

## Dispersions of magnetic nanoparticles in Water/ Ionic Liquid mixtures.

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### ELECTRONIC SUPPORTING INFORMATION

#### S1 Table of conversion of the compositions of the mixtures water EAN

Table showing the mass fractions converted in different other units, for values corresponding to the samples and some important points. The mixture is considered as ideal, which induces a very small error according to Hadded et al<sup>1</sup>. Several relevant lengths are also given (see in Lee et al<sup>2</sup>)

$w_{EAN}$	$\Phi_{EAN}$	[EAN] mol/L	[H <sub>2</sub> O] mol/L	$x_{EAN}$	$x_{H_2O}$	H <sub>2</sub> O/EAN	$\lambda_D$	$a/\lambda_D$	$l_B$
0	0	0	55.5	0	1	$\infty$	$\infty$	0	0.72
0.003	0.0025	0.028	55.4	0.0005	0.999	1994	1.82	0.24	0.72
0.0051	0.0042	0.047	55.3	0.00085	0.999	1170	1.40	0.31	0.72
0.007	0.0058	0.065	55.2	0.00117	0.999	851	1.19	0.36	0.72
0.0104	0.0086	0.096	55.1	0.0017	0.998	571	0.98	0.44	0.72
0.02	0.017	0.19	54.6	0.0034	0.997	294	0.70	0.61	0.72
0.03	0.025	0.28	54.2	0.0051	0.995	194	0.57	0.75	0.73
0.05	0.042	0.47	53.2	0.0087	0.991	114	0.44	0.98	0.74
0.106	0.09	1	50.6	0.0194	0.98	50.6	0.29	1.47	0.77
0.21	0.18	2	45.6	0.042	0.96	22.6	0.20	2.17	0.84
0.41	0.36	4.1	35.3	0.104	0.90	8.63	0.13	3.42	1.03
0.54	0.49	5.5	28.2	0.16	0.84	5.1	0.10	4.26	1.18
0.75	0.71	8	16	0.33	0.67	2	0.073	5.85	1.54
0.9	0.88	9.9	6.6	0.6	0.4	0.67	0.060	7.16	1.86
0.94	0.93	10.4	4	0.72	0.28	0.38	0.057	7.6	1.96
1	1	11.2	0	1	0	0	0.052	8.2	2.14

Table S1 - Mass fraction of EAN  $w_{EAN}$ , Volume fraction of EAN  $\Phi_{EAN}$ , concentration of EAN [EAN] mol/L, concentration of water [H<sub>2</sub>O] mol/L, molar fraction of EAN  $x_{EAN}$ , molar fraction of water  $x_{H_2O}$ , number of molecules of water per EAN H<sub>2</sub>O/EAN, Debye length  $\lambda_D$  in nm, Parameter  $a/\lambda_D$  with  $a = 0.43$  nm the size of the ions, Bjerrum length  $l_B$  in nm.

#### S2 Interaction NP/Solvent - Activation energy at large $w_{EAN}$ determined by FRS relaxation.

Fig. S1-left shows that  $D_m/T$  determined by FRS relaxation, when interparticle interaction is repulsive and weak (here at  $\Phi = 1\%$  for  $\chi \sim 1$ ) can be analysed as a function of  $1/kT$  in the framework of an Andrade model<sup>3</sup> and adjusted with an Arrhenius law, as the results obtained by DLS (see Fig. 4 of main text). The values of activation energy  $E_a$ , obtained by the fit and given in the figure caption, are fully compatible with those obtained by Zarrougui et al from viscosity measurements in<sup>4</sup>, namely  $E_a = 0.23$  eV at  $w_{EAN} = 1$  and  $E_a = 0.175$  eV at  $w_{EAN} = 0.9$ . Moreover, this is also true in the attractive area (with  $\chi > 1$ ) down to  $w_{EAN} = 0.55$ , where the interparticle interaction has been checked to be  $T$ -independent. Corresponding values of Zarrougui et al are  $E_a = 0.16$  eV at  $w_{EAN} = 0.75$  and  $E_a = 0.15$  eV at  $w_{EAN} = 0.55$ .

Note that  $D_m$  measurements at low values of  $w_{EAN}$  and  $\chi \sim 1$ , which can be realised by DLS, are not possible by relaxation of FRS, the optical signal being too low in the region where the Soret coefficient  $S_T$  is too small.

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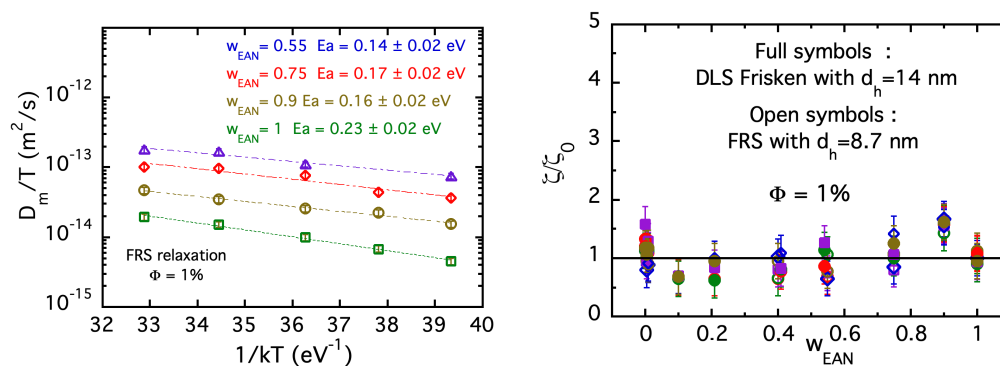


Fig. S1 - (left)  $D_m/T$  as a function of  $1/kT$  for various  $w_{EAN}$  in the range 0.55 to 1.  $D_m$  is deduced from FRS relaxation measurements at  $\Phi = 1\%$ . Full lines are fits to an Arrhenius law  $D_m/T \sim e^{-E_a/kT}$ . Values of  $E_a$  deduced from the fits are given in the figure. - (right) Reduced friction  $\zeta/\zeta_0$  experienced by the NPs in the dispersions in water/EAN mixtures at  $\Phi = 1\%$  at various temperatures:  $T = 22^\circ\text{C}$  (red symbols),  $T = 33^\circ\text{C}$  (green symbols),  $T = 47^\circ\text{C}$  (purple symbols),  $T = 64^\circ\text{C}$  (blue symbols),  $T = 80^\circ\text{C}$  (brown symbols), deduced from DLS (full symbols) and FRS relaxation (open symbols). The analysis (using Eqs. 1 and 3 of main text) is performed with the hypothesis that  $\chi$  is  $T$ -independent.

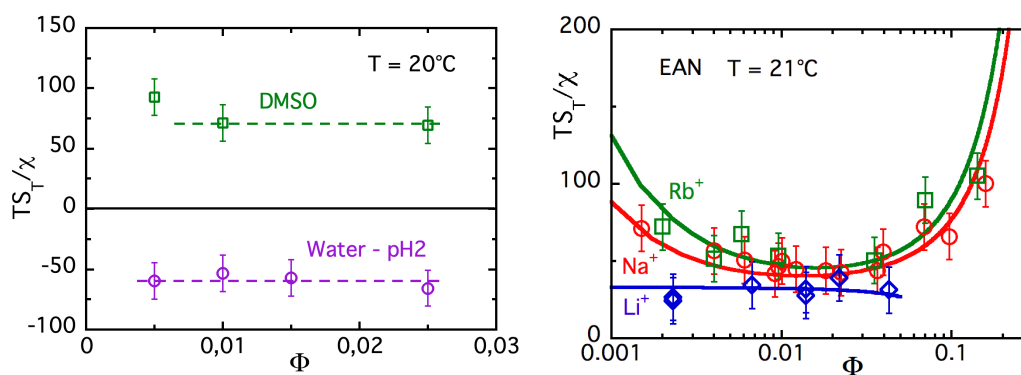


Fig. S2 - Variations of  $TS_T/\chi$  as a function of  $\Phi$  at room  $T$  - (Left) for dispersions in DMSO and in water at  $pH = 2$  of NPs with  $d_{NP} = 9$  nm and  $ClO_4^-$  counterions, probed in<sup>5</sup>. Dotted horizontal lines are guides for the eye. - (right) for dispersions in pure EAN of same NPs as in the main text with  $d_{NP} = 7.4$  nm and three kinds of counterions  $Na^+$ ,  $Rb^+$  and  $Li^+$ . Data are from<sup>6</sup> and full lines are fits of  $TS_T/\chi$  as a function of  $\Phi$  detailed in<sup>6</sup>.

### S3 Friction experienced by the NPs

$D_m$  is analysed with Eq. 1 of main text to deduce the evolution of the friction  $\zeta$  experienced by the NPs. It is presented in Fig. S1-right in a reduced representation, normalised by  $\zeta_0$  given by Eq. 3 of the main text. The analysis of the DLS and FRS data at  $\Phi = 1\%$  is done with the hypothesis that  $\chi$  is independent of  $T$  (which is experimentally verified only for  $w_{EAN} \geq 0.65$ ).  $\zeta/\zeta_0$  can be seen as the ratio  $d_h^{eff}/d_h$  where  $d_h$  is the hydrodynamic diameter determined at room temperature and  $\chi \sim 1$ .  $d_h$  is given in the legend of the figure and is different for DLS and FRS, as the experiments do not average the optical intensity in the same way.

### S4 Dependence of $TS_T/\chi$ on $\Phi$ in various kinds of NPs dispersions.

#### S4.1 Dispersions of hydroxyl coated NPs in DMSO and in water

The first example concerns (see Fig. S2-left) hydroxyl coated NPs dispersed in water at  $pH = 2$  and in DMSO with the counterion  $ClO_4^-$  from<sup>5</sup>, the third moment of the NP diameter distribution being  $d_{NP} = \sqrt[3]{\langle d^3 \rangle} = 9$  nm. Fig. S2-left shows that whatever  $\Phi$  in the experimental range  $0.01 \leq \Phi \leq 0.025$ ,  $TS_T/\Phi$  is within the error bar independent on  $\Phi$  in both solvents.

#### S4.2 Dispersions of citrate coated NPs in EAN

The second example concerns the same citrate coated NPs (the third moment of the NP diameter distribution being  $d_{NP} = \sqrt[3]{\langle d^3 \rangle} = 7.4$  nm as in the main text) dispersed in pure EAN, which have been studied in<sup>6</sup>. Fig. S2-right shows  $TS_T/\chi$  as a function of  $\Phi$  at room  $T$  for three different kinds of NPs counterions,  $Na^+$ ,  $Rb^+$  and  $Li^+$ . We reproduce here Fig. S6 of ESI of ref.<sup>6</sup>, showing that whatever the counterion, the quantity  $TS_T/\Phi$  is, at room temperature and for the counterion  $Na^+$ , independent of  $\Phi$  in the typical range  $0.6\% - 3\%$ .

### Notes and references

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