Force acting on the microparticles in electrorheological solids under the application of a nonuniform ac electric field

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Abstract

Under the application of electric fields, the structure of electrorheological (ER) solids can be changed from the body-centered tetragonal lattice to other lattices. We have derived the dipole factor for the lattice by taking into account the local-field effect through the Ewald–Kornfeld formulation, and expressed it in the spectral representation exactly. It is found that when the ER solid is subject to a nonuniform ac electric field, the force acting on the microparticle can be affected by the structure transformation, and local-field effect as well as field frequency. Our results are very well understood in the spectral representation theory.

Electrorheological (ER) fluids [1] are the suspension in which the induced dipole moment can order the suspended polarizable particles into columns under the application of a strong electric field. The rapid field-induced aggregation [2] and the large anisotropy [3] of ER fluids render this material potentially important for technological applications. As the external field exceeds a critical field, the ER fluid turns into a solid (ER solid), the ground state of which is a body-centered tetragonal (bct) lattice [4]. For the ER solid, ones [5] proposed that a structure transformation from the bct lattice to some other lattices can appear when a magnetic field is simultaneously applied perpendicular to the electric field and the particles have magnetic dipole moments. To one’s interest, this proposal was verified experimentally and a structure transformation from the bct lattice to some other lattices appears when a magnetic field is simultaneously applied perpendicular to the electric field and the particles have magnetic dipole moments. To others, induced to appear [8] which acts on the microparticles due to the interaction between the induced dipole moment inside the particle and the nonuniform field. This force $F(f)$ is a function of field frequency $f$, and given by [8]

$$F(f) = 2\pi r^3 \text{Re} \{\text{Dipole Factor} \} \left(\frac{\text{E}_{\text{rms}}}{r}\right)^2,$$

where $r$ stands for the radius of the particle, $\epsilon_2$ the real dielectric constant of the host oil, $E_{\text{rms}}$ the root-mean-square magnitude of the applied nonuniform electric field, and $\text{Re} [\text{Dipole Factor}]$ denotes the real part of Dipole Factor (also known as Clausius–Mossotti factor).

In this work, we shall investigate this kind of force acting on the particle in an ER solid, by taking into account the effect of structure transformation as well as the local-field effect. In fact, we have presented an effective medium theory (EMT) which neglects the lattice effect, for considering the local-field effect on the dielectric properties of colloidal particles in the previous Letter [9]. In other words, the EMT cannot be used to include the detailed structural information like bct or bcc lattices in real electrorheological solids, whereas the Ewald–Kornfeld formulation [10–12] can. In this regard, in the present Letter, we shall resort to the Ewald–Kornfeld formulation, in an attempt to compute the local electric field for a particle in the ER solid by taking

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into account the influence of the local-field effect arising from all the other particles in lattices, and hence obtain the desired force.

Consider the ground state of an ER solid, namely a bct lattice, which can be regarded as a tetragonal lattice, plus a basis of two particles each of which is fixed with a point dipole at its center. One of the two particles is located at a corner and the other one at the body center of the tetragonal unit cell. Its lattice constants are denoted by \( c = q \ell \) and \( a(= b) = \ell q^{-1/2} \) along the \( z \)- and \( x \)- (\( y \))-axes, respectively. In this case, the uniaxial anisotropic axis is directed along \( z \) axis. As \( q \) varies, the volume of the unit cell keeps the same, \( V_c = \ell^3 \). Thus, the degree of anisotropy of the tetragonal lattice is measured by how \( q \) is deviated from unity.

When one applies an \( x \)-directed external electric field \( \vec{E}_0 \), the dipole moments, \( \vec{p} = \vec{p}_x \), are perpendicular to the uniaxial anisotropic axis. Then, the local-field \( \vec{E} \) (e.g., \( \vec{E} = E_0 \hat{x}, \vec{E}_z = 0 \)) at the lattice point \( \vec{R} = \vec{0} \) has the following form (i.e., Ewald–Kornfield formulation)

\[
E_x = \frac{p}{V_c} \sum_{j=1}^{2} \sum_{k \neq 0} \left[ -x_1(\vec{R}_j) + x_2(\vec{R}_j) \right] - \frac{4\pi p}{V_c} \sum_{j \neq 0} \mathfrak{F}(\vec{G}) \frac{G_z^2}{G_x^2} \exp \left( -\frac{G_x^2}{4\eta^2} \right) + \frac{4\pi \eta^4}{3\sqrt{\pi}}.
\]  (2)

In this equation, \( x_1 \) and \( x_2 \) are two coefficients, given by

\[
x_1(\vec{r}) = \frac{\text{erfc}(\sqrt{r})}{r^3} + \frac{2\eta}{\sqrt{\pi}r} \exp(-\eta^2 r^2),
\]

\[
x_2(\vec{r}) = \frac{3 \text{erfc}(\eta r)}{r^3} + \left( \frac{4\eta^4}{\sqrt{\pi}r^2} + \frac{6\eta}{\sqrt{\pi}r^4} \right) \exp(-\eta^2 r^2),
\]

where \( \text{erfc}(\eta r) \) is the complementary error function, and \( \eta \) an adjustable parameter making the summation converge rapidly. In Eq. (2), \( R \) and \( G \) denote the lattice vector and the reciprocal lattice vector, respectively, \( \vec{R} = \ell(q^{-1/2} \hat{x} + q^{-1/2} m \hat{y} + q n \hat{z}), \)

\[
\vec{G} = \frac{2\pi}{\ell} \left( q^{-1/2} a \hat{x} + q^{-1/2} v \hat{y} + q^{-1/2} w \hat{z} \right),
\]

where \( l, m, n, u, v, w \) are integers. In addition, \( x_j \) and \( R_j \) in Eq. (2) are given by, respectively,

\[
x_j = l - \frac{j - 1}{2}, \quad R_j = |\vec{R} - \frac{j - 1}{2} (a \hat{x} + a \hat{y} + c \hat{z})|,
\]

and the structure factor \( \mathfrak{F}(\vec{G}) = 1 + \exp[i(u + v + w)/\pi]. \)

So far, based on the result of the local-field (Eq. (2)), we can derive the dipole factor of the particle \( b^* \) in the lattice via a self-consistent method, which admits

\[
b^* = \frac{b}{1 - b \beta^*},
\]  (3)

where \( \rho \) stands for the volume fraction of the particles, and \( \beta^* = 3EV_c/4\pi p \) the local-field factor, which was measured in computer simulations by Martin et al. [13,14].

Let us use \( \beta^*_x \) and \( \beta^*_y \) to denote the local-field factors parallel and perpendicular to the uniaxial anisotropic axis, which is actually a function of the degree of anisotropy \( q \). Note that \( \beta^*_x = \beta^*_z = 1 \) for bcc lattice, namely \( q = 1 \). Furthermore, there exists a sum rule for \( \beta^*_x \) and \( \beta^*_y \), namely \( 2\beta^*_x + \beta^*_y = 3 \). In Eq. (3), \( b \) denotes the dipole factor for an isolated particle, which is given by the known form

\[
b = \frac{\tilde{\epsilon}_1 - \tilde{\epsilon}_2}{\tilde{\epsilon}_1 + 2\tilde{\epsilon}_2},
\]  (4)

where the complex dielectric constant \( \tilde{\epsilon}_j = \epsilon_j + \sigma_j/2\pi f \), with \( j = 1, 2 \) for the particle and host, respectively. Here, \( \epsilon \) denotes the real dielectric constant, \( \sigma \) the conductivity, and \( i = \sqrt{-1} \).

For simplifying the study, let us express the dipole factor [Eq. (3)] in the spectral representation. Actually, the spectral representation approach [15] is a rigorous method, which separates the material parameters from the geometrical structure parameters, thus simplifying the study [16,17]. This approach was founded originally for two-component composites, but an extension to three-component composites was done as well [18].

For recasting the dipole factor into the spectral representation, we should introduce a material parameter \( \tilde{s} = (1 - \tilde{\epsilon}_1/\tilde{\epsilon}_2)^{-1} \). The substitution of it into Eq. (3) yields

\[
b^* = \frac{F_1}{\tilde{s} - s_1},
\]  (5)

where the residue \( F_1 = -1/3 \), and the pole \( s_1 = (1 - \rho \beta^*)/3 \). For isolated sphere case, i.e., \( \rho \to 0 \), \( s_1 = 1/3 \), as expected.

Then we introduce two (real) material parameters, i.e., the real dielectric constant ratio and conductivity ratio, as

\[
s = (1 - \epsilon_1/\epsilon_2)^{-1}, \quad t = (1 - \sigma_1/\sigma_2)^{-1},
\]

and after some manipulations we obtain

\[
b^* = \frac{F_1}{s - s_1} + \frac{\Delta b}{1 + if/f_c},
\]  (6)

where the dispersion strength \( \Delta b \) and the characteristic frequency \( f_c \) [at which the dielectric dispersion occurs for the force \( F(f) \) are, respectively, given by

\[
\Delta b = \frac{F_1}{t(s - s_1)(s - s_1)},
\]  (7)

\[
f_c = \frac{1}{2\pi} \frac{\sigma_2 s(t - s_1)}{t(s - s_1)}.
\]  (8)

Eq. (6) is the main result in the present Letter, which takes into account the local-field effect due to all the other particles in the lattice of interest and is expressed in the spectral representation exactly in terms of the dispersion strength with respect to the characteristic frequency.
We are now in a position to do some numerical calculations. Fig. 1a shows the dependence of the local-field factor on the degree of anisotropy $q$. It is evident to find that $\beta_1' (\beta_2')$ is caused to increase (decrease) for increasing $q$. And a plateau is shown at $\beta_1' = \beta_2' = 1$, which actually includes the transformations ranging from the bcc ($q = 1$) lattice to the fcc ($q = 2^{1/3}$). Accordingly, similar plateau occurs at other parameters (e.g., $s_1, \Delta b, f_c$) due to their dependence on the local-field factor. In Fig. 1b, we investigate the pole $s_1$ as a function of $q$. It is shown that increasing $q$ causes $s_1$ ($s_2$) to decreases (increase). Moreover, larger volume fraction yields a smaller pole. In fact, $\rho = 0.02$ indicates a dilute limit case, hence its pole appears near $1/3$ (which is known for the isolated particle), as expected. Fig. 1c displays the effect of $q$ on the dispersion strength $\Delta b$. We find that $\Delta b_1$ ($\Delta b_2$) is caused to increase (decrease) as $q$ increases. From this figure, it can be concluded that the corresponding force acting on the particle should be caused to increase (decrease) accordingly by the increasing $q$ (this is just verified in Fig. 2). Also, for both the dispersion strengths $\Delta b_1$ and $\Delta b_2$, larger $\rho$ yields larger $\Delta b$. The same effect can be shown for the force as well (no figures shown here). We investigate the characteristic frequency $f_c$ in Fig. 1d. Increasing $q$ causes $f_{cx}$ (or $f_{cz}$) to decrease (or increase), as shown in Fig. 2 as well. In addition, for a larger volume fraction, a smaller $f_c$ can also be achieved.

In Fig. 2, we plot the force which is given by the real part of the corresponding dipole factor $\text{Re}[h_z^s]$ (Fig. 2a) and $\text{Re}[h_z^c]$ (Fig. 2b), as a function of the field frequency $f$. Note that the curves predicted by $q = 1.0$ (bcc) and $2^{1/3}$ (fcc) are overlapped, as already predicted in the spectral representation, see Fig. 1. It is shown that the local-field has a significant effect on the force by comparing the isolated particle case with the other nonisolated particle, and that the local-field enhances the force significantly. On the other hand, increasing $q$ causes $\text{Re}[h_z^s]$ to increase, and it, however, has exactly the opposite effect for $\text{Re}[h_z^c]$. Such results has been well understood with the aid of the spectral representation (see Fig. 1).

So far, we have discussed the force acting on a microparticle in the model ER solid which is subject to the structure transformation. Based on the Ewald–Kornfeld formulation, the dipole factor for a particle was analytically derived by including the local-field effect arising

![Fig. 1](image-url)

Fig. 1. (a) Dependence of the local-field factor $\beta'$ on the degree of anisotropy $q$; (b)-(d) Dependence of the pole $s_1$, the dispersion strength $\Delta b$ and the characteristic frequency $f_c$ on $q$, respectively, for various volume fraction $\rho$ at $\sigma_2 = 2.8 \times 10^{-9}$ S/m, $t = -1/90$, $\epsilon_1 = 2.25\epsilon_0$, and $s = -0.045$ (i.e., $\epsilon_1 = 52.25\epsilon_0$). Here, $\epsilon_0$ denotes the dielectric constant of free space. For clarity, the bct (ground state), bcc and fcc lattices which are, respectively, related to $q = 0.87358$, $q = 1.0$ and $q = 2^{1/3}$ are also shown (dotted lines). Empty symbols denote the $x$- (or $y$-) component of the corresponding physical quantities, and solid symbols the $z$-component. In (b)-(d): circles, $\rho = 0.02$; triangles, $\rho = 0.1$; squares, $\rho = 0.2$. 
from all the other particles, and exactly expressed in the
spectral representation in terms of the dispersion
strength with respect to the characteristic frequency,
thus simplifying the study.

As the particles are located closely, the multipolar
interaction between them is expected to play a role
[19,20]. In this regard, it is of value to extend the present
work to investigate the effect of the multipolar interac-
tion. In addition, it is also interesting to see what hap-
pens if one takes into account the present lattice effect
on the nonlinear alternating current responses [3] of ER
solids.

To sum up, our results have shown that the force
acting on a particle in the ER solid due to the nonuni-
form ac electric field can be affected significantly by the
structure transformation, and local-field effect as well as
field frequency. Our results are very well understood by
using of the spectral representation theory. Thus, it
is possible to monitor the structure of ER solids by
detecting this force.

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