Long-Term Colloidally Stable Aqueous Dispersions of 
\( \leq 5 \) nm Spinel Ferrite Nanoparticles

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1. Surface Functionalization

1.1 FT-IR Analysis

Figure S2. FT-IR spectra of citrate-functionalized spinel ferrite and iron oxide nanoparticles, as well as trisodium citrate (spectra stacked for clarity). The peak of the symmetric stretching vibration $v_{\text{sym}}$ of the carboxyl group remains at 1386 cm$^{-1}$ for all ferrite nanoparticles, while it shifts slightly to 1380 cm$^{-1}$ for iron oxide. The peak of the asymmetric stretching vibration $v_{\text{asym}}$ shifts from 1580 to 1566 cm$^{-1}$ for citrate molecules interacting with spinel ferrite surfaces, and to 1590 cm$^{-1}$ for iron oxide particles. The separation of the symmetric and asymmetric modes is $\Delta \nu = v_{\text{asym}}(\text{COO}^-) - v_{\text{sym}}(\text{COO}^-) = (1566 - 1386) \text{ cm}^{-1} = 180 \text{ cm}^{-1}$ for the ferrites, whereas it is $\Delta \nu = (1590 - 1382) \text{ cm}^{-1} = 208 \text{ cm}^{-1}$ for the iron oxide particles. These peak shifts evidence the coordination of citrate to the surfaces of the nanoparticles, but likely different binding modes co-exist.

FT-IR measurements support ligand binding on the nanoparticles surface. The data on citrate-functionalized ferrite as well as iron oxide nanoparticles is presented exemplarily in Figure S2, highlighting the diagnostic spectral region of carbonyl resonances, 1800 cm$^{-1} > \nu > 1300$ cm$^{-1}$. Clearly the asymmetric carboxylate mode of the capping citrate is susceptible to the nature of $M^{2+}$ in $MFe_2O_4$, whereas the symmetric mode remains almost invariant. It is noteworthy that both a negative and a positive shift of the asymmetric stretching mode are observed, in comparison to bulk trisodium citrate. For the ferrite nanoparticles with $M^{2+} \neq Fe^{2+}$, the mode shifts to slightly lower wavenumbers, resulting in an overall smaller separation $\Delta \nu$ of the
symmetric and asymmetric modes of $180 \text{ cm}^{-1}$, calculated from $\nu_{\text{asym}} (\text{COO}^-) - \nu_{\text{sym}} (\text{COO}^-)$. For iron oxide nanoparticles, on the other hand, the shift is to larger separation $\Delta \nu = 208 \text{ cm}^{-1}$. Derived from computational and FT-IR studies of citrate adsorbed on hematite nanoparticles, this latter finding suggests mainly binuclear bidentate configuration of citrate on the ferrite nanoparticles, and a higher relevance of monodentate binding on the iron oxide nanoparticles.\[1\] It is known, that on metal oxide surfaces, many different binding modes – monodentate, bidentate, salicylate – often co-exist.\[2\] Since the peaks in our FT-IR spectra are broad, a unique distinction of binding modes cannot be made.

![Figure S3. FT-IR spectra of betaine-functionalized iron oxide nanoparticles, pure betaine and DEG. The nanoparticles exhibit absorption bands in the region of the carboxylic asymmetric stretching vibration of betaine between $1730 - 1520 \text{ cm}^{-1}$, confirming the coordination of betaine to the surface of the nanoparticles. Additionally, the symmetric (1124 cm$^{-1}$) and asymmetric (1050 cm$^{-1}$) C-O stretching vibrations of DEG are present. Due to the interaction between the polyol oxygen and metal cation both modes slightly shift to higher wavenumbers, as recently shown for DEG-coated cobalt ferrite nanopowder.\[3\] This confirms the coexistence of betaine and DEG on the nanoparticle surface. The existence of DEG residuals was similarly observed also for sodium citrate and phosphocholine functionalized iron oxide nanoparticle powders.

1.2 TGA and CHN Analysis

![Figure S4. TGA data of iron oxide nanoparticles, showing the percentage weight loss (blue line, left y-axis) and its derivative (dTG, black line, right y-axis) for capping with A) phosphocholine, B) betaine and C) citrate. The weight loss up to about $200^\circ\text{C}$ (highlighted by red arrows) is assigned to the loss of residual moisture (H$_2$O). The mass loss due to decomposition of the organic ligands occurs between about $200^\circ\text{C}$ and $930^\circ\text{C}$, $860^\circ\text{C}$ and $735^\circ\text{C}$, respectively.}
Table S1. Details of surface coverage with ligand and water molecules as derived by combination of elemental (CHN) analysis and TGA for iron oxide nanoparticles capped with the three different ligands, namely phosphocholine, citrate and betaine.

Elemental analysis yields the amounts of C, H, and N in the samples in wt%. The ligand content in the samples can be calculated using the molar mass of the ligand molecules (phosphocholine: C₅H₁₅NO₄P, M = 184.13 g mol⁻¹; betaine: C₅H₁₁NO₂, M = 117.14 g mol⁻¹; citrate: C₆H₅O₇, M = 189.04 g mol⁻¹). The experimental N:C and N:H ratios deviated from the corresponding nominal ratios for phosphocholine and betaine. An excess of C was identified and assumed to originate from the solvent diethyleneglycol (DEG), which obviously could not be completely removed from the nanoparticle surface during the ligand-exchange process. Presence of DEG on the nanoparticle surfaces is further suggested by FT-IR (see Figure S6). The amount of H, which is still remaining after consideration of ligand molecules and DEG, is supposed to stem from surface water. For citrate, the existence of residual DEG cannot be checked with elemental analysis, because citrate possesses no hetero atom besides O for clear distinction between ligand and DEG amounts (amount of O not accessible with elemental analysis). Hence, all C was assumed to originate from citrate. Spare H is assumed to be from surface water.

The total amount of organics as well as the amount of surface water obtained with CHN-analysis are compared to respective values from TGA. The amount of H₂O in the samples with TGA is evaluated according to Figure S3. For phosphocholine-stabilized sample, the total amount of organics and of H₂O match well. For betaine-capped samples, the amount of water determined with the two methods also agrees well, yet, the total amount of organics is 3.7 % higher in TGA analysis. This difference could be due to a loss of surface oxygens from the crystalline nanoparticle surface at elevated temperatures⁴, since the total amount of organics in the sample is relatively low suggesting a lot of uncovered surface. Results from elemental analysis and TGA analysis differ a lot for the citrate-capped sample. This is likely due to the presence of DEG on the NP surface, which however cannot be quantified.

|            | content of molecule wt% | content of molecule in μmol |
|------------|-------------------------|----------------------------|
|            | Phos  | DEG   | H₂O  | Phos  | DEG   | H₂O  |
| Phosphocholine                      |       |       |      |       |       |      |
| 25.0       | 13.7  | 3.9   | 3.2  | 3.0   | 5.1   |      |
| total organics CHN-analysis:         |       |       |      |       |       | 42.6 wt% |
| total weight loss TGA:               |       |       |      |       |       | 41.1 wt% |
| Weight loss due to H₂O TGA:          |       |       |      |       |       | 5.0 wt % |
| Citrate                                  |       |       |      |       |       |      |
| 25.9       | -     | 10.5  | 3.3  | -     | 13.8  |      |
| total organics CHN-analysis:         |       |       |      |       |       | 36.4 wt % |
| total weight loss TGA:               |       |       |      |       |       | 28.7 wt % |
| weight loss due to H₂O TGA:          |       |       |      |       |       | 4.0 wt % |
| Betaine                                   |       |       |      |       |       |      |
| 2.8        | 8.3   | 1.3   | 0.7  | 2.3   | 2.1   |      |
| total organics CHN-analysis:         |       |       |      |       |       | 12.4 wt % |
| total weight loss TGA:               |       |       |      |       |       | 16.1 wt % |
| weight loss due to H₂O TGA:          |       |       |      |       |       | 1.4 wt % |
2. Magnetic Characterization

Figure S5. SQUID magnetometry field dependence for spinel ferrite and iron oxide nanoparticles during a field cycle at 300 K (open symbols). Lines are fits to the Langevin equation.

The magnetic response of the dried citrate-stabilized iron oxide and spinel ferrite nanoparticles has been addressed by field-dependent SQUID magnetometry at temperature $T = 300$ K. As is commonly observed with magnetic nanoscale materials with $d < 10$ nm, the magnetization tends to vary steeply with the field around $|H| = 0$ without hysteresis. Results recorded with nickel ferrite nanoparticles are exemplarily shown in Figure 1 (for the complete series, see Figure S5). Magnetization levels off at fields $|H| > 30.000$ Oe with saturation magnetizations $M_s$ that are clearly influenced by the nature of the $M^{2+}$ species in the spinel ferrites. In order to quantify the influence of ad-metal ion $M^{2+}$, the magnetization curves were fitted with the Langevin equation[6]

$$\frac{M}{M_s} = \coth(\alpha) - 1 / \alpha; \quad \text{with} \quad \alpha = \mu_{\text{part}} \times H / k_B \times T$$  \hspace{1cm} (1)$$

with the Boltzmann constant $k_B$ and magnetizing field $H$. $\mu_{\text{part}}$ denotes the (formal) magnetic moment of a single nanoparticle. Satisfactory fits to the Langevin equation could be obtained in all cases, yielding $M_s$ which decreases along the series $\text{Fe}^{2+} (40.1 \text{ emu g}^{-1}) > \text{Co}^{2+} (24.0 \text{ emu g}^{-1}) > \text{Zn}^{2+} (16.0 \text{ emu g}^{-1}) > \text{Ni}^{2+} (11.5 \text{ emu g}^{-1}) > \text{Mg}^{2+} (7.7 \text{ emu g}^{-1})$. This order largely reflects the variation of the bulk saturation magnetization $M_s(\text{bulk})$, which scales negatively with decreasing particle dimensions.
3. Structure Characterization

3.1 ICP-OES Analysis

Inductively coupled plasma optical emission spectrometry (ICP-OES) measurements have been performed for all heterothallic ferrites $\text{MFe}_2\text{O}_4$ ($\text{M}^{2+} \neq \text{Fe}^{2+}$), to determine the $\text{Fe}^{3+}:\text{M}^{2+}$ ratio. As the surface functionalization was achieved at the end of the synthesis, after nanoparticle nucleation and growth, the ligands do not impact the particle composition. According to the initial precursor ratio in the synthesis, the ideal $\text{Fe}^{3+}:\text{M}^{2+}$ ratio should be close to 2.0. In fact, close-to-ideal stoichiometry prevails for $\text{M}^{2+} = \text{Mg}^{2+}$ and $\text{Co}^{2+}$ with the $\text{Fe}^{3+}:\text{M}^{2+}$ ratio between 2.0 and 2.1, except for one sample of magnesium ferrite, for which it is 2.7. For $\text{M} = \text{Ni}^{2+}$, the ratio varies for the three samples between 1.7 and 2.4, resulting in an average value of 2.0. Merely, the zinc ferrite nanoparticles deviate from the nominal composition for all investigated samples with $\text{Fe}^{3+}:\text{M}^{2+}$ ratios ranging from 2.9 to 3.2, indicating that too little $\text{Zn}^{2+}$ is incorporated into the spinel structure. This coincides with investigations by Kodama et al. on the incorporation of $\text{Zn}^{2+}$ ions into the spinel ferrite crystal structure: even a 12-fold excess of $\text{Zn}^{2+}$ to $\text{Fe}^{3+}$ ions in the precursor solution yields in a high number of $\text{Zn}^{2+}$ vacancies and thus in 27 % maghemite. Hence, the metal compositions in the ferrite nanoparticles follow the intended precursor ratios for $\text{Mg}^{2+}$ and $\text{Co}^{2+}$, rather well for $\text{Ni}^{2+}$, but deviate for $\text{Zn}^{2+}$.

3.2 PXRD Analysis

![XRD pattern](image)

**Figure S6.** Background-corrected XRD patterns of dry powders of betaine-stabilized ferrite nanoparticles (background = empty capillary; patterns stacked for clarity). All nanopowders are crystalline and can be indexed with the cubic spinel structure $Fd-3m$. Different heights and FWHM of Bragg reflexes stem from different nanoparticle diameters. Here, for clarity, the theoretical XRD pattern of CoFe$_2$O$_4$ is shown exemplarily; the lattice parameters of the different spinels vary slightly. For NiFe$_2$O$_4$, few small sharp peaks (stars) at $Q = 1.92, 2.23, 3.15, 4.45, 4.98$ and $5.45 \text{ Å}^{-1}$ stem from betaine monohydrate, as the ligand was used in excess.

XRD and PDF analysis confirm the high crystallinity of all ferrite and iron oxide nanoparticles, which results from the high temperatures maintained during synthesis. All ferrite XRD patterns can be indexed with the cubic spinel structure (see section 3.2 in SI). Despite the deviation of the zinc to iron ratio in the zinc ferrite nanoparticles, no side phases could be detected in its XRD patterns. Merely, the XRD of nickel ferrite
particles shows reflexes of an additional side phase, probably arising from betaine monohydrate. Bragg reflexes are broadest for magnesium ferrite nanoparticles, originating from the smaller particle diameter in comparison with the other spinel ferrite and iron oxide nanoparticles, and as additionally confirmed by TEM and SAXS (see Table 2).

### 3.3 PDF Analysis

**Table S2.** Refined parameters for PDF refinements of betaine-capped spinel ferrites without Occ<sub>oct</sub> (see Figure S9 A).

|                          | CoFe<sub>2</sub>O<sub>4</sub> | ZnFe<sub>2</sub>O<sub>4</sub> | NiFe<sub>2</sub>O<sub>4</sub> | MgFe<sub>2</sub>O<sub>4</sub> |
|--------------------------|-------------------------------|-------------------------------|-------------------------------|-------------------------------|
| a = b = c / Å (start)    | 8.394                         | 8.442                         | 8.347                         | 8.397                         |
| a = b = c / Å             | 8.407                         | 8.439                         | 8.368                         | 8.390                         |
| crystallite size / Å      | 43                            | 47                            | 49                            | 35                            |
| U<sub>iso,Co/Zn/Ni/Mg</sub> / Å<sup>2</sup> | 0.009                         | 0.008                         | 0.010                         | 0.023                         |
| U<sub>iso,Fe</sub> / Å<sup>2</sup> | 0.009                         | 0.011                         | 0.007                         | 0.007                         |
| U<sub>iso,O</sub> / Å<sup>2</sup> | 0.021                         | 0.025                         | 0.018                         | 0.023                         |
| z(O)                     | -0.631                        | -0.632                        | -0.631                        | -0.631                        |
| R<sub>w</sub>             | 0.15                          | 0.16                          | 0.30                          | 0.18                          |

**Table S3.** Refined parameters for PDF refinements of betaine-capped spinel ferrites including the parameter of octahedral occupancy Occ<sub>oct</sub> (see Figure S7 and S9 B). For the sample NiFe<sub>2</sub>O<sub>4</sub> the value of Occ<sub>oct</sub> is out of the physical reliable range due to a certain amount of side phase, impacting the PDF peak at 3.0 Å.

|                          | CoFe<sub>2</sub>O<sub>4</sub> | ZnFe<sub>2</sub>O<sub>4</sub> | NiFe<sub>2</sub>O<sub>4</sub> | MgFe<sub>2</sub>O<sub>4</sub> |
|--------------------------|-------------------------------|-------------------------------|-------------------------------|-------------------------------|
| a = b = c / Å (start)    | 8.394                         | 8.442                         | 8.347                         | 8.397                         |
| a = b = c / Å             | 8.407                         | 8.410                         | 8.368                         | 8.390                         |
| crystallite size / Å      | 43                            | 47                            | 49                            | 34                            |
| U<sub>iso,Co/Zn/Ni/Mg</sub> / Å<sup>2</sup> | 0.009                         | 0.010                         | 0.006                         | 0.021                         |
| U<sub>iso,Fe</sub> / Å<sup>2</sup> | 0.009                         | 0.010                         | 0.009                         | 0.007                         |
| U<sub>iso,O</sub> / Å<sup>2</sup> | 0.022                         | 0.029                         | 0.016                         | 0.029                         |
| z(O)                     | -0.631                        | -0.632                        | -0.631                        | -0.631                        |
| Occ<sub>oct</sub>        | 0.99                          | 0.84                          | 1.13                          | 0.73                          |
| R<sub>w</sub>             | 0.15                          | 0.16                          | 0.29                          | 0.19                          |
PDF refinements based on the cubic spinel structure $Fd-3m$ describe the experimental data well in a fit range of 1.7 to 50 Å. Goodness-of-fit values $R_w$ lie between 0.15 and 0.19 for betaine-capped ferrites – except for NiFe$_2$O$_4$ due to a betaine-side phase (see Figure S6 and Table S3). Refinements of the citrate-stabilized particles result in $R_w$ values of 0.18 to 0.32, and phosphocholine-capped particles yield $R_w$ values of 0.16 to 0.50 due to stabilizer residuals. Particle diameters can be extracted from the refinements (contained in Table 2), and approximate particle diameters are visible in the experimental PDFs when peak heights decay to zero (see Figure S7 for refinements of betaine-stabilized cobalt and magnesium ferrite, for instance). For all three stabilizers, magnesium ferrites have the highest $R_w$ values, i.e. lowest fit qualities, throughout.
Figure S8. PDF refinements of betaine-stabilized cobalt, zinc and magnesium ferrite nanoparticles A) without and B) with the fit parameter $\text{Occ}_{\text{oct}}$. Fit ranges are 1.7 - 50 Å for zinc and cobalt ferrites, and 1.7 - 40 Å for magnesium ferrites

Exemplarily, Figure S8a shows the short-range order up to 5 Å from the PDF refinements of betaine-capped ferrites (Nickel one excluded due to betaine side phase). For the magnesium and zinc ferrite particles, the peak height ratios of octahedral and tetrahedral sites at 3.0 and 3.5 Å are not entirely matched. Hence, we introduced a parameter into the fit to account for possible vacancies on the octahedral cation positions ($\text{Occ}_{\text{oct}}$). In both cases, the fit results in reduced occupancies of 0.73 and 0.84 for magnesium and zinc, respectively (Figure S8 B and Table S3). These octahedral vacancies seem to describe the local structure up to 5 Å slightly better, while the goodness-of-fit of the entire range up to 50 Å is not improved. This shows that the average structure is not dominated by vacancies. The initial fit quality with $R_W = 0.18$ for magnesium and 0.16 for zinc was already very high and small residual structural features in the difference curve are rather due to disorder in the very small magnesium ferrite nanoparticles, and due to non-stoichiometry for zinc ferrites (see ICP-OES results). Oxidation as source of structural changes can be ruled out, as the bivalent metal ions $M^{2+} \neq \text{Fe}^{2+}$ are stable against oxidation in air.
Figure S9. PDF refinements of betaine-stabilized iron oxide nanoparticles taking the space group A) $Fd-3m$ and B) $P4_32_22$ with refinement of the occupancy of the octahedral iron site Fe(4) ($\text{Occ}_{\text{Fe(4)}}$). Fits were carried out over 1.7 - 50 Å, yet for highlighting the essential structural feature of the octahedral iron site, only the range of 1.7 - 5 Å is displayed. Refining $\text{Occ}_{\text{Fe(4)}}$ results in an occupancy of 0.52 and an overall better quality of fit in the short range order, showing that a significant maghemite content exists in the iron oxide nanoparticles. Here, a phase ratio of 27 % magnetite and 73 % maghemite is derived according to Cooper et al.[8]

Figure S10. PDF refinements of betaine-stabilized iron oxide nanoparticles taking the space group $Fd-3m$. The complete fit-range is shown.
Figure S11. PDF refinements of betaine-stabilized iron oxide nanoparticles taking the space group $P4_32_12$ with refinement of the occupancy of the octahedral iron site Fe(4) ($\text{Occ}_{\text{Fe(4)}}$). The complete fit-range is shown.

Figure S12. PDF refinements of A) citrate- and B) phosphocholine-stabilized iron oxide nanoparticles taking the space group $P4_32_12$ with refinement of the occupancy of the octahedral iron site Fe(4) ($\text{Occ}_{\text{Fe(4)}}$). The complete fit range is shown.
Table S4. Refined parameters for PDF fits of iron oxide nanoparticles using the space group \( Fd-3m \) and \( P4_32_12 \). Starting values for lattice parameters are \( a = b = c = 8.397 \ \text{Å} \) (\( Fd-3m \)) and \( a = b = 8.337 \) and \( c = 8.322 \ \text{Å} \) (\( P4_32_12 \)). Fits are shown in figures S8 and S9.

| Sample Space group \( a=b / \text{Å} \) | \( \text{Fe}_2\text{O}_4 \) - betaine | \( \text{Fe}_2\text{O}_4 \) - betaine | \( \text{Fe}_2\text{O}_4 \) - citrate | \( \text{Fe}_2\text{O}_4 \) - phos |
|---------------------------------|----------------|----------------|----------------|----------------|
| \( c / \text{Å} \) | 8.393 | 8.364 | 8.417 | 8.404 |
| Crystallite size / Å | 8.393 | 8.435 | 8.333 | 8.348 |
| \( U_{iso,Fe} / \text{Å}^2 \) | 0.010 | 0.005 | 0.007 | 0.008 |
| \( U_{iso,O} / \text{Å}^2 \) | 0.023 | 0.007 | 0.007 | 0.011 |
| \( O_z \) | -0.623 | | | |
| \( O_{1_x} \) | 0.617 | 0.602 | 0.618 | |
| \( O_{1_y} \) | 0.855 | 0.866 | 0.873 | |
| \( O_{1_z} \) | -0.005 | -0.003 | -0.021 | |
| \( O_{2_x} \) | 0.085 | 0.134 | 0.144 | |
| \( O_{2_y} \) | 0.365 | 0.379 | 0.381 | |
| \( O_{2_z} \) | -0.006 | -0.002 | 0.002 | |
| \( O_{3_x} \) | 0.122 | 0.131 | 0.135 | |
| \( O_{3_y} \) | 0.875 | 0.875 | 0.883 | |
| \( O_{3_z} \) | -0.015 | 0.014 | 0.026 | |
| \( O_{4_x} \) | 0.383 | 0.418 | 0.384 | |
| \( O_{4_y} \) | 0.636 | 0.624 | 0.629 | |
| \( O_{4_z} \) | -0.001 | -0.031 | -0.010 | |
| \( Occ_{Fe(4)} \) | 0.52 | 0.37 | 0.33 | |
| \( R_w \) | 0.17 | 0.17 | 0.16 | 0.18 |

PDF refinements of the iron oxide nanoparticles based on the cubic \( Fd-3m \) structure cannot describe the intensity ratios for peaks at interatomic distances of 3.0 and 3.5 Å (Figure S9 A and S10). These peaks correspond to the distance between two iron ions occupying each an octahedral site (3.0 Å peak), and the distance between metal ions on an octahedral and a tetrahedral site (3.5 Å). The fraction of tetrahedral sites, i.e. height of the 3.5 Å peak, is consistently underestimated. This could, for instance, stem from surface faceting of magnetite particles with tetrahedral iron sites. Vacancies in the particles could affect the intensity ratio of the 3.0 and 3.5 Å peaks in a similar way. According to Cooper et al., the intensity ratio of these two peaks reflects the fraction of maghemite present in magnetite nanoparticles, since magnetite nanoparticles readily undergo oxidation either during synthesis or when stored in air.[8] Maghemite has unoccupied octahedral sites compared to magnetite. It can be described with the tetragonal space group \( P4_32_12 \), with an occupancy of 33 % on the Fe(4) site (\( Occ_{Fe(4)} \)), while magnetite would have an occupancy of 100 %.[9] For our citrate- and phosphocholine-capped iron oxide particles this occupancy parameter refined to 0.33, i.e. pure maghemite, and simultaneously \( R_w \) values improved from 0.21 to 0.16 for citrate- and from 0.24 to 0.18 for phosphocholine-stabilized particles, respectively (see Figure S12). Both particles feature small diameters of 3.5 nm, which more readily oxidize entirely to maghemite in contrast to larger particles. For our larger 5 nm betaine-capped iron oxide nanoparticles, the fit quality with the tetragonal (maghemite) model over a fit range from 1.7 to 50 Å was not improved compared to the cubic structure (identical \( R_w = 0.17 \), see Table S4). However, in the short range of 1.7 – 5.0 Å, peak height ratios of octahedral and tetrahedral sites


are better described within the tetragonal model with a refined $\text{Occ}_{\text{Fe}(4)} = 0.52$, and interatomic distances are better met due to fitted oxygen positions (Figure S9 B and S11). If we convert this occupancy to molar phase fractions according to Cooper et al., our 5 nm betaine-capped iron oxide particles consist of 73 % maghemite and 27 % magnetite, respectively a 0.9 nm thick maghemite shell around a magnetite core.

4. Size Determination and Stability

4.1 TEM Analysis
Figure S13. Representative TEM images of each type of spinel ferrite nanoparticles with different organic ligands, together with histograms and the lognormal fit of the size distribution of at least 100 particles.
4.2 SAXS Analysis

![Figure S14](image1.png)

**Figure S14.** SAXS data and their fits of freshly synthesized phosphocholine-stabilized ferrite nanoparticle dispersions at a concentration of 5 g L\(^{-1}\). SAXS patterns are offset for clarity. Refined particle diameters and standard deviations are given to the right.

![Figure S15](image2.png)

**Figure S15.** Experimental SAXS data and their fits of aqueous dispersions of citrate-stabilized iron oxide nanoparticles at concentrations of 5 (purple, bottom), 10 (light blue, middle) and 100 g L\(^{-1}\) (red, top curve). These iron oxide nanoparticles were synthesized with reaction conditions from Ref. 26 and 31 of the main text. Namely those reaction conditions were a precursor solution concentration of 0.08 M and a NaOH solution concentration of 0.45 M with a hold time of 2 hours. The altered reaction conditions increased the size of the obtained iron oxide nanoparticles. Curves are stacked for clarity. Data was collected 30 days after synthesis.
### 4.3 DLS Analysis

**Table S5.** Particle diameter according to DLS after storage as aqueous dispersion (c = 5 g L\(^{-1}\)).

| NP composition | Stabilizer          | Particle diameter / nm | Ageing time / months |
|----------------|---------------------|------------------------|----------------------|
| CoFe\(_2\)O\(_4\) | citrate             | 5.4 ± 1.4              | 10                   |
|                 |                     | 4.8 ± 1.1              | 2                    |
|                 | betaine             | 5.0 ± 1.5              | 2                    |
|                 |                     | 4.9 ± 1.0              | 8.5                  |
| Fe\(_3\)O\(_4\)  | citrate             | 4.4 ± 1.0              | 3                    |
|                 |                     | 4.9 ± 1.5              | 2                    |
|                 | phosphocholine      | 3.7 ± 0.9              | 2                    |
| MgFe\(_2\)O\(_4\) | citrate             | 87.1 ± 14.1            | 2                    |
|                 | betaine             | 4.6 ± 1.2              | 7                    |
|                 | phosphocholine      | 5.0 ± 1.3              | 7                    |
| NiFe\(_2\)O\(_4\) | citrate             | 5.2 ± 1.1              | 10                   |
|                 |                     | 4.5 ± 1.0              | 2                    |
|                 | betaine             | 5.1 ± 1.2              | 8                    |
|                 | phosphocholine      | 6.5 ± 1.6              | 8.5                  |
|                 |                     | 5.4 ± 1.2              | 2                    |
| ZnFe\(_2\)O\(_4\) | citrate             | 6.0 ± 1.1              | 10                   |
|                 |                     | 5.5 ± 0.9              | 2                    |
|                 | betaine             | 5.4 ± 1.4              | 8                    |
|                 |                     | 7.6 ± 2.2              | 2                    |
|                 | phosphocholine      | 6.3 ± 1.4              | 8.5                  |
|                 |                     | 6.4 ± 1.4              | 2                    |

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