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 ($\xi > 0$) generated by the magnetization along the *x*-axis of the particle results¹

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

Rearranging it, results

$$\phi = -\frac{r_x r_y r_z}{2} x(L_{r_x}(\xi) - L_{r_x}(\infty)) M_x.$$
⁽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

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

$$H_y = \frac{r_x r_y r_z}{2} x \frac{\partial L_{r_x}(\xi)}{\partial y} M_x, \qquad (4)$$

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

Similarly, the magnetic potentials due to the magnetization along the y and z particle axes result in a similar functionality to Eq. (2). The magnetic field components due to the magnetization along the y-direction results

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

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

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

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

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

$$H_y = \frac{r_x r_y r_z}{2} z \frac{\partial L_{r_z}(\xi)}{\partial y} M_z, \qquad (10)$$

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

$$H_{x} = \frac{r_{x}r_{y}r_{z}}{2} \left(\left[L_{r_{x}}(\xi) - L_{r_{x}}(\infty) \right] + x \frac{\partial L_{r_{x}}(\xi)}{\partial x} \right) M_{x} + \frac{r_{x}r_{y}r_{z}}{2} \left(y \frac{\partial L_{r_{y}}(\xi)}{\partial x} \right) M_{y} + \frac{r_{x}r_{y}r_{z}}{2} \left(z \frac{\partial L_{r_{z}}(\xi)}{\partial x} \right) M_{z}, \qquad (12)$$

$$H_{y} = \frac{r_{x}r_{y}r_{z}}{2} \left(x \frac{\partial L_{r_{x}}(\xi)}{\partial y} \right) M_{x} + \frac{r_{x}r_{y}r_{z}}{2} \left(\left[L_{r_{y}}(\xi) - L_{r_{y}}(\infty) \right] + y \frac{\partial L_{r_{y}}(\xi)}{\partial y} \right) M_{y} + \frac{r_{x}r_{y}r_{z}}{2} \left(z \frac{\partial L_{r_{z}}(\xi)}{\partial y} \right) M_{z}, \qquad (13)$$

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

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

$$\mathbf{H} = \frac{r_{x}r_{y}r_{z}}{2}\mathscr{G}\cdot\mathbf{M},\tag{15}$$

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

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

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

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 ($\beta_s = 10$) with different orientations and aspect ratios, (a1)-(g1) $r_x/r_m = 1$, (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 $r_x/r_m = 1$ 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 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 $r_x/r_m = 5$ 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 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 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 8 Normalized probability 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 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 9 Snapshots of MC simulations for suspensions composed of spheres $(r_x/r_m = 1)$ in a two-dimensional confinement as a function of α_s and β_s . The colorbar is as shown in Fig. 3.



Fig. S 10 Pair distribution function for suspensions composed of spheres $(r_x/r_m = 1)$ in a two-dimensional confinement as a function of α_s and β_s ..



Fig. S 11 Snapshots of MC simulations for suspensions composed of ellipsoids $(r_x/r_m = 5)$ in a two-dimensional confinement as a function of α_s and β_s . The colorbar is as shown in Fig. 3.



Fig. S 12 Pair distribution function for suspensions composed of ellipsoids $(r_x/r_m = 5)$ in a two-dimensional confinement as a function of α_s and β_s ..



Fig. S 13 Snapshots of MC simulations 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_s and β_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_s and β_s .