Anisotropic dielectric properties of structured electrorheological fluids

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We present an approach to monitor the structure-induced anisotropic dielectric properties of electrorheological fluids. The particles used are made from uniform glass microspheres coated with an inner magnetic layer and an outer dielectric layer. Under an applied magnetic field, the particles are found to form columnar structures with a body-centered-tetragonal crystalline arrangement inside the columns. It is shown that the structure-induced dielectric constant can be consistently explained through first-principles calculations. © *1998 American Institute of Physics.* [S0003-6951(98)04647-6]

Electrorheological (ER) fluids, consisting of fine dielectric particles suspended in an insulating liquid, make up a two-phase system whose rheological properties are controllable through the application of an external electric field. This phenomenon has been the focus of much attention experimentally and theoretically.[1–8] It is known that in the high field limit, uniform spherical particles would aggregate into columns inside which the spheres form a body-centered-tetragonal (BCT) structure, with a corresponding increase of the dielectric constant along the field direction.[5,6,8] This is therefore a nonlinear dielectric system, with the increase in the dielectric constant dictated by the principle of minimum free energy and the accompanying structural transformation. However, the dependence of the dielectric constant of the ER fluids on the structure variation is still a topic of debate in both experimental and theoretical studies. Block *et al.*[9] observed that the permittivity and dielectric loss were larger at high fields than at low fields, and pointed out the change of dielectric properties was due to the fibrillation of particles. Recent study of the structure-induced nonlinear ER fluid properties showed a tendency of the permittivity of ER fluids to increase as the electric field was increased.[10,11] However, investigation of the anisotropically dielectric constants measured along and perpendicular to the column direction is still lacking. The reason for this is due to experimental limitation. One can imagine that if two pairs of the electrodes are mounted perpendicularly (one is used to measure the dielectric constant and another is applied with high electric field) it will lead to an electrical short once the high electric field is applied.

In this work, a unique material, denoted the electromagneto (EM) microspheres, is used to investigate the anisotropic dielectric properties of ER fluids. The EM microsphere is made of a glass core onto which a magnetic layer and a dielectric layer have been sequentially coated. Since the inner layer is ferromagnetic, the application of an *external magnetic* field will lead to the aggregation of spheres into columns aligned in the field direction, with a BCT arrangement of spheres inside the columns. The structure-induced dielectric variation and anisotropic property of ER fluids can thus be determined. In contrast to the experiment performed under high electric fields, the present test is easy and more accurate. Moreover, the microspheres used here are uniform in size so that the results may be compared directly with theoretical predictions.

The particles used in our study consist of $35\pm3\ \mu$m diam glass spheres onto which an inner magnetic nickel layer and an outer dielectric layer, such as lead zirconic titanate (PZT) or TiO_2, are coated using electroless plating and sol-gel processes.[12,13] The magnetization of each individual microsphere, μ_i, can be controlled by adjusting the thickness of the nickel layer. In our experiment, the average thickness of the nickel layer is $2.5\pm0.3\ \mu$m; the magnetization of one such sphere is calculated to be $\sim$30 emu/g. It should be emphasized here that the thickness of the nickel layer should be at least 2 μm, otherwise the magnetization of such spheres is too small for suspension in the fluid, even when a relatively strong magnetic field is applied. To obtain a strong insulating outer layer, at least five repeat coatings should be made. Finally, the prepared particles were annealed at 500 °C for 2 h. The cross-sectional SEM image of a well-coated microsphere is shown in the upper inset of Fig. 1, where it can be seen that the inner Ni magnetic layer and the outer PZT insulating layer are clearly visible. The process for TiO_2 coating is the same as the PZT case.

Figure 1 gives a schematic for the dielectric measurement. The setup consists of three separate parts: (a) a computer-controlled electromagnet whose magnetic field strength can be varied from 0 to 4000 G; (b) LCR meter (HP 4284A) used to measure the dielectric properties as the structure changes with increasing magnetic field; (c) an ER fluids cell formed by mounting a plastic stripe between two brass electrodes. The dimension of the cell is $30\times30\times2$ mm, with a 2 mm gap between the two electrodes. To observe the structure changes as a function of the magnetic field, an optical microscope is applied for *in situ* monitoring. Figure 2(a) shows randomly dispersed particles in silicone oil at zero field. As the field strength is increased the particles began to align to form chains in the field direction. With further field

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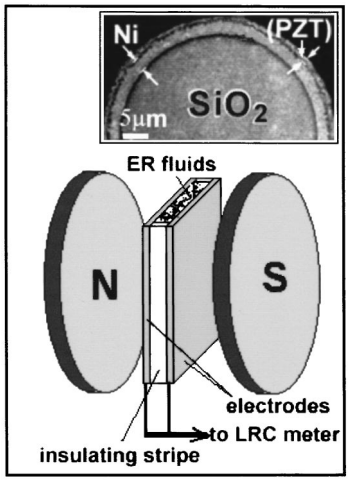

FIG. 1. Schematic diagram of the experimental setup. The upper inset shows the cross-sectional SEM imagine of double-coated microsphere.

increase the chains coarsen to form columns across the two electrodes. The coarsening process is shown in Figs. 2(b)–2(c).

Figures 3(a) and 3(b) show, corresponding to the structure changes, the measured variations of real (ϵ') and imaginary (ϵ'') parts of the dielectric constant as a function of applied magnetic field strength for two samples. In the figures, $zz(xx)$ indicates the test configuration where the two electrodes of the ER fluid cell are placed parallel (perpendicular) to the field direction. For the *zz* case, it is seen that both the real and imaginary parts of the dielectric constant *increase* monotonically as the magnetic field strength is increased from 0 to 2500 G; beyond that, there is only a slight increase. The increase in the real parts of the dielectric constant for the PZT- and TiO_2-coated ER fluids is 7% and 4%, respectively, and the imaginary parts for both cases are increased by 7%. For the *xx* case, however, both the real and imaginary parts decrease as the magnetic field strength is increased. It should be pointed out that from visual microscope observations, the columns formed when the field strength reached 150 G and beyond that almost no visual change in the column structure can be identified. We believe that the slight increase of the dielectric constant at high magnetic fields is due to the small adjustments among the particles.

To explain the experimental results described above, we use the following calculations to obtain the *zz* and *xx* components of the effect dielectric tensor. Since the ER system before the formation of the columns is a random dispersion

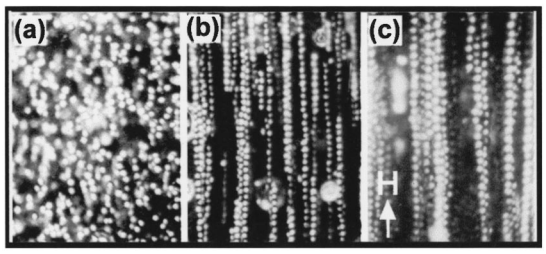

FIG. 2. Structure evolution as the magnetic field strength is increased. Here the field strengths for (a), (b), and (c) are 0, 30, and 200 G, respectively.

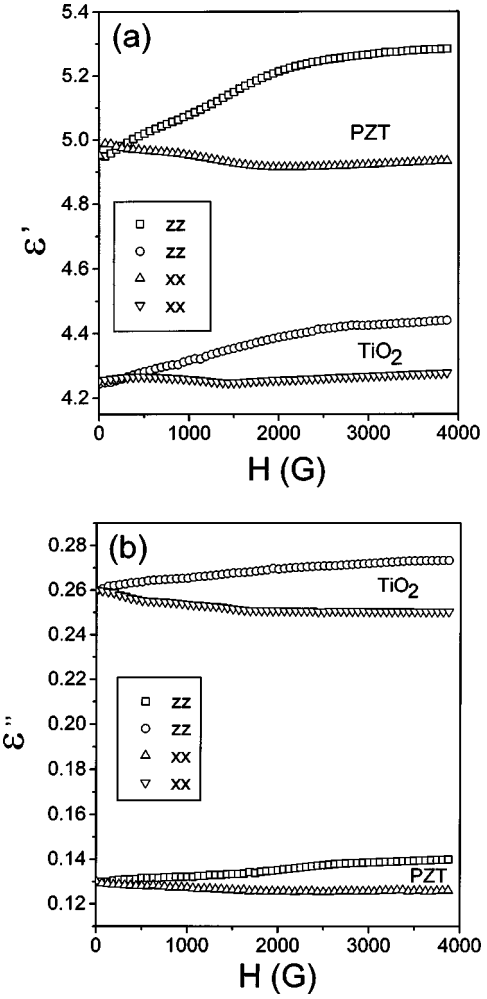

FIG. 3. Dependencies of real (a) and imaginary (b) parts of the dielectric constant on magnetic field strength. Here the volume fraction is 0.27, and the frequency of the LCR meter is fixed at 1 kHz.

of particles, its effective complex dielectric constant can be calculated by the Maxwell-Garnett formula[14]

$$\frac{\bar{\epsilon}-\epsilon_2}{\bar{\epsilon}+2\epsilon_2}=p\,\frac{\epsilon_1-\epsilon_2}{\epsilon_1+2\epsilon_2}. \tag{1}$$

Here the subscripts 1, 2 stand for particle and liquid, respectively, $\bar{\epsilon}$ is the effective dielectric constant, p is the volume fraction of solid particles. From the measured effective dielectric constant (with the frequency of the LCR meter fixed at 1 kHz) of the mixture and the dielectric constant of the oil (2.71), one can deduce from the above formula the dielectric constant of the solid particle, ϵ_1. In the high electric field limit, the particle structure which gives the largest effective dielectric constant is the body-centered-tetragonal (BCT) arrangement, which has been shown in previous theoretical and experimental studies.[5,6,8] The question here is: can we treat the particle structure in the present work as the BCT structure? Fortunately, the answer is positive and it has been proved recently, through both theoretical calculations and experimental observations.[15] Based on this consideration, we calculate the high field dielectric constant as follows.

Since particles form columns along the *z* direction, the *zz* component of the effective dielectric constant is given by

TABLE I. Experimental and theoretical results of the real and imaginary parts of ER fluid dielectric constants measured along *z* and *x* directions for the random and structured cases.

		Theory		Experiment	
		ϵ'	ϵ''	ϵ'	ϵ''
Random		Fitted	Fitted	4.95	0.13
	zz	5.67	0.23	5.29	0.14
PZT- column					
	xx	4.75	0.11	4.94	0.12
Random		Fitted	Fitted	4.26	0.26
	zz	4.56	0.37	4.44	0.28
TiO_2- column					
	xx	4.15	0.23	4.25	0.25

$$\bar{\epsilon}_{zz}=\frac{p}{p_c}\,\epsilon_{czz}+\left(1-\frac{p}{p_c}\right)\epsilon_2. \tag{2}$$

Here ϵ_{czz} is the *zz* component of the effective dielectric constant of the column, and p_c is the volume fraction inside the column. The component ϵ_{czz} can be calculated from first principles when uniform-sized spheres form regular structures, e.g., the BCT. Considering the dispersions of the sphere size in this experiment, we did the first principle calculations with uniform spheres but kept the spheres slightly separated, the separation between spheres being 0.15 (here 0.15 is chosen to fit our experimental results in the whole fitting process) of the sphere diameter. The method used in our calculation is the eigenfunction expansion method of the Bergman-Milton approach, the details of which are described in Refs. 8 and 16. The calculated results can be represented accurately by the formula

$$\epsilon_{czz}=\epsilon_2\left(1-\frac{0.4509}{s-0.1613}-\frac{0.0058}{s-0.4187}-\frac{0.0006}{s-4481}-\frac{0.0018}{s-0.4959}\right), \tag{3}$$

which is a truncated form of the Bergman-Hilton representation. Here $s=\epsilon_2/(\epsilon_2-\epsilon_1)$. The *xx* component of the effective dielectric constant can be calculated by the two dimensional Maxwell-Garnett formula

$$\frac{\bar{\epsilon}_{xx}-\epsilon_2}{\bar{\epsilon}_{xx}+\epsilon_2}=\frac{p}{p_c}\,\frac{\epsilon_{cxx}-\epsilon_2}{\epsilon_{cxx}+\epsilon_2} \tag{4}$$

and the calculated ϵ_{cxx} can be approximated by the formula

$$\epsilon_{cxx}=\epsilon_2\left(1-\frac{0.4351}{s-0.1741}-\frac{0.0157}{s-0.3944}-\frac{0.0019}{s-4451}-\frac{0.0050}{s-0.4972}-\frac{0.0013}{s-5456}\right). \tag{5}$$

In Table I we give a comparison of measured and calculated results. The agreement is satisfactory considering the idealized models used in our calculations. However, the observed increase in ϵ_{czz} is invariably smaller than the theoretical prediction. This could be due to imperfection in the crystalline BCT structure.

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