Supplementary information to: **Size and property bimodality in magnetic nanoparticle dispersions: Single domain particles vs strongly coupled nanoclusters**

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ADDITIONAL STRUCTURAL CHARACTERIZATION OF NANOCLUSTERS

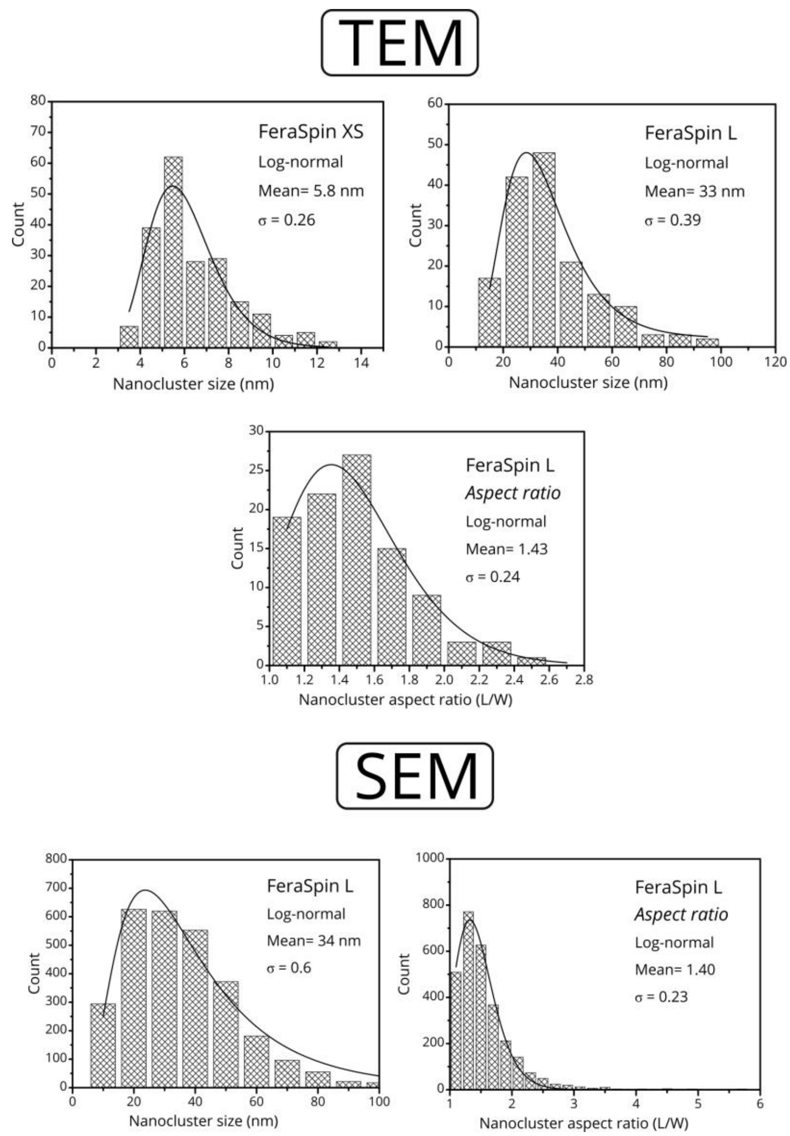

Figure S 1: Histogram of the nanocrystals (FeraSpin XS) and nanocluster (FeraSpin L) sizes, and the aspect ratio (L/W) for FeraSpin L as determined by SEM and TEM.

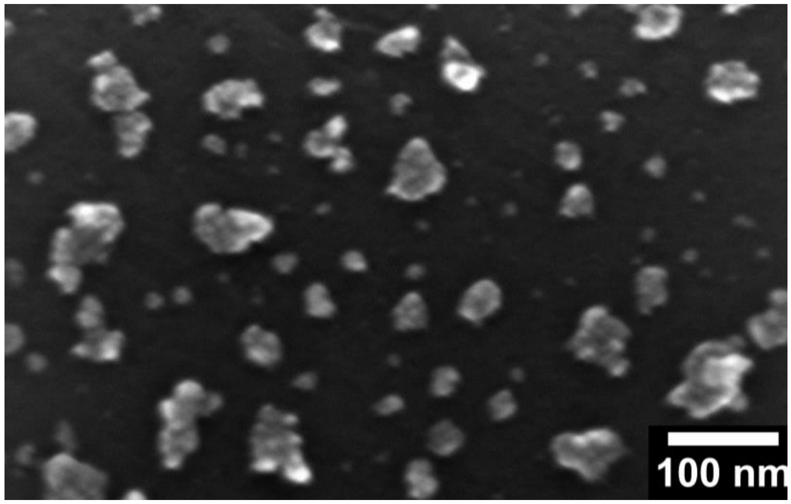

Figure S 2: SEM image of FeraSpin L nanoclusters immobilized on a Si substrate. Please refer to the experimental section for more details on the sample preparation.

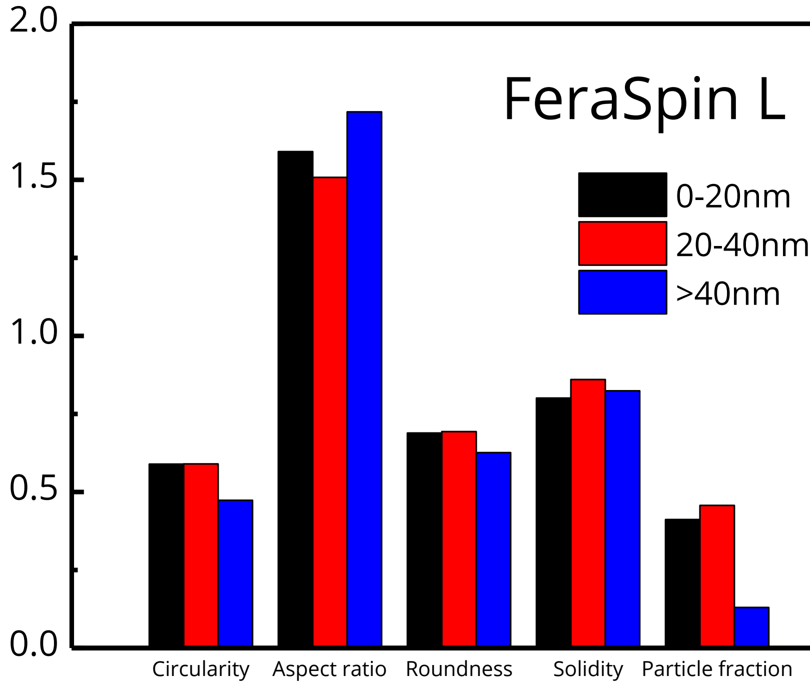

Figure S 3: Particle characteristics for Feraspin L derived from the SEM analysis using ImageJ, sorted in three different size regimes. Definitions are here as in the documentation of ImageJ. *Aspect ratio:* the aspect ratio of the fitted ellipse to the particle projection, i.e., [Major Axis] / [Minor Axis]. *Roundness:* (4 × [Area]) / (π × [Major axis]^2) or the inverse of [Aspect ratio]. *Solidity*: [Area]/[Convex hull area], where convex hull can be thought of as a rubber band wrapped tightly around the projection of the particle. For example, a circle, ellipse, triangle or rectangle would have Solidity = 1, while star- or flower-like shape would have Solidity <1. *Circularity*: (4π × [Area])/[(Perimeter]^2) with a value of 1.0 indicating a perfect circle. As the value approaches 0.0, it indicates an increasingly elongated shape.

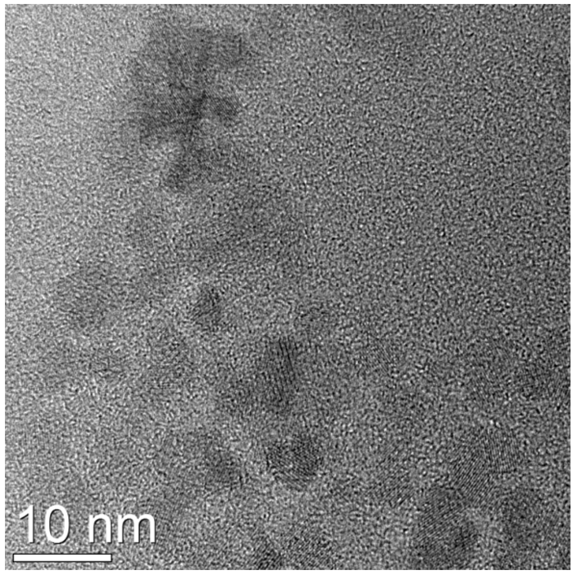

Figure S 4: Soft aggregate in FeraSpin XS, a possible drying artefact during TEM sample preparation. The absence of nanoclusters of larger sizes in the AF4 fractograms, FMR spectra and temperature dependent AC-magnetometry suggest that these agglomerates are either statistically insignificant or formed during de-wetting of the water film evaporation of the dispersion.

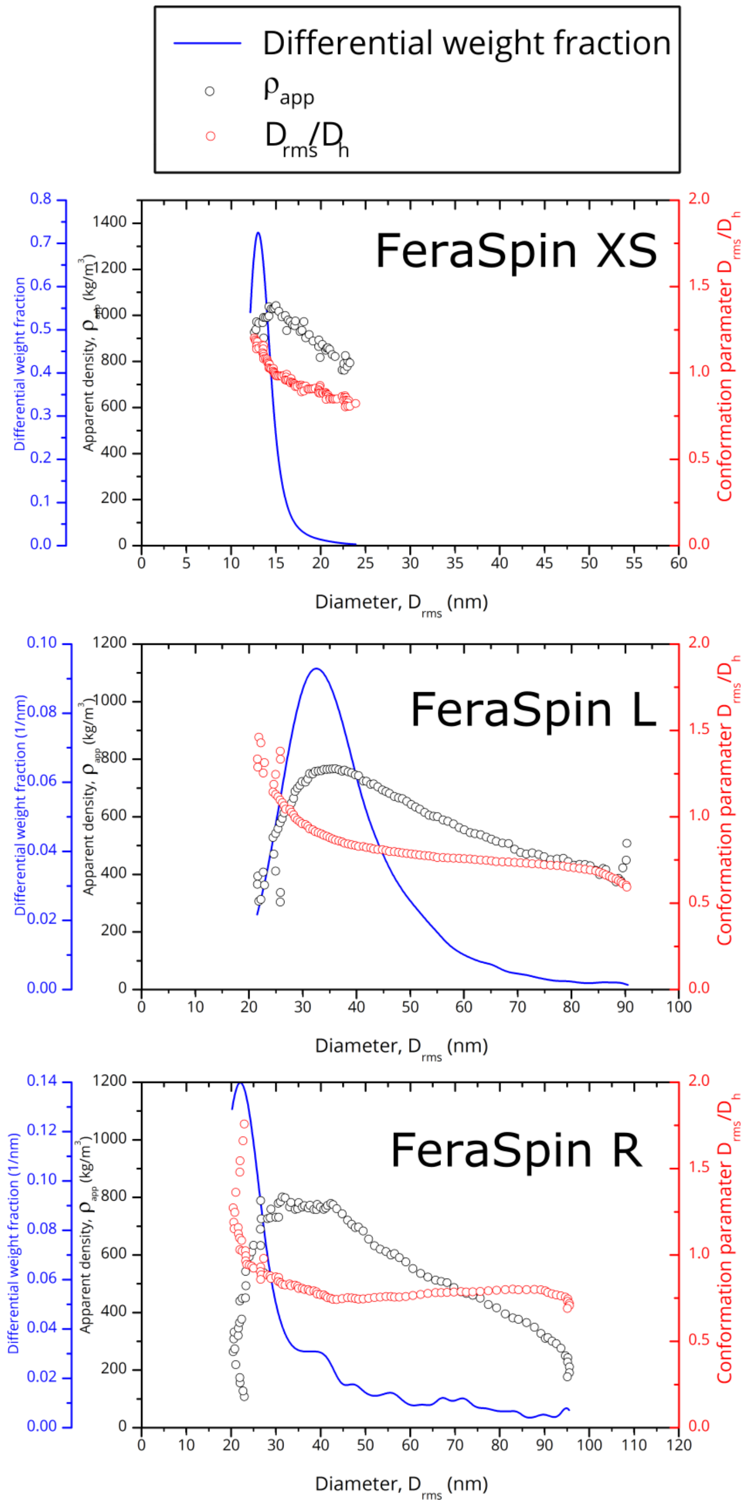

Figure S 5: Differential weight fractions, apparent density (ρ_{app}), and conformational parameter (D_{rms}/D_h) as a function of D_{rms} for dispersions of FeraSpin XS, L, and R.

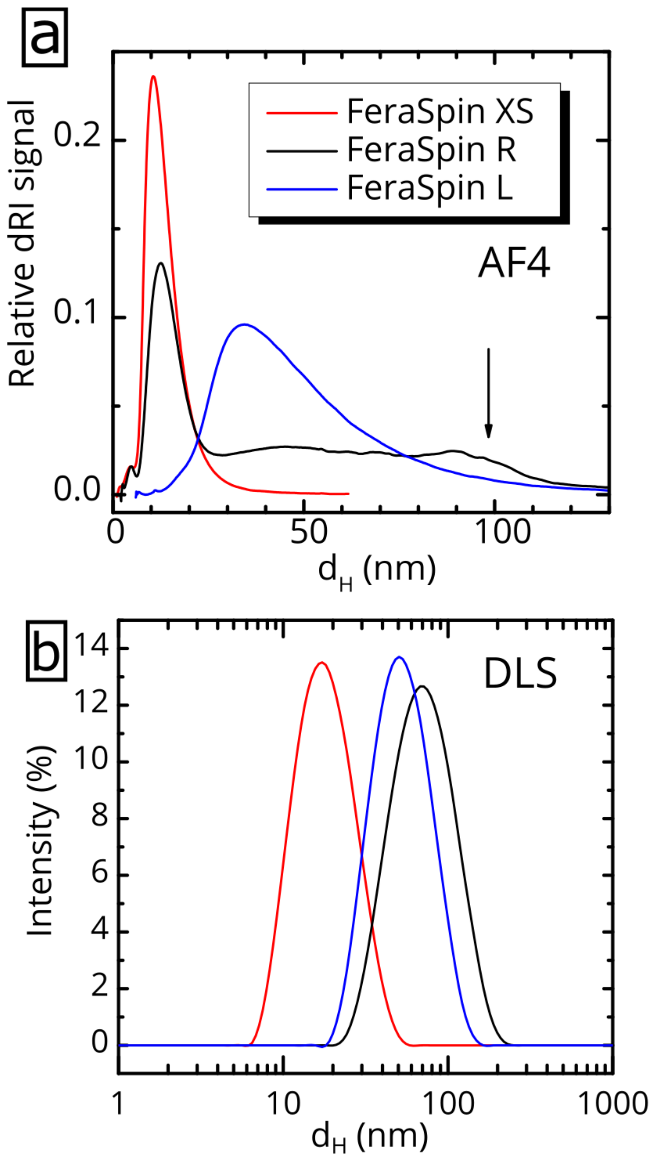

Figure S 6: Comparison of the relative signals of the (a) AF4 refractive index fractograms for the three samples FeraSpin XS, L and R with (b) DLS data. As seen in the figure, FeraSpin R has a longer tail (see arrow) than FeraSpin L, and subsequently has a slightly wider size distribution of colloidal nanoclusters than FeraSpin L.

CALCULATION OF THE DEMAGNETIZATION FACTORS FOR ELLIPSOIDAL PARTICLES

For a general (prolate) ellipsoid with semi-axes *a* > *b* = *c* we have that the components of the demagnetization tensor *N* are N_{zz}, N_{yy}, N_{zz} (for the semi-axes *a*, *b*, *c* respectively). In general, we have that, $N_{zz} + N_{yy} + N_{xx} = 4\pi$, and when $m = a/c$:

$$\frac{N_{zz}}{4\pi} = \frac{1}{m^2-1}\left(\frac{m}{2\sqrt{(m^2-1)}} \times \ln\left(\frac{m+\sqrt{(m^2-1)}}{m-\sqrt{(m^2-1)}}\right) - 1\right),$$

$$\frac{N_{xx}}{4\pi} = \frac{N_{yy}}{4\pi} = \frac{m}{2(m^2-1)}\left(m - \frac{1}{2\sqrt{(m^2-1)}} \times \ln\left(\frac{m+\sqrt{(m^2-1)}}{m-\sqrt{(m^2-1)}}\right)\right),$$

The effective demagnetization factor can then be found through $N_{eff} = N_{\perp} - N_{//}$,where $N_{xx} = N_{yy} = N_{\perp} > N_{//} = N_{zz}$. Using a value of $m = \frac{a}{c} = L/W$ (as shown if fig. S1) gives N_{eff} = 0.13.

ADDITIONAL FMR DATA

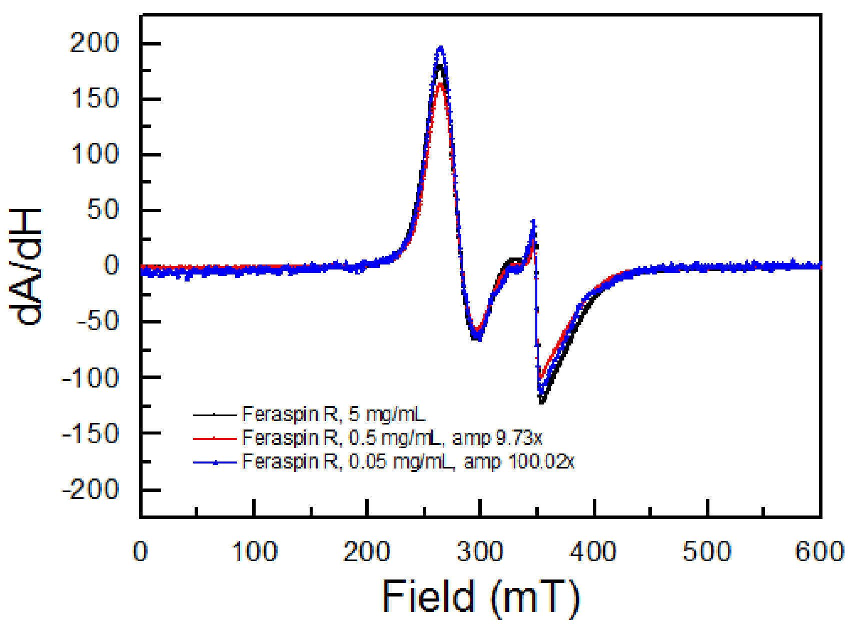

Figure S 7: A concentration series of FeraSpin R diluted ≈ 10x and 100x. The spectra have been renormalized according to the actual dilution factor, determined by weight. Although the data is rather noisy, there are no obvious concentration effects seen after renormalization.

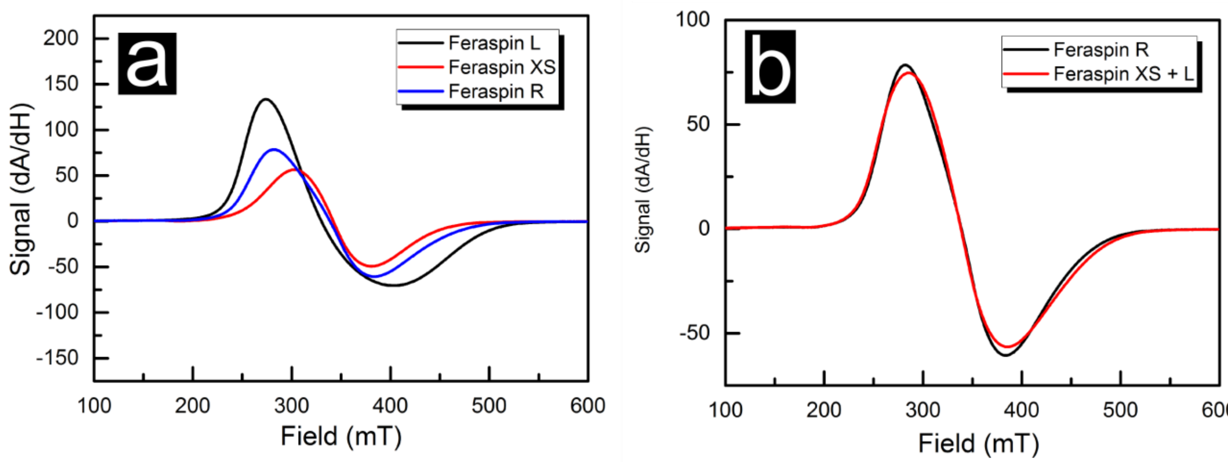

Figure S 8: Additional FMR spectra of immobilized particles. (a) FeraSpin XS, R and L adsorbed in cotton wool. (b) Linear combination fit of the FeraSpin XS + L spectra to the FeraSpin R spectrum.

ADDITIONAL MAGNETIZATION DATA

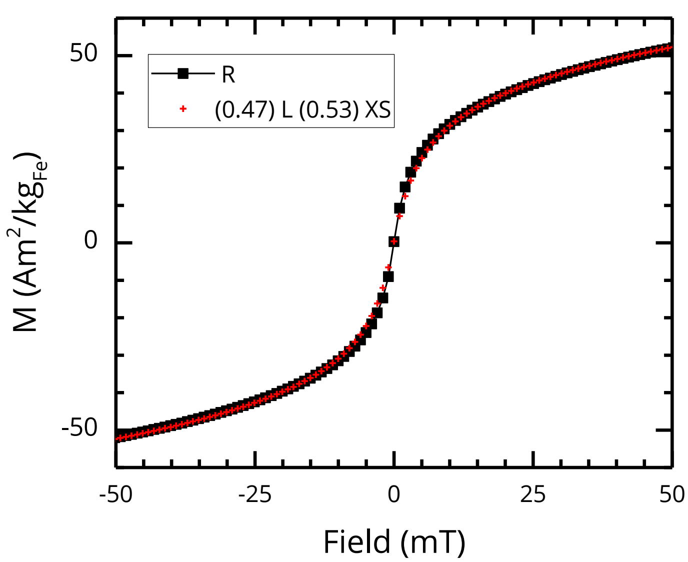

Figure S 9: Fit of the FeraSpin R magnetization (*M* vs. *H*) curve using a linear combination of the magnetization curves of FeraSpin XS and FeraSpin L. As seen in the graph, there is a slight mismatch between the original and composite curves in the low-field (< 10 mT) region.

Table S 1: Calculation of K_{eff} using the intra-well potential approximation given in the main paper (eq. 1). Other approximations used are ρ = 5 g/cm^3 and m(Fe)/m(Fe_xO_y) = 0.7 (valid for γ-Fe_2O_3). We estimated errors assuming an error in the magnetic moment of 3%.

Sample	**$M_s(0)$**	**Fitted** **$\chi_{\perp} T\rightarrow 0$** *dimensionless* *SI units*	**Effective anisotropy** *AC* ($\chi_{\perp}$ T→0) *10^4 J/m^3*
FeraSpin XS	110 Am^2/kg$_{Fe}$ 385 kA/m	2.25	2.8(2)
FeraSpin L	120 Am^2/kg$_{Fe}$ 420 kA/m	3.50	2.1(2)