Interparticle force in polydisperse electrorheological fluids:
Beyond the dipole approximation

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Abstract

We have developed a multiple image method to compute the interparticle force for a polydisperse electrorheological (ER) fluid. We apply the formalism to a pair of dielectric spheres of different dielectric constants and calculate the force as a function of the separation. The results show that the point-dipole (PD) approximation errs considerably because many-body and multipolar interactions are ignored. The PD approximation becomes even worse when the dielectric contrast between the particles and the host medium is large. From the results, we show that the dipole-induced-dipole (DID) model yields very good agreements with the multiple image results for a wide range of dielectric contrasts and polydispersity. The DID model accounts for multipolar interaction partially and is simple to use in computer simulation of polydisperse ER fluids.

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I. INTRODUCTION

Polydisperse electrorheological (ER) fluids have attracted considerable interest recently because the size distribution and dielectric properties of the suspending particles can have significant impact on the ER response [1]. Real ER fluids must be polydisperse in nature: the suspending particles can have various sizes or different permittivities. In a recent paper [2], we investigated the case when the suspending particles are of different sizes. In this work, we extend the study to that of different dielectric constants.

The point-dipole approximation [3] is routinely adopted in computer simulation [4,5] because it is simple and easy to use. Since many-body and multipolar interactions between particles have been neglected, the predicted strength of ER effects is of an order lower than the experimental results. Hence, substantial effort has been made to sort out more accurate models [6–8]. Recently, we have developed a multiple image method and an integral equation approach to compute the interparticle force. In particular, we proposed a dipole-induced-dipole (DID) model for efficient computer simulation of polydisperse ER fluids [2].

Poladian [9] claimed that the multiple image method can be used to calculate the dipole moment of a pair identical dielectric spheres in an applied electric field. In Ref. [2], we generalized the multiple image method to a pair of dielectric spheres of different sizes. We showed that the generalization yields a reasonable approximation when the spheres have a large dielectric constant. The multiple image method was widely adopted [10–12]. The approximation is reasonable because in ER fluids, the dielectric constant of the particles can be much larger than that of the host fluid. However, the results for low contrast are questionable.

In fact there is a more complicated image method for a dielectric sphere [13], which gives the exact image dipole moment of a dielectric sphere that placed in front of a point dipole. We thus modify the multiple image formula. The results of the improved formula agree with the numerical solution of an integral equation method [14,15] even when the dielectric contrast of the spheres is low.
In this work, we extend the multiple image method to compute the interparticle forces for a polydisperse mixture of dielectric spheres of different dielectric constants. The DID model will be compared with the Klingenberg’s empirical force expressions [3].

II. IMPROVED MULTIPLE IMAGE METHOD

Here we briefly review the method and extend the method slightly to handle different dielectric constants. Consider a pair of dielectric spheres, of radii \(a\) and \(b\), dielectric constants \(\epsilon_1\) and \(\epsilon'_1\) respectively, separated by a distance \(r\). The spheres are embedded in a host medium of dielectric constant \(\epsilon_2\). Upon the application of an electric field \(E_0\), the induced dipole moment inside the spheres are respectively given by:

\[
p_{a0} = \beta \epsilon_2 E_0 a^3, \quad p_{b0} = \beta' \epsilon_2 E_0 b^3,
\]

where the dipolar factors \(\beta, \beta'\) are given by:

\[
\beta = \frac{\epsilon_1 - \epsilon_2}{\epsilon_1 + 2\epsilon_2}, \quad \beta' = \frac{\epsilon'_1 - \epsilon_2}{\epsilon'_1 + 2\epsilon_2}.
\]

From the multiple image method [2], the total dipole moment inside sphere \(a\) is:

\[
p_{aT} = (\sinh \alpha)^3 \sum_{n=1}^{\infty} \left[ \frac{p_{a0}b^3(-\beta)^{n-1}(-\beta')^{n-1}}{(b \sinh n\alpha + a \sinh(n-1)\alpha)^3} + \frac{p_{b0}a^3(-\beta)^n(-\beta')^{n-1}}{(r \sinh n\alpha)^3} \right],
\]

\[
p_{aL} = (\sinh \alpha)^3 \sum_{n=1}^{\infty} \left[ \frac{p_{a0}b^3(2\beta)^{n-1}(2\beta')^{n-1}}{(b \sinh n\alpha + a \sinh(n-1)\alpha)^3} + \frac{p_{b0}a^3(2\beta)^n(2\beta')^{n-1}}{(r \sinh n\alpha)^3} \right],
\]

where the subscripts \(T\) (\(L\)) denote a transverse (longitudinal) field, i.e., the applied field is perpendicular (parallel) to the line joining the centers of the spheres. Similar expressions for the total dipole moment inside sphere \(b\) can be obtained by interchanging \(a\) and \(b\), as well as \(\beta\) and \(\beta'\). The parameter \(\alpha\) satisfies:

\[
cosh \alpha = \frac{r^2 - a^2 - b^2}{2ab}.
\]

In Ref. [3], we checked the validity of these expressions by comparing with the integral equation method. We showed that these expression are valid at high contrast. Our improved expressions will be shown to be good at low contrast as well (see below).
The force between the spheres is given by [10]:

\[ F_T = \frac{E_0}{2} \frac{\partial}{\partial r} (p_{aT} + p_{bT}), \quad F_L = \frac{E_0}{2} \frac{\partial}{\partial r} (p_{aL} + p_{bL}). \] (5)

For monodisperse ER fluids, Klingenberg defined an empirical force expression [6]:

\[ \frac{F}{F_{PD}} = (2F_\parallel \cos^2 \theta - F_\perp \sin^2 \theta) \hat{r} + F_\Gamma \sin 2\theta \hat{\theta}, \] (6)

being normalized to the point-dipole force \( F_{PD} = -3p_0^2/r^4 \), where \( F_\parallel, F_\perp \) and \( F_\Gamma \) (all tending to unity at large separation) are three force functions being determined from numerical solution of Laplace’s equation. The Klingenberg’s force functions can be shown to relate to our multiple image moments as follow (here \( a = b, \beta = \beta' \) and \( p_a = p_b \)):

\[ F_\parallel = \frac{1}{2} \frac{\partial \tilde{p}_L}{\partial r}, \quad F_\perp = -\frac{\partial \tilde{p}_T}{\partial r}, \quad F_\Gamma = \frac{1}{r} (\tilde{p}_T - \tilde{p}_L), \] (7)

where \( \tilde{p}_L = p_L/F_{PD}E_0 \) and \( \tilde{p}_T = p_T/F_{PD}E_0 \) are the reduced multiple image moments. We computed the numerical values of these force functions separately by the approximant of Table I of the second reference of Ref. [6] and by Eq.(7).

In Fig.1, we plot the multiple image results and the Klingenberg’s empirical expressions. We show results for the perfectly conducting limit (\( \beta = 1 \)) only. For convenience, we define the reduced separation \( \sigma = r/(a+b) \). For reduced separation \( \sigma > 1.1 \), simple analytic expressions were adopted by Klingenberg. As evident from Fig.1, the agreement with the multiple image results is impressive at large reduced separation \( \sigma > 1.5 \), for all three empirical force functions. However, significant deviations occur for \( \sigma < 1.5 \), especially for \( F_\parallel \). For \( \sigma \leq 1.1 \), alternative empirical expressions were adopted by Klingenberg. For \( F_\perp \), the agreement is impressive, although there are deviations for the other two functions. From the comparison, we would say that reasonable agreements have been obtained. Thus, we are confident that the multiple image expressions give reliable results.

III. DIPOLE-INDUCED-DIPOLE MODEL

The analytic multiple image results can be used to compare among the various models according to how many terms are retained in the multiple image expressions: (a) point-
dipole (PD) model: $n = 1$ term only, (b) dipole-induced-dipole (DID) model: $n = 1$ to $n = 2$ terms only, and (c) multipole-induced-dipole (MID) model: $n = 1$ to $n = \infty$ terms.

The multiple image expressions [Eqs.(3)–(6)] allows us to calculate the correction factor defined as the ratio between the DID and PD forces:

$$F^{(T)}_{\text{DID}} / F^{(T)}_{\text{PD}} = 1 - \frac{\beta a^3 r^5}{(r^2 - b^2)^4} - \frac{\beta' b^3 r^5}{(r^2 - a^2)^4} + \frac{\beta \beta' a^3 b^3 (3r^2 - a^2 - b^2)}{(r^2 - a^2 - b^2)^4}, \quad (8)$$

$$F^{(L)}_{\text{DID}} / F^{(L)}_{\text{PD}} = 1 + \frac{2\beta a^3 r^5}{(r^2 - b^2)^4} + \frac{2\beta' b^3 r^5}{(r^2 - a^2)^4} + \frac{4\beta \beta' a^3 b^3 (3r^2 - a^2 - b^2)}{(r^2 - a^2 - b^2)^4}, \quad (9)$$

where $F^{(T)}_{\text{PD}} = 3p_{a0}p_{b0}/r^4$ and $F^{(L)}_{\text{PD}} = -6p_{a0}p_{b0}/r^4$ are the point-dipole forces for the transverse and longitudinal cases respectively. These correction factors can be readily calculated in computer simulation of polydisperse ER fluids. The results show that the DID force deviates significantly from the PD force at high contrast when $\beta$ and $\beta'$ approach unity. The dipole induced interaction will generally decrease (increase) the magnitude of the transverse (longitudinal) interparticle force with respect to the point-dipole limit.

In a previous work [4], we examine the case of different size but equal dielectric constant ($\beta = \beta'$) only. Here we focus on the case $a = b$ and study the effect of different dielectric constants. In Fig.2, we plot the interparticle force in the transverse field case against the reduced separation $\sigma$ between the spheres for $\beta = 1/3$ and various $\beta'/\beta$ ratios. At low contrast, the DID model almost coincides with the MID results. In contrast, the PD model exhibits significant deviations. Similar conclusion can be drawn from the longitudinal field case as in Fig.3. It is evident that the DID model generally gives better results than PD for all polydispersity.

At higher contrast, the DID model still agrees with the MID model, except at close encounter. In Figs.4 and 5, we plot the force in the transverse and longitudinal field cases against the reduced separation $\sigma$. It is evident that the DID model generally gives better results than PD for all polydispersity. For the longitudinal field case, the DID model agrees with the MID model for $\sigma > 1.2$, except at close encounter where the MID force diverges as $\sigma \to 1$. 

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CONCLUSION

In summary, we have used the multiple image to compute the interparticle force for a polydisperse electrorheological fluid. We apply the formalism to a pair of spheres of different dielectric constants and calculate the force as a function of the separation. The results show that the point-dipole approximation is oversimplified. It errs considerably because many-body and multipolar interactions are ignored. The dipole-induced-dipole model accounts for multipolar interactions partially and yields overall satisfactory results in computer simulation of polydisperse ER fluids while it is easy to use.

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Figure Captions

Fig.1: Comparison of the multiple image results with Klingenberg’s empirical force expression.

Fig.2: Interparticle force for transverse field, $\beta=1/3$ while $\beta'/\beta$ ranges from 1.0 to 1.2.

Fig.3: Same as Fig.2. Force for longitudinal field.

Fig.4: Force for transverse field, $\beta=9/11$ while $\beta'/\beta$ ranges from 1.0 to 1.2.

Fig.5: Same as Fig.4. Force for longitudinal field.
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Fig. 1: Comparison with Klingenberg

\[ F_{\text{perp}} \]

\[ F_{\parallel} \]

\[ F_{\Gamma} \]

- **MID**
- **Klingenberg**
Fig. 2: Force for Transverse Field, $\beta=1/3$

- $\beta'=\beta$
- $\beta'=1.05\beta$
- $\beta'=1.1\beta$
- $\beta'=1.2\beta$

The graphs show the force $F_T$ as a function of $\sigma$, with three different values of $\beta'$ and three different models represented: MID, DID, and PD.
Fig. 3: Force for Longitudinal Field, $\beta = 1/3$

- $\beta' = \beta$
- $\beta' = 1.05\beta$
- $\beta' = 1.1\beta$
- $\beta' = 1.2\beta$

Graphs showing the force $F_L$ against $\sigma$ for different values of $\beta'$ with various markers and line styles.
Fig. 4: Force for Transverse Field, $\beta = 9/11$

- $\beta' = \beta$
- $\beta' = 1.05\beta$
- $\beta' = 1.1\beta$
- $\beta' = 1.2\beta$

Graphs showing $F_T$ vs $\sigma$ for different values of $\beta'$, with different markers and line styles representing MID, DID, and PD.
Fig.5: Force for Longitudinal Field, $\beta=9/11$

- $\beta'=\beta$
- $\beta'=1.1\beta$
- $\beta'=1.05\beta$
- $\beta'=1.2\beta$

Graphs showing force $F_L$ vs. $\sigma$ for different values of $\beta'$. Legend includes 'MID', 'DID', and 'PD'.