A Two-Parameter Model for Colloidal Particles with an Extended Magnetic Cap

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Self-assembly of magnetic colloidal particles in solution has successfully been simulated by hard- or soft-sphere models with a set of embedded magnetic point dipoles, and the position and orientation of each dipole are adapted to mimic the magnetization distribution. Herein, a conceptually simpler approach is introduced for magnetically capped colloidal particles, which replaces the set of dipoles by the extended magnetization distribution of a single conductive loop. Only two parameters are required to characterize the magnetization distribution: the diameter of the loop and its radial off-center shift within the sphere. This approach reflects the radial symmetry and the spatial extension of the magnetic cap. At larger distance and in the limit of very small loops that model reproduces the far field and particle arrangements obtained with the single, radially shifted dipole model. For larger loop radii, additional stable assembly patterns are obtained, which occur in experiments, but cannot be simulated with a single shifted dipole model.

1. Introduction

Magnetic colloidal particle suspensions have been used to study a wealth of physical phenomena which span from fundamental aspects to application-oriented topics, e.g., in medical science and imaging,[3–5] in microfluidic sensing,[6] or in environmental engineering.[7,8] Recent examples address directed motion in anisotropic media or along chemical, thermal, and magnetic field gradients,[9] the pattern formation under static and dynamic external field conditions[10–13] with applied shear forces,[14,15] or in conjunction with magnetic frustration,[16] as well as applications as bit-patterned magnetic media,[17] ferrofluids,[18] magnetically directed or actuated swimmers[4,19] or microscale motors.[20] Both ferromagnetic particles with a permanent magnetic moment and (super)paramagnetic particles have been studied.[4,21] Complex, even chiral, structures have been obtained from simple, structurally isotropic building blocks,[22] especially for 2D assemblies in the presence of external magnetic fields.[13,23–26] More elaborate anisotropies of the assembly pattern have been introduced via the particle shape,[27] by an inhomogeneous distribution of the magnetic phase within the particle or on its surface[12,24,28] or by both effects combined.[29]

Here, we focus on modeling the structurally very simple case of spherical particles, which become magnetically anisotropic when they are coated with a magnetic metal layer on one hemisphere. Micrometer-sized silica spheres capped with a thin, ferromagnetic Co/Pd multilayer are a specific, experimentally well-studied model system.[5,23,30,31] This special variant of so-called Janus particles exhibits a permanent magnetization which points perpendicular to the cap surface. Thus, the magnetization is rotationally symmetric and extends over the whole cap and is ferromagnetically ordered perpendicular to most of the cap surface.[32] Video microscopy on particle dimers indicates that in the far field, the dipole component parallel to the rotation axis prevails, whereas in the near field, the curved magnetization distribution leads to a canted antiparallel arrangement.[12]

The desired magnetic properties of the multilayer cap follow from shape anisotropy and exchange interaction between ferromagnetic Co layers with a thickness of less than 60 nm, which are separated by paramagnetic Pd layers of 0.27 nm. When deposited on small particles up to 300 nm, a single-domain out-of-plane magnetization is obtained.[16,30,32,33]

Caps on larger particles become single domains if they are magnetically saturated in an external field after film deposition.[16] The present model refers to magnetic caps with Co and Pd that provide strong perpendicular magnetization anisotropy. In suspension of such capped particles, the change in magnetization in the presence of nearby particles or under external fields in the same order of magnitude is negligible.[23] Therefore, for the present discussion, the microscale contributions of the single spins or the single magnetic layers can be integrated to form a
more coarse-grained radially symmetric, time-invariant magnetization distribution on the mesoscopic scale.

Detailed data have been recorded for the interaction of single, magnetically activated particles with the surrounding aqueous medium and for the interparticle interactions in 2D aggregates. In the absence of external fields, such aggregates are composed of distinct structural motifs, which result from the interplay between the favorable trigonal dense packing of spheres and the spatially more complex magnetic interactions between the extended ferromagnetic caps. From those observations, one may conclude that depending on the size and composition of the magnetic cap, different flux-closure structures are formed, which minimize the far field and hence the magnetic energy of the assembly. Systematic, continuous variations of the cap parameters required to test this assumption are not straightforward experimentally. The present analytical approach captures the salient geometric features of magnetically capped spherical colloidal particles and allows studying their interaction in a dissipative medium in dependence on the cap parameters.

Theoretical models of highly symmetric geometric objects such as spheres or cubes with an embedded magnetic point dipole have proven to be simple but suitable models to predict and reproduce the experimentally observed structures of homo- geneously magnetized dipolar particles. This approach can be extended to approximate the total stray field of particles with an anisotropic magnetization distribution by shifting the point dipole off-center (shifted dipole or sd model) radially or laterally. For capped colloidal particles, the anisotropy introduced by the extended, asymmetric distribution of the magnetic material leads to a complex assembly behavior, and some observed self-assembled structures have been reproduced by simulations with such models.

Despite the amazing success of such simple sd models, there have been experiments with Janus particles in which the overall shape and the relative interior cap orientations of the assembly deviated from predictions with the sd model. In such cases, the spatially extended form of the magnetization distribution is not captured well by the point dipole picture. Recent models consider this by assuming multipolar interactions and apply higher-order moments, a planar distribution of three point dipoles in the absence of out-of-plane external field components, or 3D arrangements of five dipoles, to model the radial symmetry of magnetization distribution better. As a consequence, such models become more complex and require an increased number of parameters as well as an increasing computational effort in comparison with the single sd model. Therefore, we have developed an alternative approach which covers near- and far-field interactions of Janus particles with only two parameters.

2. Model

Here, we introduce a new model for capped magnetic particles with an anisotropic, broad, but radially symmetric magnetization distribution. As shown in Figure 1, this model consists of a sphere with an enclosed conductive loop, which exhibits an ideal ring current. The magnetic far field of that current reproduces the stray field of a point dipole model, whereas the near field reflects the spatial extension of the magnetization distribution.

Such a model has the great advantage that it addresses ideally dipolar particles and colloids with an extended and shifted magnetization within the same conceptual framework and by the same set of only two parameters. This provides the possibility to analyze the pattern evolution as a function of the cap shape in a continuous and consistent way and correlate observed assembly patterns with the geometric parameters of the magnetization distribution. It furthermore provides the possibility to study pattern changes as a function of the cap shape in a continuous way. That is neither possible experimentally, because the cap size is fixed, nor is it accessible by the multiple shifted dipole models, because adding dipoles to mimic the extended magnetization is a discrete process. However, it is the pathway to designing cap shapes for specific applications.

The model particles are simulated as hard spheres, and the current loop is fixed inside. The size of the loop controls the width of the magnetization distribution. The position of the loop inside the sphere, , determines the degree of magnetic asymmetry. As shown in Figure 1, the model’s parameters are as follows.

The sphere is centered at and has the radius . The particle orientation is denoted by with the in-plane and out-of-plane orientations given by the angles and with respect to and directions, respectively. The plane spanned by the loop is oriented normal to , and its center is radially displaced by a shift from along . The loop itself is parameterized in polar coordinates by its radius and by the angle of the loop plane. The loop is further assigned a constant current , and one circulation direction is chosen for all particles. The reference frame of the loop is thus uniquely defined by the position of the loop center at and the radial vector , which is the normal vector of the loop plane. Together with the discretization of the loop in sections , it follows for the magnetic induction at an arbitrary point

\[
B(x) = \frac{\mu_0 I u \sum_{j=0}^{p-1} e_{\phi_j} \times (x - x'_{\phi_j})}{2p |x - x'_{\phi_j}|^3}
\]
where the considered points are located in a particle \( A \) at \( x'_\varphi \) with the current \( I \) oriented in the direction of \( e_\varphi \). The interaction between particle \( A \) and another particle \( B \) is described by the Lorentz force \( F_L \)

\[
F_L = \frac{2\pi I u}{p} \sum_{i=0}^{p-1} (e_\varphi \times B(x'_\varphi))
\]  

(2)

induced by the influence of the magnetic stray field exerted by \( A \) on the electric current in \( B \), and the resulting torque \( M_L \)

\[
M_L = \frac{2\pi I u}{p} \sum_{i=0}^{p-1} (e_\varphi \times B(x'_\varphi)) \times (x'_\varphi - x_0)
\]  

(3)

on particle \( B \). Here, in analogy to particle \( A \), the loop in \( B \) is parameterized by \( \varphi_i \), resulting in the points \( x'_\varphi \) and the direction \( e_\varphi \) of the current inside \( B \). Based on the parameters of the experimental reference systems;\(^4\) in particular, the low Reynolds number of the colloidal suspensions studied there, the particles’ inertia is negligible. As a consequence, the magnetic force and torque lead to an instantaneous change in lateral speed \( v = \lambda_i^{-1} F \) and angular speed \( \omega = \lambda_i^{-1} M \). Consequently, the system dynamics can be described as

\[
x_0(t + \Delta t) = x_0(t) + v\Delta t
\]  

(4)

\[
r_1(t + \Delta t) = R(\omega\Delta t)r_1(t)
\]  

(5)

where \( R(\omega\Delta t) \) is the rotation matrix. The length of a time step \( \Delta t \) is determined adaptively from the maximum set increment of translation and rotation with \( \Delta x_{\text{max}} = 0.005 R \) and \( \Delta \theta_{\text{max}} = 0.005 \), respectively.

\[
\Delta t = \min\left(\frac{\Delta x_{\text{max}}}{v}, \frac{\Delta \theta_{\text{max}}}{\omega}\right)
\]  

(6)

We define that two interacting particles have reached a stable configuration if the change of \( x_0 \) and \( r_1 \) over multiple time steps is below a convergence threshold \( \epsilon \) for both particles.

### 3. Results

As a first step, we studied the influence that the number of simulated points \( p \) of the loop exhibits on the rotational symmetry of the resulting stray field around \( r_1 \). The goal is to produce a field with high rotational symmetry despite mandatory discretization, to retain the advantage provided by a loop over single dipoles, without bloating computing times.

We examined the magnetic stray field of a particle with the loop parameters \( v = 0.4R \) and \( u = 0.4R \), for which the orientation angle \( \alpha = \frac{\pi}{4} \) amounts to the average of its extreme values. To obtain a reference field \( B_0 \), we determine the magnetic field at the intersection of \( r_1 \) with the sphere. The magnetic field at the sphere surface is evaluated along the radial projection of the loop along the vector \( r_1 \). This projection line is a ring with a radius \( r_{\text{ring}} = 0.7R \), centered around the loop vector \( r_1 \) and parallel to the loop itself. We calculate the difference \( \Delta_{\text{max/min}} \) between the maximum \( B_{\text{max}} \) (close to loop elements) and minimum \( B_{\text{min}} \) (between two loop elements) of the magnetic field along the ring in units of the normalized field \( B_0 \). The values of \( \Delta_{\text{max/min}} \) for different numbers of discretization points \( p \) are shown in Table 1.

| \( p \) | \( \Delta_{\text{max/min}} [B_0] \) |
|---|---|
| 4 | 0.51421 |
| 8 | 0.03096 |
| 12 | 0.00156 |
| 24 | <10^{-6} |

A small value of \( p = 4 \) produces pronounced variations along the projection line, which may be as large as 0.51\( B_0 \). Already \( p = 8 \) leads to minor deviations from the rotational symmetry, and with \( p = 12 \), the field is smooth to the per mille range. From this result, one may conjecture that multiple sd models will require about ten side dipoles to represent a similarly symmetric magnetization distribution. For \( p = 24 \) points, the difference is six orders of magnitude smaller than \( B_0 \) and thus sufficiently small to be negligible. Thus, for the present study, we will be using \( p = 24 \) for the simulated particles to investigate their interaction.

Next, we studied a system consisting of two Janus particles \( A \) and \( B \) and investigated their mutual arrangement in dependence on the loop parameters \( v \) and \( u \) in the absence of external fields. Both particles have the same loop parameters but differ in position \( x_0 \) and orientation \( r_1 \). In line with literature data from experimental and theoretical studies on 2D-confined assemblies of colloidal particles with a ferromagnetic cap, we may assume that the centers \( x_0 \), \( A \) and \( x_0 \), \( B \) of both spheres and both loop vectors \( r_{1,A} \) and \( r_{1,B} \) are located within one plane. In the present discussion, we arbitrarily assign the \( x-\gamma \) plane.

As shown in Figure 2, the relative arrangement of the particles \( A \) and \( B \) is defined by the connection line \( d_{AB} \) between the particle centers, the relative angle \( \gamma \) between the loop vectors, and the angles \( \delta_A \) and \( \delta_B \) of the loop vectors with respect to the connection line \( d_{AB} \). If an angle \( \delta = 0 \), then the cap of the given particle points exactly along the connection line \( d \) toward a neighboring particle.

**Figure 2.** Characteristic geometric parameters for the in-plane arrangement of two particles \( A \) and \( B \). The scale bar is 2 \( \mu \)m.

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\( \text{Table 1. } \) Values for \( \Delta_{\text{max/min}} \) for different numbers of discretization points \( p \).

| \( p \) | \( \Delta_{\text{max/min}} [B_0] \) |
|---|---|
| 4 | 0.51421 |
| 8 | 0.03096 |
| 12 | 0.00156 |
| 24 | <10^{-6} |
particle. This convention is chosen, because it facilitates the upscaling to larger clusters. In the presently studied field-free two-particle case, \( \delta_A \) and \( \delta_B \) amount to about \( \frac{\pi}{8} \) in the experiment. For two particles, the \( \delta \) angles can be combined to the angle \( \gamma \) between the loop vectors, as shown in sphere A, in Figure 2. A value of \( \gamma = 0 \) corresponds to the parallel arrangement of the loop vectors; for \( \gamma = \pi \), the two loop vectors are antiparallel.

Figure 3 shows which values the angles \( \gamma \), \( \delta_A \), and \( \delta_B \) obtain in the stable two-particle configuration for given loop parameters \((\nu,u)\). The inset shows the experimentally observed antiparallel dimer structure of such Janus particles, which exhibits values of \( \delta_A \) and \( \delta_B \) of about \( \frac{\pi}{8} \).

Figure 3c shows the \( \gamma-\nu-u \) diagram, which visualizes the relative arrangements which we obtained for two particles in contact \((\delta_{AB} = 2R)\) and in the field-free case. The diagram correlates the angle \( \gamma \) between the loop vectors \( r_{1,A} \) and \( r_{1,B} \) and the loop parameters \( \nu \) and \( u \). Following the definitions shown in Figure 2 for \( \gamma = 0 \), i.e., the parallel arrangement of the loop vectors and the corresponding \((\nu,u)\) pairs are indicated in black in the \( \gamma-\nu-u \) diagram. For a wide range of antiparallel arrangements with \( \gamma = \pi \), the two loop vectors are shown in yellow in the \( \gamma-\nu-u \) diagram. The transition between those two extremes via intermediate angles is visualized by the other colors of the scale.

We found three distinct stable arrangement patterns, which can be assigned to different closed parameter areas in the \( \gamma-\nu-u \) diagram. Parallel arrangements are obtained in an elliptical region for low loop radii \( u \) and shifts \( v \), antiparallel ones if the geometric mean of the two loop parameters is larger than about 0.5, and intermediate ones once the two parameters assume low-to-medium values. For small loops, i.e., along the \( v \) axis of the diagram, all possible angles of \( \gamma \) are obtained with increasing shift \( v \). Figure 4 shows the sequence of the relative particle arrangements: for smaller loops with an equally low shift, the system defaults to the parallel orientation \((\gamma = 0)\) along \( d_{AB} \). With increasing shift values, such particles undergo a transition to a zone with a continuous change of the angle \( \gamma \), for large shifts, they prefer an antiparallel orientation \((\gamma = \pi)\). Along the \( u \) axis, i.e., for very low shifts and increasing loop radius, the change from the parallel to the antiparallel arrangement is sudden, and no zone with intermediate angles is found. As shown in Figure 5, the antiparallel arrangement for large radii shows point symmetry in the point of contact \((\delta_A = \delta_B = \delta)\), and the angles \( \delta \) decrease upon transition to smaller loops and larger shifts (Figure 3a,b). In parts of the transition area, we can observe the third arrangement pattern, which is nonsymmetrical and shows sharp transition lines between the two other patterns.

A comparison for different \((\nu,u)\) parameter values within the area of the parallel arrangement (black) shows no variation of the actual arrangements of the particles \((\delta_A = 0, \delta_B = \pi)\). Thus, the parallel arrangement is unique and does not depend on the specific parameter values. This finding suggests that in the experiment, magnetic colloids tolerate quite large deviations from the ideal central dipole location and still exhibit homogeneously parallel ordering. Both of the other arrangements, on the contrary, exhibit a dependence on the \((\nu,u)\) parameter combination. For the antiparallel arrangement, we observe a sudden change of the angle \( \delta \) between \( r_1 \) and \( d_{AB} \) as shown in Figure 5. Large loops \( u \approx R \) close to the center of the sphere \( v = 0 \) show an almost perpendicular orientation \( \delta \approx \pi/2 \) to the direct connection between both particle centers, \( d_{AB} \); the respective parameter pairs...
correspond to the top-left corner of Figure 3. Moving along the parameter area for the antiparallel arrangement, the loops get smaller and their shift increases. Along that path, we observe that the angles $\delta_A$ and $\delta_B$ between the loop vectors and the connection line shrink, which means that the loops start orienting themselves closer to $d_{AB}$. At the end of that path through the parameter space, i.e., for tiny loops $u \approx 0$ and large shifts $v \approx R$ (bottom right of Figure 3), the loop vectors nearly align to $d_{AB}$. We therefore conclude that experimentally antiparallel arrangements can be realized if the magnetic center is shifted significantly off-center and that the relative arrangement of the loop vectors with respect to the geometric connection line very sensitively depends on the spatial extension of magnetization distribution.

Finally, we want to elucidate whether the two-particle arrangements obtained with the shifted loop model exhibit similarities with analytical and numerical data from earlier simulations of the two-particle system with sd models. As the far field of a small loop resembles the one of a single magnetic dipole, we focus on the transition points between stable arrangement patterns in dependence on the shift $v$ for a negligible loop radius ($u \rightarrow 0$). Comparing with the findings of Kantorovich et al., particles with a single shifted dipole \cite{18} revealed the same configurations for two particles. The values of the shift $v$ for the transitions between the three different arrangements are shown in Table 2. In addition to the shifted dipoles, we investigated the values for three different sets of particles that started either with a parallel or one of two antiparallel arrangements and differ in their respective $\delta$ values ($\delta = \frac{\pi}{4}$ and $\delta \approx \frac{3}{4}$). The first transition between the parallel arrangement and the nonparallel one is obtained from these starting arrangements. For the shifted loop model, the values of $v$ for that transition lie within an interval of $\pm 0.01R$ around the value for the shifted dipole $v_{sd} = 0.408R$ \cite{18} with $v$ being smaller for the antiparallel ($\delta = \frac{3}{4}$) and larger for the parallel arrangement. The existence of this asymmetric state and its apparent dependence on the starting conditions is a sign pointing toward bistability, which can mainly be observed in larger systems. For the second transition between the noncollinear and the antiparallel arrangements, we obtain a slightly less good agreement between the shifts. Generally, the shifted loop delivers slightly larger values $v = 0.619R$ and $v = 0.606R$ compared with $v_{sd} = 0.597R$ even while starting close to the anticipated arrangement ($\delta \approx \frac{3}{4}$). A possible reason is the fact that even a small loop compared with the radius of the particle does not equal a single point, with increased shift and the lower distance between the loops in the antiparallel arrangements even less so. In that regime, both models describe a gradual transition toward an antiparallel cap arrangement with $\delta_A = \delta_B = 0$. A quantitative difference occurs in those values which $\delta_A$ and $\delta_B$ exhibit in the vicinity of the boundary between the noncollinear and the antiparallel cap arrangement. There, the shifted dipole model suggests one specific value of $\delta \approx \frac{\pi}{4}$, whereas in Figure 3, the present loop model indicates that the angles $\delta$ span a wider range, which starts from about $\frac{\pi}{4}$ for the smallest loop diameters and increases to $\pi$ with growing radii of the loop. In comparison, the experimental value amounted to $\frac{\pi}{8}$, which is not in the range of the shifted dipole particles, but may be obtained with the present loop model if the loop is extended.

**Table 2.** Transitions between different arrangements for a small loop ($u \rightarrow 0$) because of shift $v$. Shown for parallel and two different antiparallel arrangements as starting points, with single shifted dipole model \cite{18} for reference. Shift $v$ is expressed in units of the particle radius $R$. $v \in [0, 1]$.

| Arrangement | First transition | Second transition |
|-------------|------------------|-------------------|
| Parallel →→ | $v = 0.417$      | $v = 0.619$       |
| Antiparallel | $v = 0.398$      | $v = 0.619$       |
| Antiparallel | $v = 0.415$      | $v = 0.606$       |
| Shifted dipole | $v_{sd} = 0.408$ | $v_{sd} = 0.597$ |

**4. Conclusions**

We used an ideally conductive loop with a constant current inside a hard sphere to simulate magnetic Janus particles and study the interaction between two such particles. The loop is described by
its two parameters, the radius $u$ and shift $v$, and provides a sufficiently homogeneous magnetic field to be close to one produced by the magnetic cap of the real particles.[30] We were able to model the parallel and antiparallel assembly patterns known from experiments on two particles in dependence on the chosen parameter values. For a small shifted loop, we obtain analogous points of transition between those arrangements as observed earlier for simulations with shifted dipoles.[18] With the present model, we find an additional transition to an antiparallel arrangement of the particles with increasing radius $u$, which reflects the extension of the cap. With increasing value of $u$, the relative angle $\delta$ between the cap vectors $r_1$ and the connection line $d_{AB}$ of the two particles increases until both cap vectors are perpendicular to $d_{AB}$ and antiparallel to each other at $v = 0$ and $u = 1$. For large shift values $v$, increasing the radius $u$ leads to a gradual transition from an antiparallel arrangement with both cap vectors along $d_{AB}$ to another antiparallel one, in which the cap vectors are perpendicular to $d_{AB}$. For small shift values $v$, this leads to a transition zone between the parallel, linear chain at small radius $u$ and the antiparallel, perpendicular arrangement at large $u$. In these zones, the system is bistable, i.e., the starting conditions determine which of the two adjacent states is chosen upon force minimization. In conclusion, the present particle model yields results comparable with experiments and exhibits more flexibility than standard models with only two parameters. As such, it provides a viable basis for further works with more complex systems.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

capped colloids, magnetic colloids, self-assemblies, simulations, theories

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