Numerical calculation of interaction forces between paramagnetic colloids in two-dimensional systems

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Typically the force between paramagnetic particles in a uniform magnetic field is described using the dipolar model, which is inaccurate when particles are in close proximity to each other. Instead, the exact force between paramagnetic particles can be determined by solving a three-dimensional Laplace's equation for magnetostatics under specified boundary conditions and calculating the Maxwell stress tensor. The analytical solution to this multi-boundary-condition Laplace's equation can be obtained by using a solid harmonics expansion in conjunction with the Hobson formula. However, for a multibody system, finite truncation of the Hobson formula does not lead to convergence of the expansion at all points, which makes the approximation physically unrealistic. Here we present a numerical method for solving this Laplace's equation for magnetostatics. This method uses a smoothed representation to replace all the boundary conditions. A two-step propagation is used to dramatically accelerate the calculation without losing accuracy. Using this method, we calculate the force between two paramagnetic particles in a uniform and a rotational external field and compare our results with other models. Furthermore, the many-body effects for three-particle, ten-particle, and 24-particle systems are examined using the same method. We also calculate the interaction between particles with different magnetic susceptibilities and particle diameters. The Laplace's equation solver method described in this article that is used to determine the force between paramagnetic particles is shown to be very useful for dynamic simulations for both two-particle systems and a large cluster of particles.

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I. INTRODUCTION

Paramagnetic colloidal suspensions have been utilized for a number of innovative applications in a variety of fields such as smart fluids [e.g., magnetorheological (MR) fluids] [1–3], biomedical imaging and sensing [4], unique directed assembly structures [5–7], self-propelled swimming structures [8,9], force probing [10,11], and models for statistical mechanics [12,13]. In many of these applications, the force between these paramagnetic particles needs to be accurately calculated. The accuracy is dependent on the model used [14,15], the physical properties of the particles, and the distance between particles [15].

Typically, the dipolar model (DM) has been used to calculate the force between paramagnetic particles placed in an external magnetic field [2,16]. Though this widely used model is simple to use, it noticeably starts to lose accuracy when considering systems where particles are close to each other due to negligence in including the mutual induction effect. The improved mutual dipolar model (MDM) takes into account the mutual induction, but it also fails to properly consider the multipolar effect [15]. The exact force can be calculated analytically by solving Laplace's equation for magnetostatics with multiple boundary conditions at the interfaces and then calculating the Maxwell stress tensor over the particle volume [15,17]. The solution to Laplace's equation can be analytically presented as a solid harmonics expansion, with the translation theorem applied to unify the coordinate system. This translation process is very computationally cumbersome, as a very large truncation number is required [18]. A numerical solution to the aforementioned Laplace's equation has been

difficult to obtain as well due to the difficulty in applying boundary conditions for different coordinate systems.

Recently, a smoothed profile method was used to simulate the flow field of colloidal suspensions with a hydrodynamic interaction [19], which largely simplifies the numerical calculation by replacing all the boundary conditions with a simple indicator function. This method was later used to simulate electrokinetic flow [20] and the flow of two-dimensional (2D) paramagnetic disks [17]. Here we utilize a smoothed profile method to develop a numerical solution to the three-dimensional (3D) Laplace's equation describing the magnetic field of a system that consists of superparamagnetic particles trapped in a 2D plane in an external magnetic field. 2D colloidal systems are widely used experimentally to study various thermodynamic phenomena such as melting [13] and phase behavior [21,22] since individual colloidal particles can be easily visualized using optical microscopy [23]. Therefore we focus on the 2D system, but this method can also be used for 3D systems. Additionally, we use a two-step method to significantly accelerate the solution. The force between two particles in a uniform field is calculated and compared with other models. Additionally, the forces between two particles in two-particle, three-particle, ten-particle, and 24-particle systems in a rotational field are also calculated and compared. Moreover, we calculate the force between particles with different susceptibilities and particle diameters to derive a force scaling law for these parameters.

II. METHODS

A. Dipolar model (DM) and mutual dipolar model (MDM)

Under a uniform magnetic field, a single spherical paramagnetic particle will be uniformly magnetized and acquire a

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magnetic dipole moment in the direction of the field with a magnitude

$$m = \frac{4\pi a^3 \chi_{\text{eff}} H_0}{3}, \quad (1)$$

where a is the particle radius, $\chi_{\text{eff}} = \frac{3\chi}{3+\chi}$ the effective volumetric susceptibility of the particle taking into account the demagnetization effect [2,15], χ the volumetric magnetic susceptibility of the material, and H_0 the applied magnetic field strength. The magnetic field around this particle is a dipolar field,

$$\mathbf{H}_{\text{dip}}(\mathbf{r}) = \frac{1}{4\pi} \left(\frac{3\mathbf{r}(\mathbf{m} \cdot \mathbf{r})}{r^5} - \frac{\mathbf{m}}{r^3} \right), \quad (2)$$

where $\mathbf{m}$ is the dipole moment, $\mathbf{r}$ the position of the point around the particle, and r the norm of $\mathbf{r}$. A single particle experiences no force in a uniform magnetic field [Fig. 1(a)]. If another particle is placed r distance away from this particle, the particles will have an attractive force for each other, which can be approximated by the dipolar model as

$$\begin{aligned} \mathbf{F}_1 &= -\nabla(\mathbf{m}_1 \cdot \mathbf{B}_2) \\ &= \frac{3\mu_0}{4\pi r^5} \left[(\mathbf{m}_1 \cdot \mathbf{r})\mathbf{m}_2 + (\mathbf{m}_2 \cdot \mathbf{r})\mathbf{m}_1 + (\mathbf{m}_1 \cdot \mathbf{m}_2)\mathbf{r} \right. \\ &\quad \left. - \frac{5(\mathbf{m}_1 \cdot \mathbf{r})(\mathbf{m}_2 \cdot \mathbf{r})}{r^2} \mathbf{r} \right], \end{aligned} \quad (3)$$

where $\mathbf{F}_1$ is the force exerted on particle 1, $\mathbf{m}_1$ the dipole moment for particle 1, $\mathbf{m}_2$ the dipole moment for particle 2, $\mathbf{B}_2$ the dipolar magnetic field generated by particle 2, μ_0 the vacuum permeability, and $\mathbf{r}$ the connector vector between the two particles.

The DM assumes that the dipole moments on two particles have no interaction with each other. Thus, the field surrounding the two particles is a superposition of the dipolar field from two uniformly magnetized spheres. This assumption affects the accuracy of the force calculation in two aspects. First, the surrounding magnetic field is not simply the superposition of two dipolar fields, but also includes quadrupole, octopole, and higher-order multipole terms [14,16]. Multipole terms are negligible only when the particles are far away from each other, which is usually the case for a dilute suspension. When the particles are sufficiently close, the particles are no longer uniformly magnetized, thus the magnetic field around the particles can no longer be presented as a dipolar field [Fig. 1(b)]. In this case, the force between particles calculated by Eq. (3) may deviate significantly from the actual force. Second, the dipole moments will not stay intact when two particles are close to each other. Instead they will have a mutual interaction with each other, which changes the magnitude as well as the direction of the dipoles. In this case, Eq. (1) may not accurately describe the dipole moment.

The second effect can be mitigated by adding a mutual interaction. For a suspension of several particles, the MDM [15] can be used [Eq. (4)]:

$$\mathbf{m}_n = \frac{4}{3}\pi a^3 \chi_{\text{eff}} \left(\mathbf{H}_0 + \sum_{k=1, k \neq n}^N \mathbf{H}_{\text{dip}}(\mathbf{r}_{kn}) \right), \quad (4)$$

FIG. 1. (Color online) Plot showing the calculation of the magnetic field strength around (a) one paramagnetic particle and (b) two paramagnetic particles in a uniform external magnetic field of 6 G in the x direction. The scales correspond to the value of the magnetic field in units of A/m. (c) Schematic of the two grids and parameters used in the numerical calculation of the magnetic field for a two-particle system.

where $\mathbf{H}_0$ is the external magnetic field and $\mathbf{r}_{kn}$ the connector vector from particle k to particle n . For two particles, an even simpler form can be used which self-consistently assumes the directions of the two dipoles are the same [12]. The first effect is caused by the dipole moment assumption and will not vanish unless multipole terms are added. This requires the solution for Maxwell's equations and calculation of the Maxwell stress tensor.

B. Laplace's equation and solution

Maxwell's equations require a zero divergence for the magnetic B field and a zero curl for the magnetic H field if there is no free current in the system [24]. In the perspective of a scalar magnetic potential, the above equations can be

simplified to Laplace's equation for different media [15,24],

$$\nabla^2 \phi_n = 0, \quad \nabla^2 \phi_{\text{DM}} = 0, \quad (5)$$

where ϕ_n is the scalar magnetic potential for particle $n = 1$ and 2, and ϕ_{DM} is the magnetic potential for the dispersion media. Here we only consider a two-body system for simplicity. The boundary conditions at the surfaces of the spheres $r_n = a$ are

$$\phi_n = \phi_{\text{DM}}, \quad \mu_p \frac{\partial \phi_n}{\partial r_n} = \mu_0 \frac{\partial \phi_{\text{DM}}}{\partial r_n}, \quad (6)$$

where $\mu_p = \mu_0(1 + \chi)$ is the particle permeability. The boundary condition at $r_n \rightarrow \infty$ is

$$\phi_{\text{DM}} = -\mathbf{H}_0 \cdot \mathbf{r}_n. \quad (7)$$

The analytical solution to the Laplace's equation above can be expressed in terms of a solid harmonics expansion [15,24],

$$\begin{aligned} \phi_n &= \sum_{l=0}^{\infty} \alpha_{nl} r_n^l P_l(\cos \theta_n), \\ \phi_{\text{DM}} &= -H_{\perp} x - H_{\parallel} y + \sum_{n=1}^2 \sum_{l=0}^{\infty} \beta_{nl} \frac{P_l(\cos \theta_n)}{r_n^{l+1}}, \end{aligned} \quad (8)$$

where (r_n, θ_n, φ) represents a spherical coordinate system with an origin at the center of particle n , $P_l(x)$ are associated Legendre polynomials, and $H_{\perp}$ and $H_{\parallel}$ are components of $\mathbf{H}_0$ in the x and y direction, respectively, depending on the spherical coordinate system. The φ term is neglected due to the azimuthal symmetry for the two-body problem. Applying boundary conditions requires a coordinate translation, which can be done using the translation theorem for solid harmonics [25] or the Hobson formula [15,26]. However, this approach is not able to give an acceptable approximation with finite truncation on the Legendre expansion [18].

As the analytical solution to Laplace's equation is difficult to obtain, we therefore adopt a numerical approach to solving Eqs. (5)–(7). Considering the efficiency of discretization as well as the accuracy of boundary condition propagation, we use the finite volume method with a cubic grid. The most difficult step is the application of the boundary conditions, since the boundary conditions in Eq. (6) need to be applied on a spherical coordinate system. The smoothed profile method largely simplifies this process by using an indicator function to replace all the boundary conditions at the interfaces [17,20]. The indicator function $\lambda_i(\mathbf{r})$ for the i th particle is

$$\lambda_i(\mathbf{r}) = \frac{1}{2} \left[\tanh \left(\frac{1 - |\mathbf{r} - \mathbf{r}_{c,i}|}{\xi} \right) + 1 \right], \quad (9)$$

where ξ is the interface thickness. An overall indicator function for the entire system is given by summing up the indicator functions over all particles,

$$\lambda(\mathbf{r}) = \sum_{i=1}^n \lambda_i(\mathbf{r}). \quad (10)$$

The discontinuous physical property in the equation is in the permeability μ , therefore the overall indicator function is used to smooth the profile of it:

$$\frac{1}{\mu(\mathbf{r})} = \frac{1}{\mu_p(\mathbf{r})} \lambda(\mathbf{r}) + \frac{1}{\mu_0(\mathbf{r})} [1 - \lambda(\mathbf{r})]. \quad (11)$$

By using the smoothed representation, the boundary conditions at the interfaces will be automatically satisfied. Thus all one has to do to solve Laplace's equations, given by Eq. (5), is to iteratively propagate the infinite boundary conditions from the boundary of the grid box.

C. Force calculation

The solution of Laplace's equation gives the distribution of the magnetic field in the grid box. The Maxwell stress tensor is then used to calculate the magnetic force with the known magnetic field distribution. First, the divergence of Maxwell stress tensor σ_m is used to calculate the force density [17]:

$$\mathbf{f}_m = \nabla \cdot \sigma_m = \nabla \cdot \left[\mu_0 (\mathbf{H}\mathbf{H} - \frac{1}{2} H^2 \mathbf{I}) \right]. \quad (12)$$

Here $\mathbf{H}\mathbf{H}$ is a second-order tensor, H is the norm of magnetic field vector $\mathbf{H}$ obtained from the solution to Laplace's equation, and $\mathbf{I}$ is the identity tensor. Then we sum the force density over the volume of a sphere to calculate its total magnetic force,

$$\mathbf{F} = \int_{\Omega_i} \mathbf{f}_m dV = \int_{\Omega_i} \nabla \cdot \sigma_m dV, \quad (13)$$

where Ω_i is the total volume of sphere i .

D. Numerical calculation acceleration

The accuracy of the numerical Laplace's equation solver is dependent on both the size of the grid box L and the number of grid points N in each dimension. Since the infinite boundary conditions are applied at the edge of the box, the larger the box size is, the closer the edge is to infinity and thus the more accurate the solution will be. Unfortunately if one only increases the size of the grid box without accordingly increasing the number of grid points, fewer grid points will be located inside the sphere, which leads to a poorer and noisier estimate of the force from numerical integration. Increasing either one will largely increase the computing time due to the dimension of the system and increasing both will drastically do so. We initiate the system using the bulk magnetic potential (magnetic potential in the absence of any particles), which will significantly decrease the computing time. In order to further decrease the computational time, we use a two-step method, similar to a multigrid method, but more efficient in solving a partial differential equation (PDE) that has discontinuous boundaries. An illustration of the grid schematic for this method is shown in Fig. 1(c). In the first step, a coarse grid in a large box is used to propagate the boundary conditions from approximated infinity. After enough steps, a fine grid in a small box is used to propagate the boundary conditions interpolated from the coarse grid results. There are two processes in this two-step method that may arouse additional errors compared to the original solver: the coarse grid solver at the first step and the interpolation when switching grids. Since the far field is a smooth dipolar field, running the solver on the coarse grid will only yield a negligible error. The interpolation from the coarse grid to the fine grid will introduce certain noise to the final force calculation, which can be smoothed out by the polynomial fit. Therefore the two-step method does not induce significant errors. A comparison between the convergence of the two-step method and the original method is discussed in

FIG. 2. (Color online) (a), (b) The contour plots of the ΔH_x and ΔH_y middle x - y plane for $r = 1.4D$ and $\alpha = 0$. (c), (d) The contour plots of the ΔH_x and ΔH_y middle x - y plane for $r = 1.1D$ and $\alpha = 0$. The scales correspond to the value of the magnetic field in units of A/m.

Part II of the Supplemental Material [18]. In the calculation the approximated infinity is set to be eight particle diameters away from the surface of the edge particle in each direction, over which the calculated force does not change by more than 0.1%. The grid numbers in each direction, N_x , N_y , and N_z , are therefore not the same but related to each other. Later, when we analyze the grid numbers, we only consider N_x as the variable. The inset in Fig. 1(c) shows parameters used for this system, where r is the center-to-center distance and α is the angular orientation of the external magnetic field for all configurations.

III. RESULTS AND DISCUSSION

A. Magnetic field

Paramagnetic particles (Dynabeads M-280, Invitrogen) with a particle radius $a = 1.4 \mu\text{m}$ and $\chi_{\text{eff}} = 0.73$ [23,27,28], which corresponds to $\chi = 0.96$, are chosen to be used in the calculations unless otherwise mentioned. Figure 1 shows how the magnetic field distributes in spheres for a particle pair. This nonuniformity leads to the increase of higher-order multipole moments. A two-body superposition analysis proceeds to investigate how the multipolar field significantly affects the near field, shown in Fig. 2. In Fig. 2, $\Delta H_x = H_{lx} + H_{rx} - H_{0x} - H_x$, where H_{lx} is the magnetic field in the x direction with a single particle on the left side, H_{rx} is the magnetic field in the x direction with a single particle on the right side, H_{0x} is the magnetic field in the x direction in the absence of either particle, and H_x is the magnetic field in the x direction when both particles are present. ΔH_y is defined analogously. ΔH_x and ΔH_y indicate how the magnetic field around a particle pair differs from the superposition of two dipolar fields. Figures 2(a) and 2(b) show for $r = 1.4D$, where D

is the diameter of the particle, that there is a slight multipolar effect near the surface of the particles when using an applied magnetic field of $H_0 = 477 \text{ A/m}$. The range of the multipolar field decreases much faster than that of the dipolar field, as shown by the Legendre expansion $\frac{P_l(\cos \theta_a)}{r^{l+1}}$ in Eq. (8). However, for $r = 1.1D$, it is shown in Figs. 2(c) and 2(d) that both ΔH_x and ΔH_y can be quite large. For example, at the center point between two spheres, ΔH_x is 17% of the applied magnetic field.

B. Force between particles in a uniform magnetic field

For a particle pair in a uniform magnetic field, the magnetic force calculated using the numerical Laplace's equation solver (LES) at various magnetic field strengths is plotted in Fig. 3. The LES method provides the most accurate representation of the magnetic force between particles. The results obtained from LES are compared to those calculated from DM and MDM. As the multipolar field is only significant in the near field, more data points are collected when the two particles are close. Note that since both magnetic force and total force are in the x direction, F is a scalar and can represent either the magnitude of the force or F_x . From Fig. 3 it is shown that for both 0° attraction and 90° repulsion, the forces calculated from LES are larger than MDM. The comparisons between Figs. 3(a) and 3(c) and between Figs. 3(b) and 3(d) both show that the shape of the curve $F(r)$ is dependent on angle α but independent of the external magnetic field strength B_0 . More specifically, as shown in the insets, the ratio of the LES result over the DM result is always fixed when changing B_0 , indicating that, as DM, the magnetic force calculated using LES must also scale as B_0^2 . The insets in Figs. 3(a) and 3(c) are not identical for the far field results as the noise introduced by

FIG. 3. (Color online) Magnetic force and total force on a particle in a particle pair system in a uniform field for (a) $B_0 = 6$ G and $\alpha = 0^\circ$, (b) $B_0 = 6$ G and $\alpha = 90^\circ$, (c) $B_0 = 2$ G and $\alpha = 0^\circ$, and (d) $B_0 = 2$ G and $\alpha = 90^\circ$. A dashed line (cyan) is used for DM, a solid line (blue) for MDM, a large dotted line (magenta) for LES, and a small dotted line (black) for the polynomial fit. All insets show the ratio of the LES results over the DM results for a large r/D range.

numerical errors in the two-step method become pronounced when the force is small [18]. The insets indicate that multipolar effects are minimized when the particle separation is far and LES always converges to DM. The polynomial used for the fit is given by

$$F_{\text{LES}} = F_{\text{DM}} + \frac{\beta_1}{r^5} + \frac{\beta_2}{r^6} + \frac{\beta_3}{r^7}, \quad (14)$$

where $F_{\text{DM}} = -\frac{3\mu_0 m^2}{2\pi r^4}$ represents the force from the dipolar field. As F_{DM} is defined as the dipolar force, $\frac{\beta_1}{r^5}$ is defined as the quadrupolar force, $\frac{\beta_2}{r^6}$ as the octupolar force, and $\frac{\beta_3}{r^7}$ as the hexadecapolar force following the definition given in Ref. [16]. Expanding the polynomial to include the hexadecapolar force is sufficient to achieve a good fit.

C. Force between particles in a rotational magnetic field

The magnetic force between a particle pair in a rotational magnetic field becomes isotropic if the rotation frequency is higher than a critical value [12]. This isotropic force can be calculated by averaging the angular force over one revolution. The angular dependence of the magnetic force for a particle pair in a rotational magnetic field is shown in Figs. 4(a) and 4(b).

Figure 4(a) shows at a relatively large separation of $r/D = 1.4$, the magnetic force calculated from LES and MDM are almost the same for all angles. However, when separation r/D decreases below 1.1, as shown in Fig. 4(b), LES yields an evidently larger attraction and smaller repulsion than both MDM and DM. This results in changing the averaged force as well as angle θ_0 , at which there is neither attraction nor repulsion. At $r/D = 1.4$, θ_0 is almost equal to the magic angle

FIG. 4. (Color online) The pair magnetic force in a rotational magnetic field (a) at $r/D = 1.4$ for different angles for a particle pair, (b) at $r/D = 1.1$ for different angles for a particle pair, (c) angular average for a particle pair, and (d) angular average for a particle pair and three-particle system. In (c), a dashed line (cyan) is used for the DM results, a solid line (blue) for the MDM results, a line with circular markers (magenta) for the LES results, and a small dotted line (black) for the polynomial fit. The inset shows the ratio of results from LES over those from DM. In (d), the difference between a pair and a three-particle system is compared by a solid line (blue) for the MDM pair results and a small dotted line (green) for MDM three-particle results, and a line with circular markers (magenta) for the LES pair results line with square markers (yellow) for the LES three-particle results. The inset shows the ratio of effective pair force results from LES on the three-particle system over pair force results from LES.

described in Ref. [6], $\theta_m = \arccos \frac{1}{\sqrt{3}} \approx 54.7^\circ$. However, at $r/D = 1.1$, $\theta_0 \approx 60^\circ$. More importantly, for LES, there is a greater net attractive force compared to MDM. As shown in Fig. 4(c), deviation from MDM emerges when r/D is smaller than 1.3 and reaches more than 20% when r/D is smaller than 1.1. The inset in Fig. 4(c) shows that when the separation increases, the magnetic force calculated by LES converges to that calculated by DM.

As shown earlier, the ratio between the magnetic force calculated using LES or DM does not depend on applied magnetic field strength, thus the magnetic force calculated using LES also scales as B_0^2 . Using the polynomial fit given by Eq. (14), the expression for the magnetic force between a particle pair in a rotational magnetic field using LES is

$$F_{\text{LES}}(r/D) = F_{\text{DM}} + \frac{B_0^2}{36} \left(\frac{-0.309 \times 10^{-13}}{(r/D)^5} + \frac{1.154 \times 10^{-13}}{(r/D)^6} + \frac{-1.229 \times 10^{-13}}{(r/D)^7} \right), \quad (15)$$

where $F_{\text{DM}} = -\frac{3\mu_0 m^2}{8\pi r^4}$. The addition of another particle will affect the magnetic force between particles. The magnetic force for a particle pair and the effective pair magnetic force for a three-particle system are calculated using LES and MDM and compared in Fig. 4(d). Here the effective pair force for a three-particle system is defined as the magnetic force on particle 3 [see the inset at the top right of Fig. 4(d)] divided by $\sqrt{3}$, which gives the decomposed pair force exerted by particle 1 or 2 only. The difference between the two-body pair force and the effective pair force calculated for a three-body system is mostly referred to as the three-body effect. The three-body effect widely exists in other systems besides paramagnetic colloid suspension in a rotational field, such as charged colloidal suspension with the Yukawa potential [29,30] and paramagnetic colloid suspension in a uniform vertical field [31]. Here a significant three-body effect of 20% is observed for LES results when $r/D = 1.06$, indicating the pair magnetic force from LES is not sufficient to describe the magnetic force for oligomer aggregates. The effective pair force calculated by MDM also deviates noticeably from that calculated by LES and the three-body effect predicted by MDM is less than that by LES. The inset at the top right of Fig. 4(d) shows that, as the multipolar effect, the three-body effect for a three-particle system stands out only in the near field and converges to unity at distance. The addition of another particle leads to a nonlinear mutual interaction, resulting in a nonlinear Laplace's equation and the three-body effect shown above. For a larger cluster, this is of particular importance for particles at the edge, which are affected much more than those in the center. This edge condition caused by the many-body effect is further demonstrated by calculating the force on cluster systems, specifically a ten-particle system and a 24-particle system shown in Figs. 5(a) and 5(b), respectively.

Figures 5(c) and 5(d) show the many-body effects on different particles for these two systems with $r/D = 1.1$. Here many-body effects are defined as the relative difference between the LES result calculated over the entire cluster and the superposition of the two-body LES results. For symmetry considerations, we only calculate forces on certain particles

FIG. 5. (Color online) Schematics of a (a) ten-particle system and (b) 24-particle system used to calculate force. The many-body effect on different particles with $r/D = 1.1$ in a rotational magnetic field for a (c) ten-particle system and (d) 24-particle system. (e) Magnetic field strength on particles in a three-particle chain. The scales correspond to the value of the magnetic field in units of A/m. Magnetic force in the 24-particle system in a rotational magnetic field for different angles with $r/D = 1.1$ for (f) exterior particle 1 and (g) interior particle 8. In (f) and (g), dashed lines (cyan) are used for the DM results, solid lines (blue) for the MDM results, and circular markers (magenta) for the LES results. The insets in (f) and (g) show the angular averaged force calculated using a different method, where D represents DM, M represents MDM, S represents superposition of LES, and L represents LES.

on a symmetric axis. For a ten-particle system, the magnetic force on exterior particles 1 and 3 shows significant many-body effects while interior particle 4 shows only a 6% deviation. Similarly, for the 24-particle system, the magnetic force for interior particles has smaller deviations between LES and the superposition results. For example, particle 8 has a 2% deviation. The many-body effects' mitigation is due to the symmetric positions of neighboring particles. As shown in Fig. 1(b), a neighboring particle is the source of nonuniform internal magnetization and the multipolar field. However, the many-body effects on the magnetic field are largely dependent on their positions. Figure 5(e) shows that the magnetic field strength distribution of the end particle in a three-particle-chain system is wider than that of the center particle, which has a smaller multipolar field and therefore should exhibit less many-body effects when calculating the magnetic force. Though the many-body effects are calculated for a hexagonally packed array of particles, the superposition method will work for any configuration which entails neighboring particles being situated symmetrically. Figures 5(f) and 5(g) further compare the difference in the magnetic force calculated by LES, MDM, and DM for particles 1 and 8, respectively. For interior particle 1, the magnetic force calculated using MDM or DM leads to a significant underestimation, as shown in Fig. 5(f) and its inset. However, as shown in Fig. 5(g) and its inset, for exterior particle 8 MDM captures the force curve very well with a net deviation of only 5% while DM continues to underestimate the magnetic force. This comparison confirms the accuracy of using MDM in dynamic simulations with periodic boundary conditions [14].

D. Changes in particle physical properties

In this section, we study how changes in the physical properties of the particles affect the magnetic force calculated using the different methods. First we consider changes in the magnetic susceptibility χ . Figure 6(a) shows the angular averaged force for the configuration shown in Fig. 4 except the susceptibility is increased to $\chi = 5$. The differences between DM and other calculations have significantly increased compared to Fig. 4(c), showing an increased multipolar effect for increased χ . The inset shows that the three-body effect for a three-particle system also increases in both magnitude and range. Therefore a higher χ yields larger multipolar effects and a larger three-body effect, indicating no apparent scaling relationship between the magnetic force and χ for LES. However, the LES force equation [Eq. (15)] scales with the particle diameter as D^2 . Figure 6(b) shows that the force scaled by D^2 from LES calculation does not change with the particle diameter. Note that $F_{DM}(r/D)$ and $F_{MDM}(r/D)$ also scale as D^2 , as derived from the expression of these two dipolar models [12]. From this scaling law, Eq. (15) can be rewritten as

$$F_{LES}(r/D) = -\frac{\pi\chi^2 D^2 B_0^2}{96\mu_0(r/D)^4} + \frac{D^2 B_0^2}{36} \left(\frac{-0.395 \times 10^{-2}}{(r/D)^5} + \frac{1.472 \times 10^{-2}}{(r/D)^6} + \frac{-1.568 \times 10^{-2}}{(r/D)^7} \right). \quad (16)$$

FIG. 6. (Color online) (a) Angular average of the pair magnetic force in a rotational magnetic field for $B_0 = 6$ G and $\chi = 5$ from DM (cyan dashed line), MDM (blue line), and MDM on a three-particle system (green dotted line), LES (magenta line with circular markers), and LES on a three-particle system (yellow line with square markers). (b) Angular average of the pair magnetic force over the particle diameter in a rotational field for $B_0 = 6$ G, $\chi = 0.96$, and different D : DM for both $D = 2.8$ and $6 \mu m$ (cyan dashed line), MDM for both $D = 2.8$ and $6 \mu m$ (blue line), LES for $D = 2.8 \mu m$ (red line with circular markers), LES for $D = 6 \mu m$ (green line with square markers), and Eq. (16) (black small dotted line).

This equation is the highlight of our results. It can be used to accurately describe the magnetic force between paramagnetic particles with $\chi = 0.96$ and $\chi_{eff} = 0.73$. For particles of a different susceptibility, a different polynomial fit can be derived from the LES calculation result for that particular susceptibility.

IV. CONCLUSION

We demonstrate that by using a smoothed profile method, the magnetic force on magnetic particles can be precisely calculated by numerically solving the magnetostatic Laplace's equation and calculating the Maxwell stress tensor. With this method, we examined the performance of conventional DM and MDM methods compared to the numerical LES solution for Dynabeads M-280. In a uniform magnetic field, MDM

gives an acceptable estimation to LES for a particle pair. In a rotational magnetic field, the MDM fails to capture the two-body force when the distance is small. For a three-particle system, neither MDM nor the superposition of two-body LES force gives a good approximation. Thus for oligomer aggregates it is suggested that LES calculation should be used. For a large particle cluster, the many-body effect was shown to affect particles at the edge. For interior particles in a cluster, both MDM and the superposition of the two-body force result from LES give a good approximation to the many-body force, as confirmed by the force calculation for a ten-particle system and a 24-particle system. Lastly, though the LES force results show no apparent scaling law for susceptibility, it has a square

scaling with the particle diameter just as DM and MDM. The pair additivity of the two-body LES force result is very useful for dynamic simulations of a paramagnetic particle system with periodic boundary conditions which require an accurate force calculation.

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