel rotational mechanisms as shown in Eqs.6 and 7 (see Materials and Methods section). This means that Brownian and Néel relaxation times become shorter when increasing the intensity of $\vec{H}_{ex}$. In this manner, the field-dependent Brownian and Néel relaxation times are implicitly taken into account in the numerical simulation.

The dynamic behavior of the magnetic moment as well as the easy axis for a single MNP can be calculated by solving Eqs.S8 and 4 (at Materials and Methods section) simultaneously. In the simulation, Eqs. S8 and 4 were
discretised with respect to time $t$. The discretisation interval $\Delta t$ was set to $0.01\tau_0 = 0.01M_s/2\gamma \lambda K = 10^{-11}$ s, where $\lambda = 0.1$, $\gamma = 1.74 \times 10^{11}$ rad/T·s for 14 nm IONCs and $\gamma = 0.955 \times 10^{11}$ rad/T·s for 21 nm CoFeNCs were used. The dynamics of the magnetic moment vector $\mathbf{m}$ and unit vector along the easy axis $\mathbf{n}$ were calculated for $N = 7168$ MNPs. Consequently, the average values $\langle \mathbf{m} \rangle$ and $\langle \mathbf{n} \rangle$ over the ensemble were obtained. The numerical simulation was carried out until an equilibrium magnetization was obtained.

In Figure S3 and S4, numerical simulation results of AC $M - H$ and $\langle \cos \theta_k \rangle - H$ curves for an external AC magnetic field with amplitude $\mu_0 H_{\text{MAX}} = 5.2$ kA/m and frequency $f = 1$ kHz are shown, respectively. Here, $\theta_k$ represents the angle between the direction of the easy axis $\mathbf{n}$ of the $k$-th particle and the external field $\mathbf{H}$. $\langle \cos \theta_k \rangle$ is an ensemble average over $N$ particles.

Figure S3 A and S4 A show the simulation results for MNPs with $m = 6.33 \times 10^{-19}$ Am$^2$, $K = 70$ kJ/m$^3$, $V = 1.15 \times 10^{-24}$ m$^3$, $V_H = 1.44 \times 10^{-24}$ m$^3$, and $\eta = 0.957$ mPa·s. In this case, Brownian process is dominant, since Brownian and Néel relaxation times are calculated as $\tau_B = \frac{3\eta V_H}{k_B T} = 10^{-6}$ s and $\tau_N = \frac{\sqrt{\pi}}{2\sqrt{kV/k_BT}} \tau_0 \exp \left( \frac{kV}{k_BT} \right) = 0.078$ s. As seen in Figure S4A, $\langle \cos \theta_k \rangle$ changes depending on the external field, which indicates that the magnetization occurs via Brownian rotational process. The dotted line in Figure S3 A shows a Langevin function, which is given by $L \left( \frac{\mu_0 m H}{k_B T} \right) = \coth \left( \frac{\mu_0 m H}{k_B T} \right) - \frac{k_B T}{\mu_0 m H}$. It is well recognized that the $M - H$ curve is given by Langevin function when $\tau_B$ is much faster than the change of the AC external field, i.e., $\tau_B \ll 1/2\pi f$. As can be seen, the $M - H$ curve nicely fits the Langevin function. This result indicates the validity of the numerical simulation.

Figure S3 B and S4 B show the simulation results for MNPs with $m = 6.33 \times 10^{-19}$ Am$^2$, $K = 28.6$ kJ/m$^3$, $V = 1.15 \times 10^{-24}$ m$^3$, $V_H = 2.72 \times 10^{-22}$ m$^3$, and $\eta = 0.1$ Pa·s. In this case, Néel process is dominant, since their relaxation times are calculated as $\tau_B = 0.020$ s and $\tau_N = 10^{-6}$ s, respectively. As can be seen, $\langle \cos \theta_k \rangle$ is almost constant, which indicates that the Brownian process is blocked and the magnetization reversal follows
Néel process. As shown above, by simultaneously solving Eqs. S8 and 4 (at Material and Methods Section), we can calculate the magnetisation of MNPs dispersed in different viscosity media.

For numerical simulations of AC magnetisation for 14 nm IONCs and 21 nm CoFeNCs, some parameters were set to the values obtained from static magnetisation, TEM, and AC susceptibility measurements. The following values were used for 21 nm CoFeNCs: $M_s = 546$ kA/m, and $D_H = 36$ nm, and for 14 nm IONCs: $M_s = 374$ kA/m, and $D_H = 38$ nm. Since MNP volume depends on the edge MNP size, $l_C$ defines the Néel relaxation time. For 21 nm CoFeNCs, the edge size distribution will not much affect the viscosity dependence of AC magnetisation. This is because the Néel relaxation time is sufficiently large for such large edge size, i.e., the Brownian relaxation mechanism is dominant (independently of the edge size). For this reason, a single edge size $l_C = 21$ nm was used in the simulation for 21 nm CoFeNCs. However, for 14 nm IONCs, the edge size distribution was taken into account since the dominancy of the relaxation mechanism will change depending on the edge size as well as anisotropy constant, or field intensity. A lognormal distribution -as depicted by the dotted line in Figure S1D- was used for including edge size distribution of 14 nm IONCs in the numerical simulations. Such modeling was performed for each edge size over the range of $10$ nm $\leq l_C \leq 21.5$ nm with an interval of $\Delta l_C = 0.5$ nm for 14 nm IONCs. Then, the AC magnetization was calculated as:

$$\frac{M}{M_s} = \frac{1}{\nu_T} \sum_i n_i V_i \langle m_i \rangle \Delta l_C$$  \hspace{1cm} (Eq.S12)

Here, $n_i$ is a Gaussian function and $V_T = \sum_i n_i V_i \Delta l_C$ is the total volume of the IONCs. Finally, we considered $K$ as a free fitting parameter. Figure S5 shows the comparison between experimental and simulated AC hysteresis loops at 100 kHz and 24 kA/m for MNPs dispersed in water.

**7.3.2- Field dependent relaxation times**

In the numerical simulations given by Eqs. S8 and 4 (at Material and Methods Section), field-dependent relaxation times are implicitly included (see Figure S6). Those field-dependent expressions are Eqs. 6 and 7 (see Material and Methods Section).
Figure S3: Numerical simulation results of AC normalized $M - H$ curves for MNPs with (A) $\tau_B = 10^{-6}$ s and $\tau_N = 0.078$ s and (B) $\tau_B = 0.02$ s and $\tau_N = 10^{-6}$ s. Dotted lines represent a Langevin function.

Figure S4: Numerical simulation results of $<\cos \theta>$-H curves for MNPs with (A) $\tau_B = 1\mu$s and $\tau_N = 78\mu$s and (B) $\tau_B = 20\mu$s and $\tau_N = 1\mu$s.
Figure S5: Comparison of experimental (solid line) and simulated (dashed line) AC hysteresis loops under $H_{dc}$ (100 kHz and 24 kA/m) at $1g_{Fe}/L$ for 21 nm CoFeNCs, and 14 nm IONCs dispersed in water.

Figure S6: Viscosity dependences of A) the field-dependent relaxation times calculated by Eqs.6 and 7, B) AC normalized $M-H$ curves calculated by Eq.5 for CoFeNCs under $H_{dc}$ (100kHz and 24 kA/m).

References