Structure-induced nonlinear dielectric properties in electrorheological fluids

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The structure-induced nonlinear dielectric properties of electrorheological (ER) fluids have been studied in anhydrous $KNbO_3$–silicone oil and $SrTiO_3$–silicone oil systems. We find that the permittivities of ER fluids increase linearly with the increase of the field strength as the electric field exceeds a threshold value E_1, and tend to saturated constants beyond a high field strength E_2. The alignment of particles in the direction of the electric field causes variation of the dielectric properties of the ER fluids. Below E_1, when the particles are randomly dispersed, and beyond E_2, when almost all the particles are form chains and columns, the permittivities do not vary with E and attain their lowest and highest values, respectively. Correspondingly, the current densities of ER fluids no longer follow Ohm's law between E_1 and E_2, whereas they still show Ohmic behavior in the regions below E_1 and beyond E_2. [S1063-651X(97)05302-6]

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I. INTRODUCTION

An electrorheological (ER) fluid consists of fine solid particles suspended in an insulating oil. The effective viscosity of an ER fluid increases dramatically and the particles form chains and columns when an electric field is applied and exceeds a critical value. These phenomena occur rapidly and are reversible. Much attention has been attracted to study the properties and the physical mechanism of ER fluids [1–6]. The basic issues are the field-induced interactions and behaviors of the particles in the fluids, which are related to the dielectric properties of these fluids. Although some studies of dielectric properties of ER fluids have been reported in literature, these mostly involve measurements under low electric field. However, the ER effect usually appears at high fields when rearrangement of the particles occurs. On the other hand, the treatment of the dielectric properties in conventional heterogeneous two-phase mixture systems cannot be used here any longer, because the particles are movable and may aggregate in ER fluids. The most commonly noticed nonlinear electric property is that the current density increases more rapidly with the field strength E than would be expected in Ohmic behavior [6–8]. To date the explanation of this phenomenon is still insufficient. Block *et al.* [9] observed that the permittivity and dielectric loss were larger at high field than those at low field, and pointed out that the behavior was due to the fibrillation of particles in the field direction. Conrad *et al.* [10] found that the degree of alignment of the particles in the field direction increased with the field and then leveled off with further increase in the field. However, the relationship between the particle arrangement in the fluid and the nonlinear dielectric properties has not been established.

We have studied the nonlinear dielectric properties of some anhydrous ER fluids and found that the nonlinear behavior is mainly caused by the rearrangement of the particles in the fluids. The measured results show that the permittivities of ER fluids increase linearly with the increase of the field strength when the electric field exceeds a threshold value E_1, and then tend to saturated constants beyond a high field strength E_2. Below E_1 and above E_2, the permittivities

are constant at their lowest and highest values, respectively. Correspondingly, the current density no longer follows Ohm's law in the region between E_1 and E_2, while Ohm's law is obeyed outside this region. A model based on the fact that the alignment of the particles in the electric field direction causes the permittivity to increase is proposed to explain the anomalous electric properties in ER fluids. The analysis is consistent with the measured results.

II. PERMITTIVITY AND CONDUCTIVITY OF A HETEROGENEOUS MIXTURE

The permittivity of a heterogeneous mixture has been studied by various authors [11]. In the case of spherical particles distributed in a medium, a useful formula for the permittivity was given by Looyenga [12]. The permittivity of an ER fluid under an electric field lower than the threshold value, in which the particles are randomly distributed, can be described approximately by Looyenga's formula:

$$\varepsilon_1^{\text{spher}} = [\varepsilon_f^{1/3} + \phi(\varepsilon_p^{1/3} - \varepsilon_f^{1/3})]^3, \tag{1}$$

where ε_p and ε_f are the permittivities of the particles and fluid, respectively, and ϕ is the volume fraction of the particles in the fluid.

Any aggregation of the particles along the direction of the electric field must lead to the increase of the permittivity of the system. This can be schematically illustrated as Fig. 1 and described as follows. For simplicity, we assume that all the particles dispersed in the oil are cubic. In Fig. 1(a), the particles are homogeneously distributed, and in Fig. 1(b), the particles are totally aligned in the electric-field direction. The permittivity of the system can be derived by regarding the admittance as the sum of all resistances and capacitances of solid and liquid blocks in series and in parallel, respectively.

The permittivity in the high frequency limit ($\omega \rightarrow \infty$) in the homogeneously distributed case is

$$\varepsilon_{1,\infty} = \varepsilon_f \frac{\varepsilon_p + (\phi - \phi^{1/3})(\varepsilon_p - \varepsilon_f)}{\varepsilon_p - \phi^{1/3}(\varepsilon_p - \varepsilon_f)}, \tag{2}$$
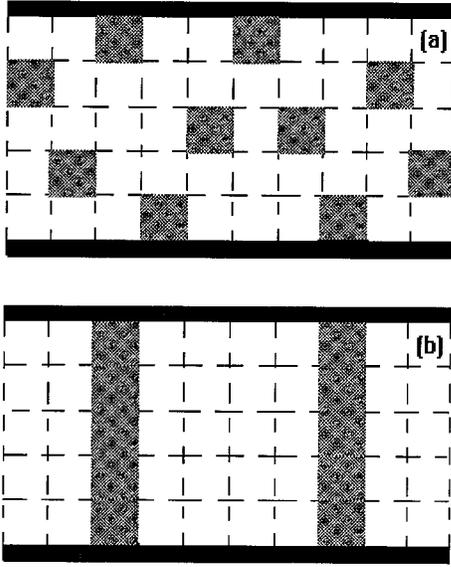
and in the aligned case is

FIG. 1. Schematic arrangement of cubic particles dispersed in a medium between two electrodes. (a) Randomly distributed, (b) aligned in the field direction.

$$\varepsilon_{2,\infty} = \phi\varepsilon_p + (1-\phi)\varepsilon_f. \qquad (3)$$

On the other hand, the conductivity σ in a dc field ($\omega=0$) in the homogeneously distributed case has an identical behavior with $\varepsilon_{1,\infty}$, namely

$$\sigma_1 = \sigma_f \frac{\sigma_p + (\phi-\phi^{1/3})(\sigma_p-\sigma_f)}{\sigma_p - \phi^{1/3}(\sigma_p-\sigma_f)}. \qquad (4)$$

In the aligned case, the conductivity is

$$\sigma_2 = \phi\sigma_p + (1-\phi)\sigma_f. \qquad (5)$$

The derivations of Eqs. (2) to (5) are given in the Appendix. We should emphasize that a certain percentage of oil in the chains must contribute to the calculations in the aligned case, i.e., Eq. (3) and Eq. (5) are not appropriate for quantitatively describing the behavior, although they are commonly accepted in the literature [13]. More reliable representations are given in the Appendix, and a discussion will be made in a later part of this paper.

Our treatment with the cubic model is more intuitive. The behaviors of ε and σ changing with ϕ, described by Eqs. (1) to (5), are plotted in Fig. 2.

In Fig. 2, curve (1) and curve (2) show the differences of the spherical particle model and the cubic particle model in the homogeneously distributed case, while curve (3) shows the permittivity and the conductivity of the system changing linearly with the volume fraction ϕ in the aligned case. It can be seen that for a certain volume fraction ϕ the permittivity ε or the conductivity σ of the system must increase when the particles are arranged in the electric-field direction.

III. EXPERIMENTAL DETAILS

In the measurement of the permittivities of ER fluids, an ac high voltage power supply with sinusoidal voltage output was applied. Two parallel electrodes filled with an ER fluid
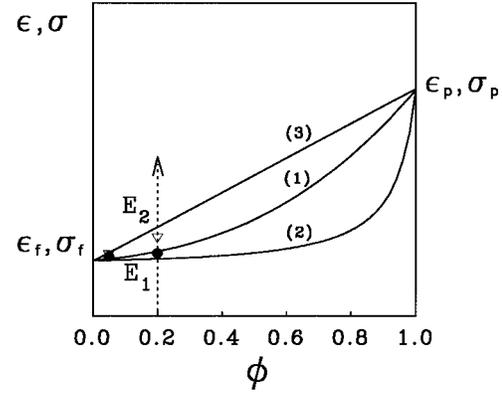

FIG. 2. Relation between the permittivity or conductivity and the volume fraction in a heterogeneous mixture in two extreme regions $E \leqslant E_1$ and $E \geqslant E_2$. The filled and hollow symbols represent the experimental results of the KNbO$_3$–silicone oil system at $\phi=0.05$ and 0.2 in these two regions, respectively. The solid lines are the theoretical values. The arrow indicates increase of the electric field strength. (1) Random distribution with spherical particle model in the region $E \leqslant E_1$, (2) random distribution with cubic particle model in the regime $E \leqslant E_1$, (3) particles aligned in the field direction in $E \geqslant E_2$.

constituted a capacitor in which the permittivity of the ER fluid was obtained by measuring the capacities. The currents passing through the ER fluid were calculated by measuring the voltage on a resistance in series with the capacitor. The impedance of the resistance was three orders smaller than that of the capacitor. Special care was taken to avoid errors resulting from ac-circuit resonance effects. All digital multimeters and oscilloscopes applied in the experiments had high enough input impedances to ensure the reliability of all measured values.

Measurements were performed on two kinds of silicone oil ER fluids, one containing KNbO$_3$ particles and the other SrTiO$_3$ particles. Particles of average size 35 μm were prepared by grinding single crystals and drying above 200 °C in an oven to evaporate any water. At room temperature KNbO$_3$ crystal is a ferroelectric material while SrTiO$_3$ is not. The volume fractions of the particles for both KNbO$_3$ and SrTiO$_3$ in the oil were 0.05 and 0.2. The ER fluid container was installed in a silicone oil bath, the temperature of which could be controlled. Each value of the permittivity and the current density at different field strength was measured after the electric field had reached a fixed value for 20 s. All the current densities and field strengths were mean effective values measured under a 50-Hz ac field at room temperature.

IV. RESULTS AND DISCUSSIONS

The dependences of the current density and the permittivity of the KNbO$_3$–silicone-oil ER fluid on an electric field are shown in Fig. 3(a) and Fig. 3(b), while those of the SrTiO$_3$–silicone oil system are shown in Fig. 4(a) and Fig. 4(b), respectively. It is obvious that in a certain range of the electric field the permittivity increases linearly with the increasing field and the current density behaves nonlinearly in this region. In Fig. 3, the region is from about $E_1=50$ V/mm to $E_2=2800$ V/mm in the case of $\phi=0.2$, while E_1 and E_2
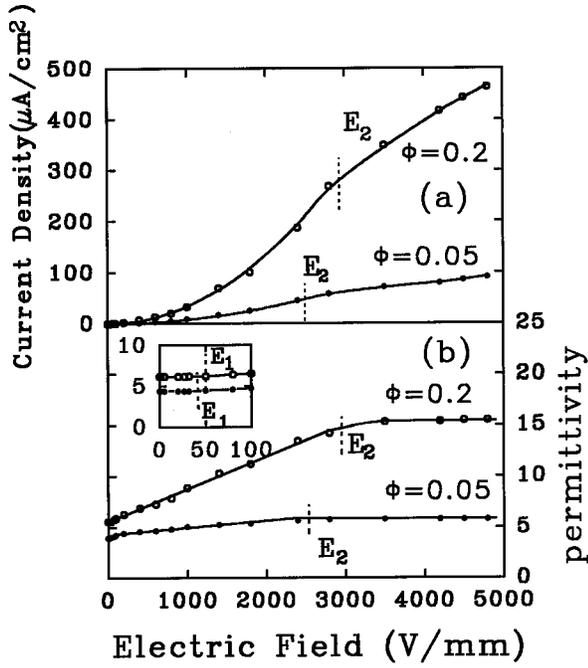
FIG. 3. Relation of (a) the current density and (b) the permittivity versus field strength in KNbO$_3$–silicone oil ER fluids (ϕ=0.05 and 0.2). Inset shows the permittivity in the low field region. The solid lines across the symbols are guides to the eye.

are lower at ϕ=0.05. Below E_1, the permittivities are constants, and beyond E_2, they tend to saturated values. This fact is also explicit in the SrTiO$_3$–silicone oil system as shown in Fig. 4, where E_1 and E_2 are about 45 V/mm and 450 V/mm, respectively. In Fig. 3(a) and Fig. 4(a), the values of E_1 are not shown because they are hardly detectable.
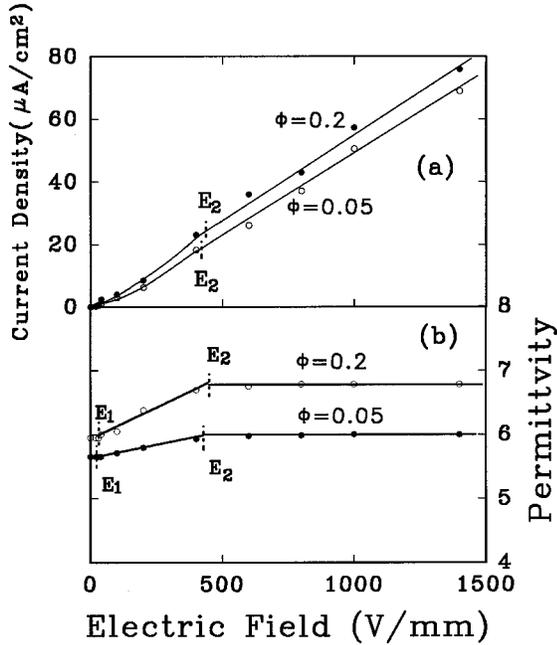

FIG. 4. Relation of (a) the current density and (b) the permittivity versus field strength in SrTiO$_3$–silicone oil ER fluids (ϕ=0.05 and 0.2). The solid lines across the symbols are guides to the eye.

The variation of permittivity is mainly due to the arrangement of the particles in the oil. When the field strength is lower than E_1, the particles in the fluid are randomly dispersed without rearrangement and the permittivity of the system does not change with the electric field, i.e., it is a constant ε_1. However, when the field strength is larger than E_2, all the particles almost form chains and columns in the direction of the field. Then the permittivity reaches ε_2 and maintains this value when the field strength is further increased. Equations (1) [or (2)] and (3), as well as Fig. 2, express the distinctness of the permittivities of these two extreme situations. In the region between E_1 and E_2, more and more particles are aligned along the direction of the field as the field strength increases, which causes the permittivity to increase. All the predictions have been demonstrated in our experiments through observation of the arrangement of the particles in ER fluids with an optical microscope. The observation was made simultaneously with the measurement of the permittivity by varying the electric field. Below threshold field E_1, no rearrangement of the particles could be observed. The aggregation of the particles in the direction of the field occurred at E_1, beyond which the particles were gradually aligned with the increase of the electric field. Finally, all particles formed chains or columns at E_2, above which the pattern was fixed and no more change could be observed.

Conrad *et al.* [10] measured the degree of alignment of the particles with an orientation parameter and found that the orientation parameter increases linearly with the electric field in a certain region of E and tends to a constant for a larger field. It is reasonable to use Conrad's results to explain the linear behavior of the permittivity increase with the electric field between E_1 and E_2 observed in our measurement. A further theoretical analysis about this phenomenon is still required.

These facts demonstrate that the polarization of the ER fluid between E_1 and E_2 is no longer linear, i.e., the ER fluid permittivity is E dependent. The relation of the permittivity and electric field can be expressed as

$$\varepsilon = \begin{cases} \varepsilon_1, & E \leqslant E_1 \\ \varepsilon_1 + k(E-E_1), & E_1 < E < E_2 \\ \varepsilon_2 = \varepsilon_1 + k(E_2-E_1), & E \geqslant E_2, \end{cases} \quad (6)$$

where k is a constant.

By the same analysis the conductivity of the ER fluid in a dc field is also E dependent, i.e.,

$$\sigma = \begin{cases} \sigma_1, & E \leqslant E_1 \\ \sigma_1 + k'(E-E_1), & E_1 < E < E_2 \\ \sigma_2 = \sigma_1 + k'(E_2-E_1), & E \geqslant E_2, \end{cases} \quad (7)$$

where k' is a constant different from k.

If we use the complex permittivity $\varepsilon_0(\varepsilon+\sigma/i\varepsilon_0\omega)$ in Eq. (6), the effective permittivity should be $\varepsilon_0(\varepsilon^2+\sigma^2/\varepsilon_0^2\omega^2)^{1/2}$, where ω is the angular frequency. The magnitude of the current density in the ER fluid, which consists of displacement and conductance, can be written as

$$j = \omega\varepsilon_0\left(\varepsilon^2 + \frac{\sigma^2}{\varepsilon_0^2\omega^2}\right)^{1/2} E. \quad (8)$$
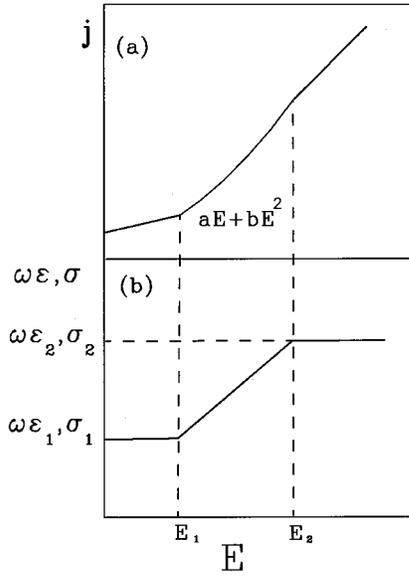
FIG. 5. Relation of (a) j and (b) $\omega\varepsilon$ or σ versus field strength.

The current density in different regions corresponding to the permittivity and conductivity can then be expressed as follows. In the high frequency ac-field limit,

$$j = \omega\varepsilon_0\varepsilon E$$
$$= \begin{cases} \omega\varepsilon_0\varepsilon_1 E, & E \leq E_1 \\ \omega\varepsilon_0[\varepsilon_1 + k(E - E_1)]E, & E_1 < E < E_2 \\ \omega\varepsilon_0\varepsilon_2 E = \omega\varepsilon_0[\varepsilon_1 + k(E_2 - E_1)]E, & E \geq E_2, \end{cases} \quad (9)$$

while in the case of a dc-field limit,

$$j = \sigma E = \begin{cases} \sigma_1 E, & E \leq E_1 \\ [\sigma_1 + k'(E - E_1)]E, & E_1 < E < E_2 \\ \sigma_2 E = [\sigma_1 + k'(E_2 - E_1)]E, & E \geq E_2. \end{cases} \quad (10)$$

Because of the nonlinear polarization or the nonlinear conductivity of the ER fluid in the region between E_1 and E_2, the E dependence of the current density behaves as $j = aE + bE^2$, where $a = \omega\varepsilon_0\varepsilon_1 - k\omega\varepsilon_0 E_1$ and $b = k\omega\varepsilon_0$ in an ac field or $a = \sigma_1 - k'E_1$ and $b = k'$ in a dc field.

A common expression for the current density can be obtained by combining Eqs. (6) to (8) and will be a little more complicated. It is obvious that the current density should also exhibit nonlinear behavior between E_1 and E_2.

In the regions below E_1 and above E_2, the current densities still follow Ohm's law, but with different slopes. The slope in the region $E \geq E_2$ is usually much larger than that in $E \leq E_1$; in other words, the current density increases with E more steeply beyond E_2 than below E_1.

The field dependence of the current density j and permittivity ε or conductivity σ are schematically plotted in Figs. 5(a) and 5(b), respectively. With this analysis the measured behaviors of the permittivity and current density shown in Figs. 3 and 4 can be well understood. A quantitative comparison between calculation and measurement is difficult because of the arbitrary shape and the inhomogeneous distribution of the particles in the oil. In practice, even at $E \geq E_2$, not all chains and columns consisting of particles are fully filled

in between electrodes, i.e., there are some chains and columns shorter than the distance between the two electrodes and there is more oil in these aligned blocks. However, in our model, by adjusting the parameter x, which represents the average percentage of oil in each chain as described in the Appendix, we can calculate the permittivity to fit the measured value of the permittivity. For example, according to Eqs. (A3) and (A9) to (A12) in the Appendix, when the electric field strength is larger than E_2, the calculated permittivities of $KNbO_3$-silicone-oil ER fluid for $\phi = 0.05$ and 0.2 are 6.103 and 15.340, respectively, corresponding to two values of x (0.01 and 0.0145), which quantitatively agree with the experimental results: 6.17 and 15.48. However, if we let $x = 0$, the calculated results would be 7.38 and 22.0, which are quite different from the measured values. This fact indicates that Eqs. (3) and (5) in the text or Eqs. (A14) and (A16) in the Appendix are not appropriate for describing the real case in which the particles are not cubic and not all the chains or columns are spanning the two electrodes. A better approximation should be Eqs. (A9) to (A12) in the Appendix. However, when the field strength is smaller than E_1, the calculated results do not fit so well as those at $E \geq E_2$. The main reason is that the particles cannot be homogeneously distributed in the oil, and their shapes are arbitrary. Nevertheless, the model we have described can explain the reason why the permittivity and the conductivity maintain linearity when $E \leq E_1$ and $E \geq E_2$ and should behave nonlinearly when $E_1 < E < E_2$.

In our experiment, because ER fluids consist of anhydrous particles and silicone oil, the relaxation is not important in the case when the field frequency is not very high. The Looyenga equation and the model we have proposed are valid. However, if ER fluids contain water or mobile ions, the relaxation problem may be serious and should be taken into account to study the nonlinear behavior. Although other factors may also contribute to the nonlinearity of the dielectric property, the structure-induced nonlinear behavior is always most important in ER fluids.

V. CONCLUSIONS

In this article the structure-induced nonlinear dielectric properties of anhydrous ER fluids have been studied. The field dependence of the permittivities and the current densities of $KNbO_3$-silicone oil system as well as $SrTiO_3$-silicone oil ER fluids were measured. The nonlinear behavior is mainly caused by the arrangement of the particles in the fluids. We have found that the permittivities of the ER fluids increase linearly with the increase of the field strength in the region between a threshold field E_1 and a critical field E_2. The enhancement of the permittivities is induced by gradual alignment of particles in the direction of the electric field. Below E_1, when the particles are randomly dispersed, and beyond E_2, when the particles almost totally form chains and columns, the permittivities do not vary with E and reach their lowest and highest values, respectively. Correspondingly, the current densities of the ER fluids behave as $j = aE + bE^2$ between E_1 and E_2, whereas they still follow Ohm's law in the regions below E_1 and beyond E_2. This analysis about the structure-induced nonlinear behavior is valid in anhydrous ER fluids and can also be applied in other
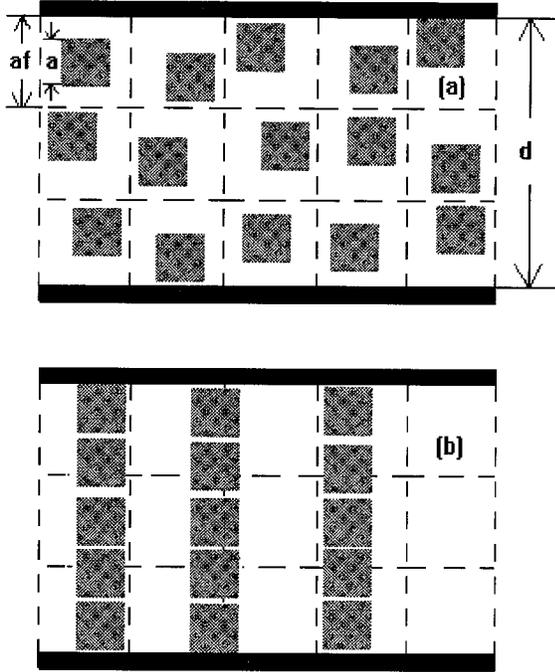
FIG. 6. Schematic arrangement of cubic particles dispersed in a medium between two electrodes. (a) Randomly distributed, (b) aligned in the direction of the electric field with a thin layer of oil between neighboring particles.

cases, although more factors should be considered.

ACKNOWLEDGMENTS

APPENDIX

Assuming that N cubic particles of side length a are randomly dispersed in the oil, the ER fluid is divided into N cubes with side length a_f, and each cube encloses a cubic particle; d is the distance between the two parallel electrodes of which the surface areas are both S [Fig. 6(a)]. The total volume of the ER fluid and the solid particles are $V = N a_f^3$ and $V_p = N a^3$, respectively. The volume fraction is $\phi = V_p / V$, and $a_f = \phi^{-1/3} a$.

The total admittance Y_t of the ER fluid can be represented as a resistance R and a capacitance C in parallel, i.e.,

$$Y_t = \frac{1}{R} + i\omega C = \sigma \frac{S}{d} + i\omega \varepsilon_0 (\varepsilon' - i\varepsilon'') \frac{S}{d}, \quad \text{(A1)}$$

where σ, ε', and ε'' are the dc conductivity, and the real and imaginary parts of the dielectric constant of the fluid, respectively. According to Fig. 6(a), the cubes are in series in the field direction, but in parallel in the plane vertical to the direction of the electric field. Assuming that both compo-

nents are loss-free, and their permittivities keep constant in the frequency region considered, then the total admittance Y_t can be rewritten

$$Y_t = \frac{S}{d}\left[\sigma + \omega\varepsilon_0 \frac{(\varepsilon_s - \varepsilon_\infty)\omega\tau}{1 + \omega^2\tau^2} + i\omega\varepsilon_0\left(\varepsilon_\infty + \frac{\varepsilon_s - \varepsilon_\infty}{1 + \omega^2\tau^2} \right) \right].$$
$$\text{(A2)}$$

Comparing Eq. (A2) with Eq. (A1) leads to the Debye equation [11]:

$$\varepsilon' = \varepsilon_\infty + \frac{\varepsilon_s - \varepsilon_\infty}{1 + \omega^2\tau^2}, \quad \text{(A3)}$$

$$\varepsilon'' = \frac{(\varepsilon_s - \varepsilon_\infty)\omega\tau}{1 + \omega^2\tau^2}, \quad \text{(A4)}$$

where

$$\varepsilon_s = \frac{\sigma_p^2\varepsilon_f - \phi^{1/3}(\sigma_p^2\varepsilon_f - \sigma_f^2\varepsilon_p)}{[\sigma_p - \phi^{1/3}(\sigma_p - \sigma_f)]^2} + (\phi - \phi^{1/3})$$
$$\times \frac{(\sigma_p^2\varepsilon_f + \sigma_f^2\varepsilon_p - 2\sigma_p\sigma_f\varepsilon_f) - \phi^{1/3}\varepsilon_f(\sigma_p - \sigma_f)^2}{[\sigma_p - \phi^{1/3}(\sigma_p - \sigma_f)]^2},$$
$$\text{(A5)}$$

$$\varepsilon_\infty = \varepsilon_f \frac{\varepsilon_p + (\phi - \phi^{1/3})(\varepsilon_p - \varepsilon_f)}{\varepsilon_p - \phi^{1/3}(\varepsilon_p - \varepsilon_f)}, \quad \text{(A6)}$$

$$\sigma = \sigma_f \frac{\sigma_p + (\phi - \phi^{1/3})(\sigma_p - \sigma_f)}{\sigma_p - \phi^{1/3}(\sigma_p - \sigma_f)}, \quad \text{(A7)}$$

and

$$\tau = \varepsilon_0 \frac{\varepsilon_p - \phi^{1/3}(\varepsilon_p - \varepsilon_f)}{\sigma_p - \phi^{1/3}(\sigma_p - \sigma_f)}. \quad \text{(A8)}$$

In the above equations ε_s and ε_∞ are the static and high-frequency dielectric constant of the ER fluid, τ is the relaxation time, while σ_p, ε_p and σ_f, ε_f are the conductivity and permittivity of the particles and the oil, respectively.

In fact, not all particles are properly enclosed by the corresponding oil cubes with side length a_f. In order to improve the precision of the model, we discard the notation of oil cube and consider that the particles also form chains such as that in the regime $E \geqslant E_2$ just with more number of chains. If y is the total thickness of the oil layers in one chain, then the number of chains m can be derived by substituting $d = na + y$ in $Sd = ma^2na/\phi$. Let $x = y/(na)$, which represents the percentage of oil occupying one chain. In terms of this parameter, the total admittance of the ER fluid can also be expressed as Eq. (A2). Consequently, one can still obtain the Debye equation with the following parameters:

$$\varepsilon_s = \frac{1}{[x\sigma_p + (1-x)\sigma_f]^2}\{x\sigma_p^2\varepsilon_f + (1-x)\sigma_f^2\varepsilon_p$$
$$+ [\phi - (1-x)][\sigma_f^2(\varepsilon_p - \varepsilon_f) + x\varepsilon_f(\sigma_p - \sigma_f)^2]\},$$
$$\text{(A9)}$$

$$\varepsilon_\infty = \varepsilon_f \frac{\varepsilon_p + [\phi - (1-x)](\varepsilon_p - \varepsilon_f)}{x\varepsilon_p + (1-x)\varepsilon_f}, \quad (A10)$$

$$\sigma = \sigma_f \frac{\sigma_p + [\phi - (1-x)](\sigma_p - \sigma_f)}{x\sigma_p + (1-x)\sigma_f}, \quad (A11)$$

and

$$\tau = \varepsilon_0 \frac{x\varepsilon_p + (1-x)\varepsilon_f}{x\sigma_p + (1-x)\sigma_f}. \quad (A12)$$

With the increase of the field strength, the particles aggregate along the field direction gradually. Thus, the number of the particles in one chain increases, whereas the the chain number and the total thickness of the oil layers (thus the parameter x) in one chain decreases. As the electric field exceeds the critical value E_2, the structure reaches its steady state and no longer changes with the external electric field.

Assuming that each chain is completely filled with particles, i.e., $x \to 0$ [Fig. 1(b)], then Eqs. (A9) to (A12) become

$$\varepsilon_s = \varepsilon_\infty = \phi\varepsilon_p + (1-\phi)\varepsilon_f, \quad (A13)$$

$$\sigma = \phi\sigma_p + (1-\phi)\sigma_f, \quad (A14)$$

and

$$\tau = \varepsilon_0 \frac{\varepsilon_f}{\sigma_f}, \quad (A15)$$

and Eqs. (3) and (4) also transform into

$$\varepsilon' = \varepsilon_\infty = \phi\varepsilon_p + (1-\phi)\varepsilon_f, \quad (A16)$$

and

$$\varepsilon'' = 0. \quad (A17)$$

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