

## Supporting Information

# One-Pot Synthesis of Water-Soluble and Biocompatible Superparamagnetic Gadolinium-doped Iron Oxide Nanoclusters

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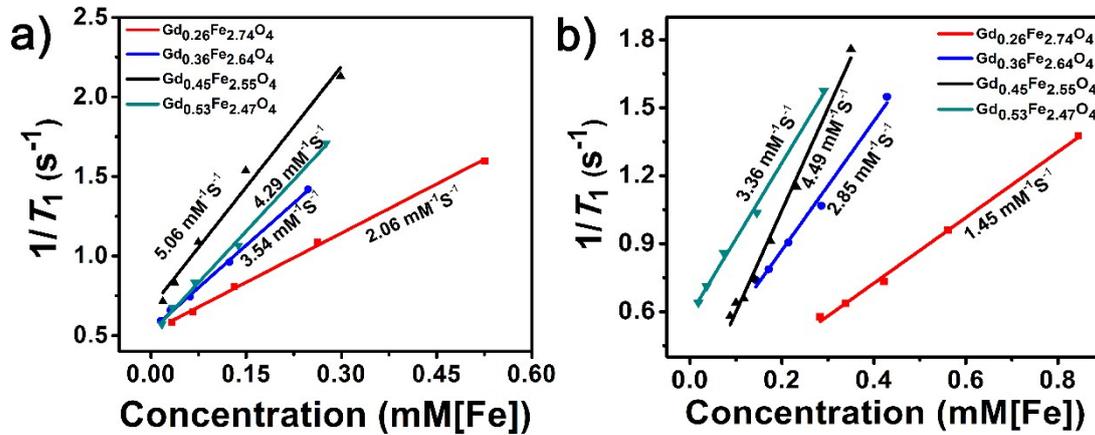
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Table S1. The  $r_2$  and  $r_1$  of GdIO NCs with different Gd doping amounts at 1.5 T

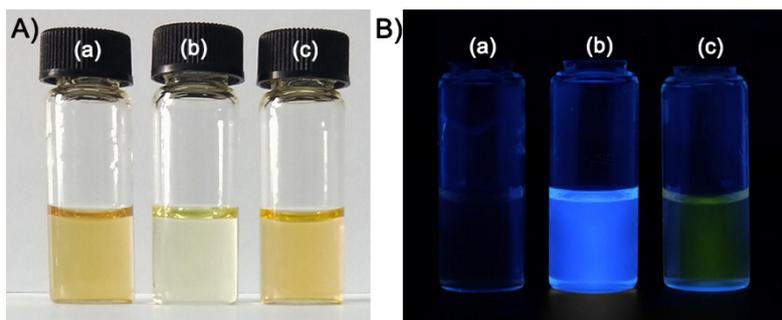
GdIO NCs	$r_1$ ([Fe] mM·s <sup>-1</sup> )	$r_2$ ([Fe] mM·s <sup>-1</sup> )	$r_2/r_1$
Gd <sub>0.26</sub> Fe <sub>2.74</sub> O <sub>4</sub>	2.06	844.8	410.1
Gd <sub>0.36</sub> Fe <sub>2.64</sub> O <sub>4</sub>	3.54	974.0	275.1
Gd <sub>0.45</sub> Fe <sub>2.55</sub> O <sub>4</sub>	5.06	859.7	169.9
Gd <sub>0.53</sub> Fe <sub>2.47</sub> O <sub>4</sub>	4.29	703.8	164.1

Table S2. The  $r_2$  and  $r_1$  of GdIO NCs with different Gd doping amounts at 7 T

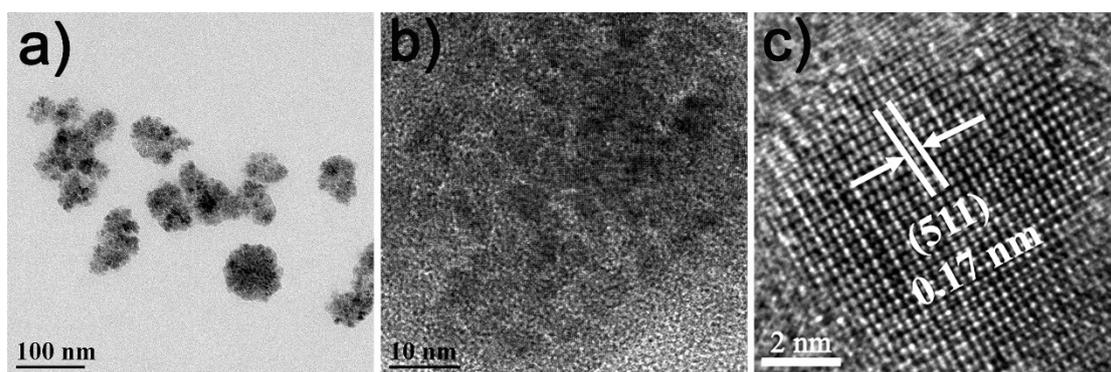
GdIO NCs	$r_1$ ([Fe] mM·s <sup>-1</sup> )	$r_2$ ([Fe] mM·s <sup>-1</sup> )	$r_2/r_1$
Gd <sub>0.26</sub> Fe <sub>2.74</sub> O <sub>4</sub>	1.45	765.7	528.1
Gd <sub>0.36</sub> Fe <sub>2.64</sub> O <sub>4</sub>	2.85	768.5	269.6
Gd <sub>0.45</sub> Fe <sub>2.55</sub> O <sub>4</sub>	4.49	633.6	141.1
Gd <sub>0.53</sub> Fe <sub>2.47</sub> O <sub>4</sub>	3.36	566.3	168.5



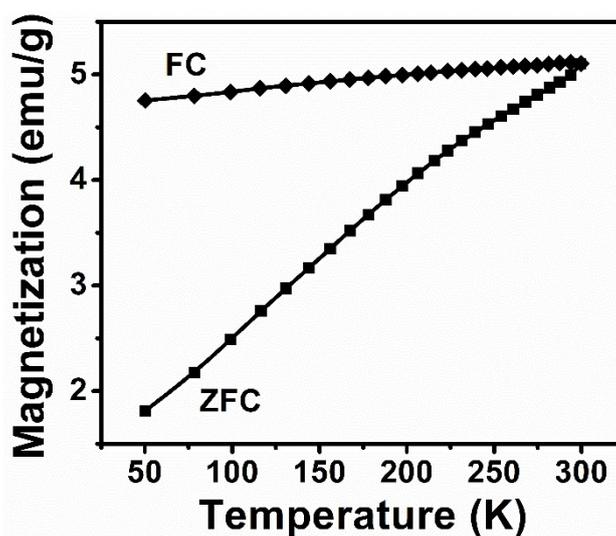
**Fig. S1.** Plots of the inverse longitudinal relaxation times ( $1/T_1$ ) versus Fe concentrations at (a) 1.5 T and (b) 7 T, respectively.



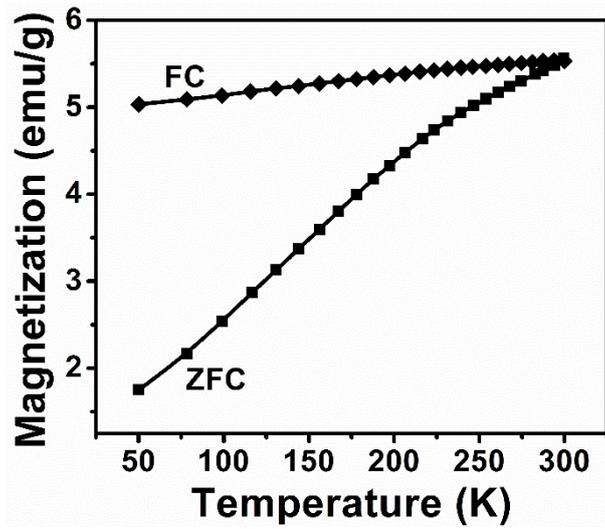
**Fig. S2.** A) Photograph of aqueous suspensions of (a)  $\text{Gd}_{0.45}\text{Fe}_{2.55}\text{O}_4$  NCs, (b) FITC alone and (c) FITC@ $\text{Gd}_{0.45}\text{Fe}_{2.55}\text{O}_4$  NCs. B) Photograph under 365 nm UV irradiation.



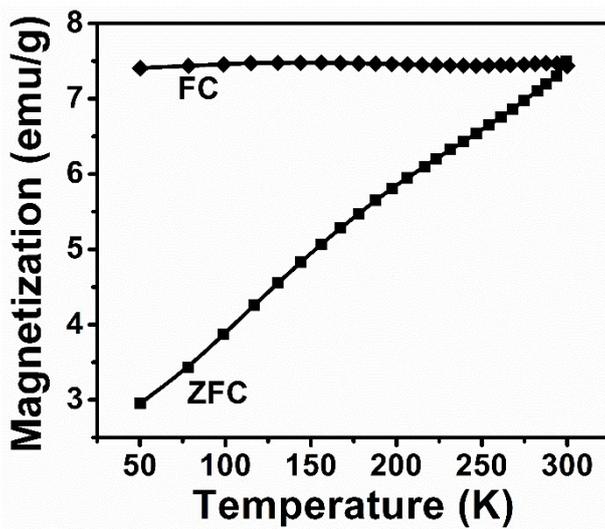
**Fig. S3** TEM and high-resolution TEM images of  $\text{Gd}_{0.53}\text{Fe}_{2.47}\text{O}_4$  NCs.



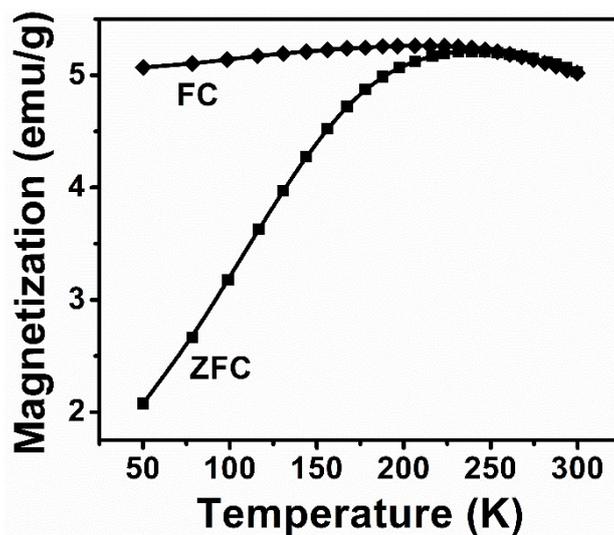
**Fig. S4** The field-cooling (FC) and zero-field-cooling (ZFC) magnetization of  $\text{Gd}_{0.26}\text{Fe}_{2.74}\text{O}_4$  NCs as a function of temperature measured in an external magnetic field (50 Oe).



**Fig. S5** The field-cooling (FC) and zero-field-cooling (ZFC) magnetization of  $\text{Gd}_{0.36}\text{Fe}_{2.64}\text{O}_4$  NCs as a function of temperature measured in an external magnetic field (50 Oe).



**Fig. S6** The field-cooling (FC) and zero-field-cooling (ZFC) magnetization of  $\text{Gd}_{0.45}\text{Fe}_{2.55}\text{O}_4$  NCs as a function of temperature measured in an external magnetic field (50 Oe).



**Fig. S7** The field-cooling (FC) and zero-field-cooling (ZFC) magnetization of  $Gd_{0.53}Fe_{2.47}O_4$  NCs as a function of temperature measured in an external magnetic field (50 Oe).