## **Synergistic interactions of binary suspensions of magnetic anisotropic particles - Supplementary Information**†

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## 1 The magnetic field of a uniformly magnetized ellipsoid

The magnetic potential outside the ellipsoidal particle (ξ > 0) generated by the magnetization along the *x*−axis of the particle results $1$ 

$$
\phi = \frac{r_x r_y r_z}{2} x M_x \int_{\xi}^{\infty} \frac{1}{(\alpha + r_x^2) \sqrt{(\alpha + r_x^2)(\alpha + r_y^2)(\alpha + r_z^2)}} d\alpha.
$$
 (1)

Rearranging it, results

<span id="page-0-0"></span>
$$
\phi = -\frac{r_x r_y r_z}{2} x (L_{r_x}(\xi) - L_{r_x}(\infty)) M_x.
$$
\n(2)

The magnetic field at position ξ outside the particle results equal to the negative gradient of the magnetic potential. The components of the magnetic field due to the magnetization along the *x*−direction results

$$
H_x = \frac{r_x r_y r_z}{2} (L_{r_x}(\xi) - L_{r_x}(\infty)) M_x + \frac{r_x r_y r_z}{2} x \frac{\partial L_{r_x}(\xi)}{\partial x} M_x
$$
 (3)

$$
H_{y} = \frac{r_{x}r_{y}r_{z}}{2}x\frac{\partial L_{r_{x}}(\xi)}{\partial y}M_{x}, \qquad (4)
$$

$$
H_z = \frac{r_x r_y r_z}{2} x \frac{\partial L_{r_x}(\xi)}{\partial z} M_x. \tag{5}
$$

Similarly, the magnetic potentials due to the magnetization along the *y* and *z* particle axes result in a similar functionality to Eq. [\(2\)](#page-0-0). The magnetic field components due to the magnetization along the *y*−direction results

$$
H_x = \frac{r_x r_y r_z}{2} y \frac{\partial L_{r_y}(\xi)}{\partial x} M_y, \tag{6}
$$

$$
H_{y} = \frac{r_{x}r_{y}r_{z}}{2}(L_{r_{y}}(\xi) - L_{r_{y}}(\infty))M_{y} + \frac{r_{x}r_{y}r_{z}}{2}y\frac{\partial L_{r_{y}}(\xi)}{\partial y}M_{y},
$$
\n(7)

$$
H_z = \frac{r_x r_y r_z}{2} y \frac{\partial L_{r_y}(\xi)}{\partial z} M_y. \tag{8}
$$

The magnetic field components due to the magnetization along the *z*−direction results,

$$
H_x = \frac{r_x r_y r_z}{2} z \frac{\partial L_{r_z}(\xi)}{\partial x} M_z, \tag{9}
$$

$$
H_{y} = \frac{r_{x}r_{y}r_{z}}{2}z\frac{\partial L_{r_{z}}(\xi)}{\partial y}M_{z},
$$
\n(10)

$$
H_z = \frac{r_x r_y r_z}{2} M_z \left( L_{r_z}(\xi) - L_{r_z}(\infty) \right) + \frac{r_x r_y r_z}{2} z \frac{\partial L_{r_z}(\xi)}{\partial z} M_z.
$$
 (11)

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Therefore, the external field to the particle due to a magnetization  $\mathbf{M} = (M_x, M_y, M_z)$  in an arbitrary direction in the particle frame results

$$
H_{x} = \frac{r_{x}r_{y}r_{z}}{2} \left( [L_{r_{x}}(\xi) - L_{r_{x}}(\infty)] + x \frac{\partial L_{r_{x}}(\xi)}{\partial x} \right) M_{x}
$$
  
+  $\frac{r_{x}r_{y}r_{z}}{2} \left( y \frac{\partial L_{r_{y}}(\xi)}{\partial x} \right) M_{y}$   
+  $\frac{r_{x}r_{y}r_{z}}{2} \left( z \frac{\partial L_{r_{z}}(\xi)}{\partial x} \right) M_{z},$   

$$
H_{y} = \frac{r_{x}r_{y}r_{z}}{2} \left( x \frac{\partial L_{r_{x}}(\xi)}{\partial y} \right) M_{x}
$$
  
+  $\frac{r_{x}r_{y}r_{z}}{2} \left( [L_{r_{y}}(\xi) - L_{r_{y}}(\infty)] + y \frac{\partial L_{r_{y}}(\xi)}{\partial y} \right) M_{y}$   
+  $\frac{r_{x}r_{y}r_{z}}{2} \left( z \frac{\partial L_{r_{z}}(\xi)}{\partial y} \right) M_{z},$   

$$
H_{z} = \frac{r_{x}r_{y}r_{z}}{2} \left( x \frac{\partial L_{r_{x}}(\xi)}{\partial z} \right) M_{x}
$$
  
+  $\frac{r_{x}r_{y}r_{z}}{2} \left( y \frac{\partial L_{r_{y}}(\xi)}{\partial z} \right) M_{y}$   
+  $\frac{r_{x}r_{y}r_{z}}{2} \left( y \frac{\partial L_{r_{y}}(\xi)}{\partial z} \right) M_{y}$   
+  $\frac{r_{x}r_{y}r_{z}}{2} \left( [L_{r_{z}}(\xi) - L_{r_{z}}(\infty)] + z \frac{\partial L_{r_{z}}(\xi)}{\partial z} \right) M_{z}.$  (14)

Thus, the field outside the ellipsoidal particle with an arbitrarily oriented magnetization M can be expressed as

$$
\mathbf{H} = \frac{r_x r_y r_z}{2} \mathcal{G} \cdot \mathbf{M},\tag{15}
$$

where  $\mathscr G$  is a tensor, which is equivalent to the Green tensor in ellipsoidal coordinates with components

$$
\mathscr{G}_{ij} = \delta_{ij} \left[ L_{r_j}(\xi) - L_{r_j}(\infty) \right] + x_j \frac{\partial L_{r_j}(\xi)}{\partial x_i},\tag{16}
$$

where  $\delta_{ij}$  represents the identity tensor and  $L_{r_j}$  is a scalar function. Additionally,  $\frac{\partial L_{r_j}(\xi)}{\partial x_i}$  $\frac{\partial \partial x_i}{\partial x_i} = \frac{\partial L_{r_j}(\xi)}{\partial \xi}$ ∂ ξ  $\frac{\partial \xi}{\partial x_i}$ , and

$$
\frac{\partial \xi}{\partial x_i} = \frac{2x_i}{(r_i^2 + \xi)} / \left( \frac{x^2}{(r_x^2 + \xi)^2} + \frac{y^2}{(r_y^2 + \xi)^2} + \frac{z^2}{(r_z^2 + \xi)^2} \right),\tag{17}
$$

$$
\frac{\partial L_{r_j}(\xi)}{\partial \xi} = F_{r_j}(\xi). \tag{18}
$$



Fig. S 1 Excluded volume of (a)-(f) monodisperse ellipsoids and (g)-(l) binary ellipsoid-sphere systems with different orientations of particle J.

## Notes and references

<span id="page-2-0"></span>1 J. A. Stratton, *Electromagnetic Theory*, McGraw-Hill Book Company, Inc., New York, 1941, p. 615.



Fig. S 2 Dipolar interaction energy using point-dipole model between two magnetized ellipsoids (β*<sup>s</sup>* = 10) with different orientations and aspect ratios, (a1)-(g1) *rx*/*r<sup>m</sup>* = 1, (a2) - (g2) 5, and (a3) - (g3) 5 and 1. In both particles, the magnetization is aligned along the *x*−axis of the particle. The white area represents the excluded volume between particles at the corresponding orientation.



Fig. S 3 Dipolar interaction energy between two magnetized spheres  $r_x/r_m = 1$  as a function of dipole-dipole interaction parameter  $β_s$  and dipole-field interaction parameter  $α_s$ . In both particles, the magnetization is aligned along the *x−*axis of the particle. The field is along the *x*−axis in the laboratory space.



Fig. S 4 Normalized probability between two magnetized spheres *rx*/*r<sup>m</sup>* = 1 as a function of dipole-dipole interaction parameter β*<sup>s</sup>* and dipole-field interaction parameter α*<sup>s</sup>* . In both particles, the magnetization is aligned along the *x*−axis of the particle. The field is along the *x*−axis in the laboratory space.



Fig. S 5 Dipolar interaction energy between two magnetized ellipsoids  $r_x/r_m = 5$  as a function of dipole-dipole interaction parameter  $β_s$  and dipole-field interaction parameter  $α_s$ . In both particles, the magnetization is aligned along the *x−*axis of the particle. The field is along the *x*−axis in the laboratory space.



Fig. S 6 Normalized probability between two magnetized ellipsoids *rx*/*r<sup>m</sup>* = 5 as a function of dipole-dipole interaction parameter β*<sup>s</sup>* and dipole-field interaction parameter α*<sup>s</sup>* . In both particles, the magnetization is aligned along the *x*−axis of the particle. The field is along the *x*−axis in the laboratory space.



Fig. S 7 Dipolar interaction energy between a magnetized ellipsoid  $r_x/r_m = 5$  and a magnetized sphere  $r_x/r_m = 1$  as a function of dipole-dipole interaction parameter  $\beta_s$  and dipole-field interaction parameter  $\alpha_s$ . In both particles, the magnetization is aligned along the *x*−axis of the particle. The field is along the *x*−axis in the laboratory space.



Fig. S 8 Normalized probability between a magnetized ellipsoid *rx*/*r<sup>m</sup>* = 5 and a magnetized sphere *rx*/*r<sup>m</sup>* = 1 as a function of dipole-dipole interaction parameter  $\beta_s$  and dipole-field interaction parameter  $\alpha_s$ . In both particles, the magnetization is aligned along the *x*−axis of the particle. The field is along the *x*−axis in the laboratory space.



Fig. S 9 Snapshots of MC simulations for suspensions composed of spheres (*rx*/*r<sup>m</sup>* = 1) in a two-dimensional confinement as a function of  $\alpha_{\text{s}}$  and  $\beta_{\text{s}}$ . The colorbar is as shown in Fig. 3.



Fig. S 10 Pair distribution function for suspensions composed of spheres (*rx*/*r<sup>m</sup>* = 1) in a two-dimensional confinement as a function of α*<sup>s</sup>* and β*<sup>s</sup>* ..



Fig. S 11 Snapshots of MC simulations for suspensions composed of ellipsoids (*rx*/*r<sup>m</sup>* = 5) in a two-dimensional confinement as a function of  $\alpha_{\text{s}}$  and  $\beta_{\text{s}}$ . The colorbar is as shown in Fig. 3.



Fig. S 12 Pair distribution function for suspensions composed of ellipsoids (*rx*/*r<sup>m</sup>* = 5) in a two-dimensional confinement as a function of  $\alpha_{\text{\tiny S}}$  and  $\beta_{\text{\tiny S}}$ ..

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Fig. S 13 Snapshots of MC simulations for suspensions composed of a mixture of spheres (*rx*/*r<sup>m</sup>* = 1) and ellipsoids (*rx*/*r<sup>m</sup>* = 5) in a two-dimensional confinement as a function of  $\alpha_{\scriptscriptstyle \! S}$  and  $\beta_{\scriptscriptstyle \! S}$ . The colorbar is as shown in Fig. 3.



Fig. S 14 Pair distribution function for suspensions composed of a mixture of spheres  $(r_x/r_m = 1)$  and ellipsoids  $(r_x/r_m = 5)$  in a two-dimensional confinement as a function of α*<sup>s</sup>* and β*<sup>s</sup>* .