Vol 13 No 7, July 2004
1009-1963/2004/13(07)/1065-05

Chinese Physics

© 2004 Chin. Phys. Soc.
and IOP Publishing Ltd

Many-body dipole-induced dipole model for electrorheological fluids*

Huang Ji-Ping(黄吉平)[a)b)†] and Yu Kin-Wah(余建华)[a)‡]

[a)] *Department of Physics, The Chinese University of Hong Kong, Shatin, NT, Hong Kong, China*

[b)] *Laboratory of Computational Engineering, Helsinki University of Technology, PO Box* 9203, *FIN*-02015 *HUT, Finland*

(Received 16 July 2003; revised manuscript received 24 December 2003)

Theoretical investigations on electrorheological (ER) fluids usually rely on computer simulations. An initial approach for these studies would be the point-dipole (PD) approximation, which is known to err considerably when the particles approach and finally touch each other due to many-body and multipolar interactions. Thus various works have attempted to go beyond the PD model. Being beyond the PD model, previous attempts have been restricted to either local-field effects only or multipolar effects only, but not both. For instance, we recently proposed a dipole-induced-dipole (DID) model which is shown to be both more accurate than the PD model and easy to use. This work is necessary because the many-body (local-field) effect is included to put forth the many-body DID model. The results show that the multipolar interactions can indeed be dominant over the dipole interaction, while the local-field effect may yield a correction.

Keywords: electrorheological fluid, many-body (local-field) effect, multipolar interaction
PACC: 4660H, 6480G, 7730

1. Introduction

An electrorheological (ER) fluid consists of polarizable particles in a nonconducting host fluid. If an external electric field is applied to an ER fluid, the particles aggregate and form chains parallel to the applied field. These chains may further aggregate to column-like structures.[1] To discuss the ER effect, early theory is often based on the point-dipole (PD) approximation, e.g. Ref.[2]. Also, the PD approximation is often adopted in computer simulation [3] because it is simple and easy to use. Since many-body and multipolar interactions between particles have been neglected, the predicted strength of ER effect is of an order lower than the experimental results. Hence, much work has been done to sort out more accurate models.[4–8] Although those methods are accurate, they are relatively complicated to use in dynamic simulation of ER fluids.[9]

In a recent paper, we put forth the dipole-induced dipole (DID) model to improve the PD model.[10] In that work, we considered pair interaction between polarized dielectric particles through the multiple image formula, but neglected the many-body interactions because it was believed that the multipolar interactions can be dominant over the many-body (local-field) effects. The claim was based on the calculation for a pair of particles. We should really question the quantitative accuracy of this claim, since in ER fluids chain and sheet structures are known to occur. In a long particle chain the local field at a dipole site is much larger than for a pair of dipoles, whereas for multipolar interactions this is probably not the case. In fact, this issue has already been quantified, for particles with large dielectric contrast with the liquid phase, by Martin and Anderson.[11] Thus we have to do some additional analysis to validate the claim.

As an initial model, we may adopt an effective dipole factor in the multiple image formula.[10] More precisely, we regard each polarized particle in an ER suspension as embedded in an effective medium with an effective dielectric constant ϵ_e. Thus, the usual dipole factor $b = (\epsilon_1 - \epsilon_2)/(\epsilon_1 + 2\epsilon_2)$ in the multiple image theory should be replaced by $b' = (\epsilon_1 -$

*Project supported by the Research Grants Council of the Hong Kong SAR Government under project nos CUHK 4245/01P and CUHK 403303.

†E-mail: jphuang@mpip-mainz.mpg.de
‡E-mail: kwyu@phy.cuhk.edu.hk

$\epsilon_e)/(\epsilon_1 + 2\epsilon_e)$,[12,13] where ϵ_e is the effective dielectric constant of the surrounding medium, which is conveniently calculated in the Maxwell–Garnett approximation (MGA). Here ϵ_1 and ϵ_2 are the dielectric constants of the suspended particles and host fluid, respectively. However, the situation is further complicated by the fact that the suspended particles in an ER fluid usually form anisotropic structures, e.g. chain and sheet structures, along the applied field and that ϵ_e can be anisotropic. Thus, ϵ_e should be calculated within the anisotropic MGA.[14−16] A preliminary calculation shows that ϵ_e should be increased and b' should be decreased as compared to b. Thus the DID interaction would be reduced. But, of course, the actual reduction should depend on the depolarization factor of the anisotropic structures. Thus the many-body effects have been included through the volume-fraction dependent ϵ_e.

So, in the presence of the surrounding particles, the interaction between two particles will be modified. By means of an effective dielectric constant, we can derive an analytic formula for the many-particle DID model. And more importantly, we can access the importance of the many-body (local-field) effects against the multipolar interactions. We believe this work is necessary because being beyond the fixed dipole model, the previous work dealt with either local-field effects only [17] or multipolar effects only, but not both.

2. Formalism

We concentrate on the case where highly polarized dielectric particles of diameter d, dielectric constant ϵ_1 are embedded in a host fluid of ϵ_2.

In the dilute limit, the dipole factor for an isolated particle may be given by

$$b = \frac{\epsilon_1 - \epsilon_2}{\epsilon_1 + 2\epsilon_2}. \tag{1}$$

On the other hand, based on the multiple image method [10] the dipole factor for a pair of particles with separation s has the following form:

$$\begin{aligned} b_T^* &= b\sum_{n=0}^{\infty}(-b)^n\left[\frac{\sinh\alpha}{\sinh[(n+1)\alpha]}\right]^3, \\ b_L^* &= b\sum_{n=0}^{\infty}(2b)^n\left[\frac{\sinh\alpha}{\sinh[(n+1)\alpha]}\right]^3, \end{aligned} \tag{2}$$

for the transverse field (T) and longitudinal field (L) case, respectively, where α satisfies the relation $\cosh\alpha = s/d$.

When a suspension containing many dielectric particles is subjected to an intense electric field, the induced dipole moments may cause the particles to form chains along the applied field, resulting in complex anisotropic structures. In this case, we may invoke the MGA for anisotropic composites [14−16] to obtain the effective dielectric function ϵ_e of the system. For the transverse field case when the electric field is applied perpendicular to the uniaxial anisotropic axis, the MGA has the form

$$\frac{\epsilon_{eT} - \epsilon_2}{\beta_T\epsilon_{eT} + (3-\beta_T)\epsilon_2} = f\frac{\epsilon_1 - \epsilon_2}{\epsilon_1 + 2\epsilon_2}; \tag{3}$$

whereas for a longitudinal field case when the field is applied along the uniaxial anisotropic axis, the MGA reads as

$$\frac{\epsilon_{eL} - \epsilon_2}{\beta_L\epsilon_{eL} + (3-\beta_L)\epsilon_2} = f\frac{\epsilon_1 - \epsilon_2}{\epsilon_1 + 2\epsilon_2}, \tag{4}$$

where β_T and β_L denote the local field factors perpendicular and parallel to the uniaxial anisotropic axis, and f the volume fraction of particles. These local field factors are defined as the ratio of the local field in the particles to the Lorentz cavity field.[14] For isotropic composites, $\beta_L = \beta_T = 1$, while both β_L and β_T will deviate from unity for an anisotropic distribution of particles in composites. These β factors have been evaluated in a tetragonal lattice of dipole moments [14] and in various field-structured composites [11] and they satisfy the sum rule $\beta_L + 2\beta_T = 3$. In this case, the dipole factor for an individual particle may have the form [12,13]

$$b' = \frac{\epsilon_1 - \epsilon_e}{\epsilon_1 + 2\epsilon_e}. \tag{5}$$

Note that to obtain this equation, we have replaced ϵ_2 in Eq.(1) with ϵ_e. That is, we assume the particle to be embedded in an effective medium. Thus the many-body (local-field) effect is included already in Eq.(5). To put forth the many-body DID model, we may consider a pair of dielectric spheres as well. The spheres are placed in an effective medium. A constant electric field $\boldsymbol{E}_0 = E_0\hat{\boldsymbol{z}}$ is applied to the spheres, which contributes to each sphere a dipole moment given by p_{10} and $p_{20}(= p_{10} = \epsilon_e E_0 d^3 b'/8)$. Thus the multiple image formula can be developed in a similar way.[10] The dipole moment p_{10} induces an image dipole p_{11} in sphere 2, while p_{11} induces yet another image dipole in sphere 1. As a result, multiple images are formed. Similarly, p_{20} induces an image p_{21} inside sphere 1, and hence another infinite series of image dipoles are

formed. Then we may obtain the sum of dipole moment inside each sphere, and hence the desired expressions for dipole factors:

$$b'^{*}_{\mathrm{T}} = b' \sum_{n=0}^{\infty} (-b')^n \left[\frac{\sinh\alpha}{\sinh[(n+1)\alpha]}\right]^3,$$
$$b'^{*}_{\mathrm{L}} = b' \sum_{n=0}^{\infty} (2b')^n \left[\frac{\sinh\alpha}{\sinh[(n+1)\alpha]}\right]^3, \qquad (6)$$

for transverse field and longitudinal field case, respectively. The two equations are nontrivial results indeed, which include the multiple image as well as many-body (local-field) effect.

To discuss the interparticle force, we may take one step forward to calculate the force between two dielectric spheres. We have already obtained the dipole factors b'^{*} and b^{*} and hence the dipole moments, and the force can be calculated by an energy approach. In this case, the dipole energy (E_n) of a pair of particles may be determined by the dot product of dipole moment and the electric field, and hence the force between a pair of particles is given by the derivative of dipole energy with respect to separation, namely $-\mathrm{d}E_n/\mathrm{d}s$. So, based on this relation, we may obtain the expressions for interparticle forces, respectively:

$$F^{*}_{\mathrm{T}} = -(1/8)\epsilon_2 E_0^2 d^3 b \sum_{n=0}^{\infty} (-b)^n \Phi_n,$$
$$F^{*}_{\mathrm{L}} = -(1/8)\epsilon_2 E_0^2 d^3 b \sum_{n=0}^{\infty} (2b)^n \Phi_n,$$
$$F'^{*}_{\mathrm{T}} = -(1/8)\epsilon_{\mathrm{eT}} E_0^2 d^3 b'_{\mathrm{T}} \sum_{n=0}^{\infty} (-b'_{\mathrm{T}})^n \Phi_n,$$
$$F'^{*}_{\mathrm{L}} = -(1/8)\epsilon_{\mathrm{eL}} E_0^2 d^3 b'_{\mathrm{L}} \sum_{n=0}^{\infty} (2b'_{\mathrm{L}})^n \Phi_n, \qquad (7)$$

with

$$\Phi_n = \frac{\sinh\alpha\cosh\alpha\sinh[(n+1)\alpha] - (n+1)\sinh^2\alpha\cosh[(n+1)\alpha]}{(d/3)\sinh^4[(n+1)\alpha]}, \qquad (8)$$

where F^{*}_{T} (F^{*}_{L}) indicates the interparticle force between a pair of particles in fluid host for transverse (longitudinal) field case, and F'^{*}_{T} (F'^{*}_{L}) in an effective medium for transverse (longitudinal) field case. It is shown that the multipolar effect as well as many-body effects have been taken into account. Setting n up to 1 will yield directly the point-dipole forces, namely F_{T} and F_{L}.

So far, we have put forth a many-body DID model. We are now in a position to do some numerical calculations to discuss the β effect on reduction factor $R = b'^{*}/b^{*}$, which indicates the correction of the many-body (local-field) effect on the multiple image effect, as well as on the ratio of b^{*} or b'^{*} to b. Then, to investigate such effect on interparticle force, we also investigate the interparticle force normalized by point-dipole force.

3. Numerical results

For numerical calculations, we choose $\epsilon_1 = 10\epsilon_0$, $\epsilon_2 = 2.5\epsilon_0$, $d = 2.0 \times 10^{-7}$ and $s/d = 1.01$, where ϵ_0 is the dielectric constant of free space. From the numerical calculations, we find that a domination of the multipolar interactions is possibly over the many-body (local-field) effects. More precisely, in Fig.1, for the transverse field case, the correction due to the local-field effect is always small, because the reduction factor is close to unity. However, for the longitudinal field case, it is evident that the local-field effect can be large, especially at high volume fractions and/or large β_{L}. In this case, the correction due to local field cannot be neglected. In Fig.2, we compute the ratio of b^{*} or b'^{*} to b versus the local field factor β_{L} for different volume fractions. In Fig.3, we discuss the dependence of β_{L} on the interparticle force normalized by PD force for different volume fractions. From Figs.2 and 3, it is obvious that the multipolar interactions can indeed be dominant over the dipole interaction, while the local-field effect may yield an important correction.

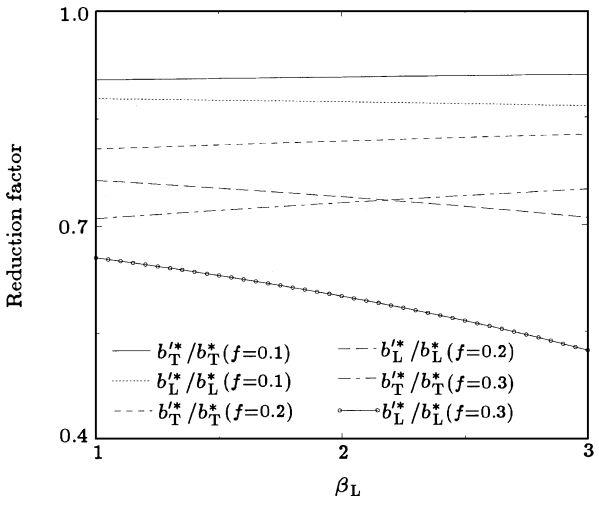


Fig.1. The reduction factor versus local field factor β_{L} for different volume fractions, f=0.1, 0.2 and 0.3.

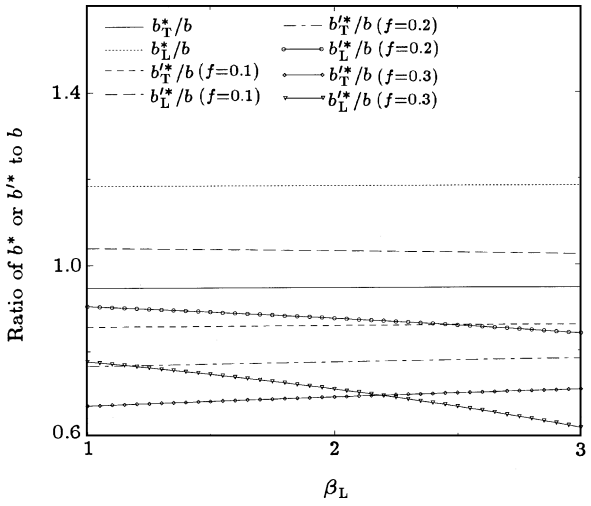


Fig.2. The ratio of b^* or b'^* to b versus local field factor β_L for different volume fractions, f=0.1, 0.2 and 0.3.

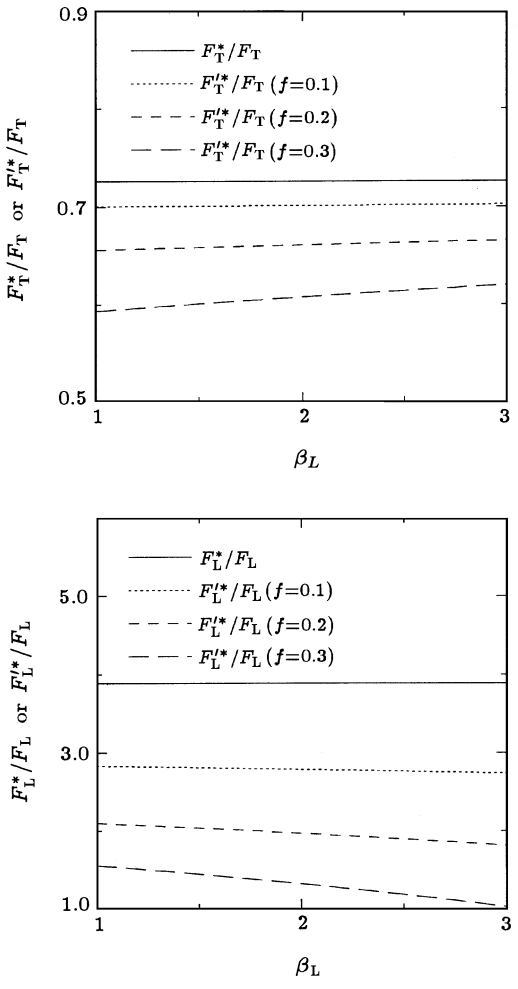


Fig.3. The ratio of interparticle force to PD force for different volume fractions, f=0.1, 0.2 and 0.3.

The force expression is reasonable, as the $n = 0$ term of Eq.(8) vanishes while the $n = 1$ term gives the correct dipole interaction force r^{-4} dependence. The divergence of the summation may be found for the longitudinal field case, due to the small distance $s/d = 1.01$ being used.

From the above results, it is clear that the correction due to many-body (local-field) effects can be very large, especially at large volume fractions. Nevertheless, for small volume fractions, the correction due to the local field can be neglected. The reduction of the magnitude of the effective interaction between two particles can be understood in a simple geometry in which the two particles interact in the presence of a third particle. An average over all possible positions of the third particle gives the desired reduction which is valid both for longitudinal and transverse fields. So, we believe our results are correct.

4. Discussion and conclusion

Here a few comments are in order. The ER effect arises from the polarization forces produced by a mismatch in the dielectric constants of the disperse and continuous phases. Typical ER fluids contain a disperse phase with volume fractions in the range of 0.05–0.50. In this sense, it is reasonable to set $f = 0.1, 0.2, 0.3$ in our calculations. On the other hand, we set $\epsilon_2 = 2.5\epsilon_0$, which is the actual value for silicon oil and commonly employed as the host fluid. Also, the disperse phases are generally composed of solid, nonconducting, or semiconducting materials with dielectric constants in the range of 2–40. Thus, $\epsilon_1 = 10\epsilon_0$ is also physical in our numerical calculations. In addition, the anisotropic local field factor β_L should satisfy $1 \leq \beta_L \leq 3$, in order to ensure $\beta_T \geq 0$. Regarding the separation between a pair of particles, various different values have been used as well, e.g. $s/d = 1.03$ etc., and similar results are obtained (not shown here). However, for the case where the separation is too large, e.g. $s/d > 2$, the multiple image effect will become small enough to be neglected.[18,19]

Our many-body DID model may be used in various studies of the behaviour of ER fluids, e.g., in a theory describing the ground state of ER solids or in a computer simulation of ER fluids. By including the many-body effects, the DID model can be used with higher accuracy. Also, it is interesting to apply the present model to the optical properties [20] of ER fluids by taking into account the anisotropic effect. Finally, it is straightforward to extend our model to deal with coated [21] or graded colloidal particles,[22,23] polydis-

perse ER fluids, and to discuss the magnetorheological suspensions.

In summary, we have developed a multiple DID model by taking into account the many-body (local-field) effect, to compute the interparticle force for an ER fluid. We apply the formalism to a pair of touching particles embedded in an effective medium, and calculate the force as a function of the anisotropic local field factor. The results show that the multipolar interactions can indeed be dominant over the dipole interaction, while the local-field effect may yield a correction.

Acknowledgments

J. P. Huang thanks Dr Mikko Karttunen for his invitation to the Laboratory of Computational Engineering, Helsinki University of Technology.

References

[1] Halsey T C 1992 *Science* **258** 761
[2] Tao R and Sun J M 1991 *Phys. Rev. Lett.* **67** 398
[3] Klingenberg D J, Swol F V and Zukoski C F 1991 *J. Chem. Phys.* **94** 6160
[4] Klingenberg D J, Swol F V and Zukoski C F 1991 *J. Chem. Phys.* **94** 6170
[5] Davis L C 1992 *Appl. Phys. Lett.* **60** 319
[6] Clercx H J H and Bossis G 1993 *Phys. Rev.* E **48** 2721
[7] Yu K W, Wan J T K and Sun H 2000 *Physica* B **279** 78
[8] Zhao H P, Liu Z Y and Liu Y Y 2001 *Chin. Phys.* **10** 35
[9] Siu Y L, Wan J T K and Yu K W 2001 *Phys. Rev.* E **64** 051506
[10] Yu K W and Wan J T K 2000 *Comput. Phys. Commun.* **129** 177
[11] Martin J E and Anderson R A 1999 *J. Chem. Phys.* **111** 4273
[12] Huang J P, Yu K W and Gu G Q 2002 *Phys. Lett.* A **300** 385
[13] Gao L, Huang J P and Yu K W 2003 *Phys. Rev.* E **67** 021910
[14] Lo C K, Wan J T K and Yu K W 2001 *J. Phys.: Condens. Matter* **13** 1315
[15] Huang J P, Wan J T K, Lo C K and Yu K W 2001 *Phys. Rev.* E **64** 061505(R)
[16] Huang J P, Gao L and Yu K W 2003 *J. Appl. Phys.* **93** 2871
[17] Wang Z W, Lin Z F and Tao R B 1996 *Int. J. Mod. Phys.* B **10** 1153
[18] Huang J P, Yu K W and Gu G Q 2002 *Phys. Rev.* E **65** 021401
[19] Huang J P, Karttunen M, Yu K W and Dong L 2003 *Phys. Rev.* E **67** 021403
[20] Gu Y and Yu K W 2002 *Chin. Phys.* **11** 601
[21] Huang J P and Yu K W 2002 *J. Phys.: Condens. Matter* **14** 1213
[22] Huang J P, Yu K W, Gu G Q and Karttunen M 2003 *Phys. Rev.* E **67** 051405
[23] Li Y, Song J and Zhang Z M 2003 *Chin. Phys.* **12** 1187