

# Theory of Electrical Conductivities of Ferrogels

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Conductive organic polymers can be formulated with polymers that incorporate fine dispersed metallic particles. In this work, we present a general model for ferrogels which are chemically cross-linked polymer networks swollen with a ferrofluid. Our aim is to study the effect of the shape and/or material (conductivity) anisotropy on the effective electrical conductivity of the ferrogel in the presence of an external magnetic field. Our theory can reproduce the known results and provides a link between the particle properties and orientation distribution and the effective electrical conductivity. To this end, we find that material (conductivity) anisotropies are more important for yielding a high effective electrical conductivity than shape anisotropies, while magnetic fields can offer a correction.

## I. Introduction

Conductive polymers<sup>1</sup> have received much attention due to their extensive industrial applications (for a review, see ref 2), such as in nonlinear optical waveguides, vapor-phase detectors, and twisted nematic liquid crystal displays. In fact, the conductive organic polymers<sup>1</sup> can be formulated with polymers that incorporate conductive additives such as carbon black<sup>2</sup> or fine dispersed metallic particles.<sup>3</sup> This is because the presence of certain additives in the polymer can enhance the effective electrical conductivity. In particular, recently, organic polymer composites filled with conductive metallic particles received much attention in scientific research.<sup>4–6</sup> For such polymer composites, their electrical characteristics are close to those of metals, whereas their mechanical properties and processing methods are typical for plastics.<sup>3–6</sup> The effective electrical conductivity of a metal–polymer composite should depend on the conductivity of particles, the particle shape, the volume fraction of particles, and the spatial orientation or alignment of particles in the composite.

Ferrogels<sup>7–10</sup> are a new class of magneto-controlled elastic materials, which are chemically cross-linked polymer networks swollen with a ferrofluid. A ferrofluid<sup>11</sup> is a colloidal dispersion of monodomain ferromagnetic particles. In the ferrogel, the finely distributed ferromagnetic particles are located in the swelling liquid and attached to the flexible network chains by adhesive forces. In the absence of an applied field, the magnetic moments are randomly oriented, and thus, the gel has no net magnetization. If an external field is applied, the magnetic moments tend to align with the field, thus yielding a bulk magnetic moment. In case of ordinary field strengths, the tendency of the dipole moments to align with the applied field is partially overcome by thermal agitation. As the strength of the field increases, all the particles eventually align their moments along the direction of the field, and as a result, the magnetization saturates. If the field is turned off, the magnetic dipole moments quickly randomize, and thus, the bulk magnetization is again reduced to zero. In a zero magnetic field, a ferrogel presents a mechanical behavior very close to that of a swollen network filled with nonmagnetic colloidal particles. In uniform magnetic fields, a ferrogel experiences no net force, and hence, no macroscopic

shape changes and motion are observed. Ferrogels have many applications ranging from soft actuators, micromanipulators, and artificial muscles<sup>7</sup> to cancer therapy<sup>8</sup> or an apparatus for immunoblotting.<sup>9</sup>

Anisotropy is a common phenomenon in most materials, and it can be an intrinsic material property or induced by the application of fields. Also, in real applications, the shape of particles may deviate from a perfect spherical shape during fabrication. In ferrogels, due to the anisotropic particle shape and/or conductivity of the ferromagnetic particles, the specific spatial orientation of the particles can be realized by using an external magnetic field. Thus, the electric properties of ferrogels can be controlled by reorientation of the particles in an applied magnetic field.

In this paper, we shall focus on an anisotropic ferrogel in which both the particle shape and conductivity possess a tensorial form. Our aim is to study the effects of the shape and/or material (conductivity) anisotropy on the effective electrical conductivity of the ferrogel, by taking into account the spatial orientation of the particles, which can be affected by the external magnetic field. To this end, we find both the shape anisotropy and the external magnetic field can cause the effective electrical conductivity to increase. Furthermore, the material (conductivity) anisotropy can increase the effective electrical conductivity significantly.

This paper is organized as follows. In section II, we put forth a general model for ferrogel containing ellipsoidal ferromagnetic particles with anisotropic conductivity. In section III, we numerically calculate two cases, namely, spherical particles with anisotropic conductivity and spheroidal particles with isotropic conductivity, in an attempt to focus on the influences of the shape and material anisotropy, respectively. This paper ends with a discussion and conclusion in section IV.

## II. A General Model for Ellipsoidal Particles with Anisotropic Conductivities

Let us start by considering an ellipsoidal particle with depolarization factors  $L_x$ ,  $L_y$ , and  $L_z$  along the  $x$ ,  $y$ , and  $z$  axes, respectively. Namely, the depolarization factor has a tensor form

$$\tilde{L} = \begin{pmatrix} L_x & 0 & 0 \\ 0 & L_y & 0 \\ 0 & 0 & L_z \end{pmatrix} \quad (1)$$

The three components satisfy a sum rule,  $L_x + L_y + L_z = 1$ .<sup>12</sup> In the case of crystalline anisotropy, the conductivity of the particle has the following tensor form

$$\tilde{\sigma}_1 = \begin{pmatrix} \sigma_{11} & 0 & 0 \\ 0 & \sigma_{22} & 0 \\ 0 & 0 & \sigma_{33} \end{pmatrix} \quad (2)$$

The particles are embedded in a host with a scalar conductivity,  $\sigma_2$ . In this paper, the principal axes of  $\tilde{\sigma}_1$  are assumed to be parallel to the geometric axes of the ellipsoidal particle. The orientation of the conductivity tensor,  $\tilde{\sigma}_1$ , differs from inclusion to inclusion, such that  $\tilde{\sigma}_1$  is transformed to

$$\tilde{\tilde{\sigma}}_1 = \tilde{v} \tilde{\sigma}_1 \tilde{v}^T \quad (3)$$

in a common coordinate system, where  $\tilde{v}$  denotes the rotation matrix of a particle. For the volume average of the field of the whole system,  $\langle \mathbf{E} \rangle$ , one has<sup>13</sup>

$$p\langle \mathbf{E}_1 \rangle + (1-p)\langle \mathbf{E}_L \rangle = \langle \mathbf{E} \rangle \quad (4)$$

where  $p$  stands for the volume fraction of particles and  $\langle \mathbf{E} \rangle$  is equal to the applied field,  $\mathbf{E}_0$ , under appropriate boundary conditions.<sup>14</sup> Here,  $\langle \cdot \cdot \cdot \rangle$  denotes the volume average of  $\cdot \cdot \cdot$ .  $\langle \mathbf{E}_L \rangle$  represents the volume average of the Lorentz field (namely, local field in the vicinity of the particles) which includes the contribution from the dipole moments of all the other particles, and  $\langle \mathbf{E}_1 \rangle$  represents the volume average of the local field inside the particles. Thus, we can rewrite eq 4 as<sup>13,14</sup>

$$p\langle \mathbf{E}_1 \rangle + (1-p)\langle \mathbf{E}_L \rangle = \mathbf{E}_0 \quad (5)$$

Solving the electrostatic equation  $\nabla^2 \Phi_1 = 0$  ( $\Phi_1$  denotes the electrical potential inside the particles), we obtain  $\langle \mathbf{E}_1 \rangle$  such that<sup>13</sup>

$$\langle \mathbf{E}_1 \rangle = \langle \tilde{\tilde{\rho}} \rangle \langle \mathbf{E}_L \rangle \quad (6)$$

with

$$\langle \tilde{\tilde{\rho}} \rangle = \langle \tilde{v} \tilde{\rho} \tilde{v}^T \rangle \quad (7)$$

Here

$$\tilde{\rho} = \begin{pmatrix} \frac{\sigma_2}{L_x \sigma_{11} + (1-L_x)\sigma_2} & 0 & 0 \\ 0 & \frac{\sigma_2}{L_x \sigma_{22} + (1-L_x)\sigma_2} & 0 \\ 0 & 0 & \frac{\sigma_2}{L_x \sigma_{33} + (1-L_x)\sigma_2} \end{pmatrix} \quad (8)$$

On the other hand, we average the electric current density,  $\langle \mathbf{J} \rangle$ , over the volume of the whole system and obtain

$$\langle \mathbf{J} \rangle = p\langle \tilde{\tilde{\sigma}}_1 \rangle \langle \mathbf{E}_1 \rangle + (1-p)\sigma_2 \langle \mathbf{E}_L \rangle \quad (9)$$

Further, the effective electrical conductivity,  $\tilde{\sigma}_e$ , of the ferrogel can be defined as

$$\tilde{\sigma}_e = \frac{\langle \mathbf{J} \rangle}{\langle \mathbf{E} \rangle} = \frac{\langle \mathbf{J} \rangle}{\mathbf{E}_0} \quad (10)$$

In view of eqs 5, 6, and 9, we obtain

$$\tilde{\sigma}_e = \frac{p\langle \tilde{\tilde{\sigma}}_1 \rangle \langle \tilde{\tilde{\rho}} \rangle + (1-p)\sigma_2 \tilde{I}}{p\langle \tilde{\tilde{\rho}} \rangle + (1-p)\tilde{I}} \quad (11)$$

which can be expressed in a tensorial form such that

$$\tilde{\sigma}_e = \begin{pmatrix} \sigma_{e11} & 0 & 0 \\ 0 & \sigma_{e22} & 0 \\ 0 & 0 & \sigma_{e33} \end{pmatrix} \quad (12)$$

In eq 11,  $\tilde{I}$  represents a unit matrix.

Equation 11 is a nontrivial equation, which can reproduce many other known results. For instance, setting  $L_x = L_y \neq L_z$  and  $\sigma_{11} = \sigma_{22} = \sigma_{33}$  and seeing the particles *randomly* oriented in a host,  $\sigma_{e11}$  obtained from eq 11 is identical to eq 14 of ref 15. In this case, if the metallic particles are subjected to an external magnetic field (i.e., no longer randomly oriented),  $\sigma_{e11}$  ( $=\sigma_{e22}$ ) and  $\sigma_{e33}$  predicted by eq 11 are identical to eqs 3 and 4 of ref 16, respectively. Finally, for spherical particles (namely,  $L_x = L_y = L_z$ ) with a scalar conductivity (i.e.,  $\sigma_{11} = \sigma_{22} = \sigma_{33}$ ), eq 11 can produce the expression for the well-known Maxwell–Garnett theory,<sup>17,18</sup> namely,

$$\sigma_e = \sigma_2 + \frac{3p\sigma_2(\sigma_1 - \sigma_2)}{3\sigma_2 + (1-p)(\sigma_1 - \sigma_2)} \quad (13)$$

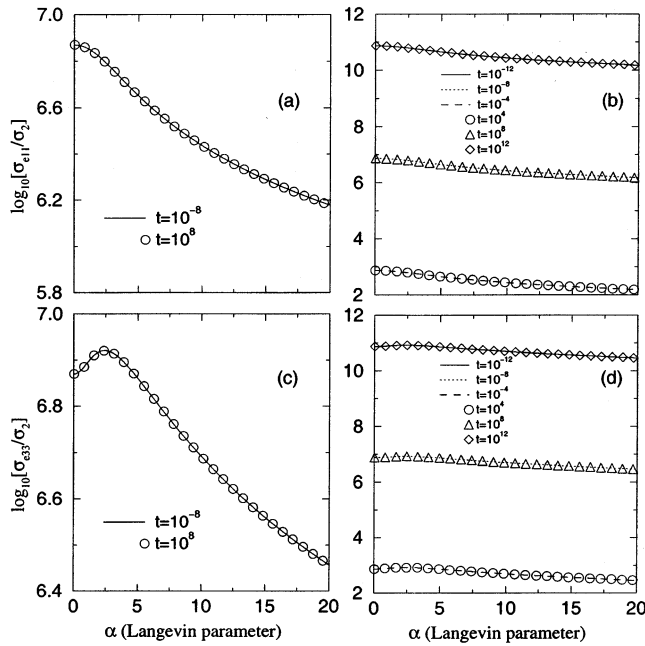
### III. Numerical Results

We are now in a position to do some numerical calculations. We shall discuss two cases: case I, a spherical particle with tensorial conductivity; case II, an ellipsoidal particle with scalar conductivity. In doing so, we can focus on the effect of shape or material (conductivity) anisotropies, respectively.

To start, we have to derive the expression for  $\langle \tilde{\tilde{\rho}} \rangle$ . For  $L_x = L_y \neq L_z$  and  $\sigma_{11} = \sigma_{22} \neq \sigma_{33}$ ,  $\langle \tilde{\tilde{\rho}} \rangle$  can be obtained as<sup>19</sup>

$$\langle \tilde{\tilde{\rho}} \rangle = \begin{pmatrix} \frac{1}{2}[\rho_{11} + \rho_{33} - (\rho_{33} - \rho_{11})\zeta] & 0 & 0 \\ 0 & \frac{1}{2}[\rho_{22} + \rho_{33} - (\rho_{33} - \rho_{22})\zeta] & 0 \\ 0 & 0 & \rho_{11} + (\rho_{33} - \rho_{11})\zeta \end{pmatrix} \quad (14)$$

where  $\zeta = \langle \cos^2 \theta \rangle$ , with  $\theta$  being the polar orientation angel of a particle and  $0 \leq \theta \leq \pi/2$ . Since the orientation of the ferromagnetic particles in ferrogels can be affected by the external magnetic field, there is  $\zeta = 1 - 2L(\alpha)/\alpha$ .<sup>16</sup> Here,  $L(\alpha)$  denotes the Langevin function and  $\alpha = mH/k_B T$  the Langevin parameter, where  $m$  represents the magnetic moment of a particle,  $H$ , the magnetic field,  $k_B$ , the Boltzmann constant, and  $T$ , the temperature. For the sake of convenience,  $\alpha$  will be used to denote the magnetic field in the following numerical calculations. In a word,  $\zeta$  contains the information of the spatial orientation of the particles in the host. In particular, the zero field ( $\alpha = 0$ ) yields  $\zeta \rightarrow 1/3$  naturally due to the random orientation of the particles. As  $\alpha > 0$ ,  $\zeta$  deviates from  $1/3$  accordingly. In other words, the specific spatial orientation of the particles in the ferrogel can be achieved by adjusting the external magnetic field.



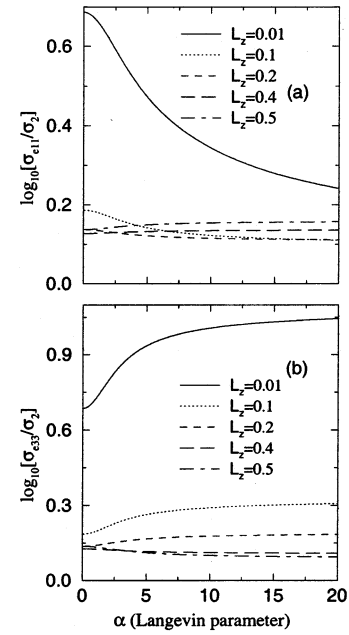
**Figure 1.** Case I: Effective electrical conductivity, (a and b)  $\sigma_{e11}/\sigma_2$  and (c and d)  $\sigma_{e33}/\sigma_2$ , vs the Langevin parameter,  $\alpha$ , for different conductivity ratios,  $t$ 's ( $=\sigma_{33}/\sigma_{11}$ ). Parameters:  $p = 0.1$  and  $\sigma_{11}/\sigma_2 = 10^{22}$ .

**A. Case I: Spherical Particles with Anisotropic Conductivity.** In this case, there are  $L_x = L_y = L_z = 1/3$  and  $\sigma_{11} = \sigma_{22} \neq \sigma_{33}$ . In Figure 1, we display the effective electrical conductivity versus the Langevin parameter for different conductivity ratios  $t$ 's ( $=\sigma_{33}/\sigma_{11}$ ). In fact,  $t$  represents the degree of anisotropy of conductivity. We find increasing the Langevin parameter,  $\alpha$ , causes  $\sigma_{e11}$  to decrease; see Figure 1a and b. In contrast, in Figure 1c and d, as  $\alpha$  increases,  $\sigma_{e33}$  increases first and decreases after a peak. In particular, at  $\alpha = 2.4$ , there is always a peak for  $\log_{10}[\sigma_{e33}/\sigma_2]$ . In detail, for  $t = 10^{12}$ ,  $10^8$ , and  $10^4$ , the corresponding peak values are  $10.92$ ,  $6.92$ , and  $2.92$ , respectively. In other words, an optimal magnetic field exists which leads to a maximum conductivity,  $\sigma_{e33}$ . It is worth noting that both  $t = 10^{-N}$  and  $t = 10^N$  ( $N$  is a number) can predict the same  $\sigma_{e11}$  or  $\sigma_{e33}$  concerned; see Figure 1b and d. This behavior stays unchanged up to  $N = 19$  or so. Moreover, a larger  $N$  value and hence a stronger conductivity anisotropy can lead to a higher effective electrical conductivity,  $\sigma_{e11}$  or  $\sigma_{e33}$ .

**B. Case II: Ellipsoidal Particles with Isotropic Conductivity.** For this case,  $L_x = L_y \neq L_z$  and  $\sigma_{11} = \sigma_{22} = \sigma_{33}$ . Figure 2 shows the effective electrical conductivity against the Langevin parameter,  $\alpha$ , for a different depolarization factor,  $L_z$ . For prolate spheroidal particles ( $L_z < 1/3$ ), a smaller  $L_z$  value can lead to a higher  $\sigma_{e11}$  or  $\sigma_{e33}$  value; see Figure 2. In contrast, for oblate spheroidal particles ( $L_z > 1/3$ ), a larger  $L_z$  value can yield a higher  $\sigma_{e11}$  or lower  $\sigma_{e33}$  value. Also, for the prolate spheroidal cases, increasing the magnetic field causes  $\sigma_{e11}$  (or  $\sigma_{e33}$ ) to decrease (or increase). However, for the oblate spheroidal cases, as the magnetic field increases,  $\sigma_{e11}$  increases but  $\sigma_{e33}$  decreases. In a word, the stronger shape anisotropy (namely, how  $L_z$  deviates from  $1/3$ ) can yield a higher effective electrical conductivity.

From cases I and II, it is concluded that material (conductivity) anisotropies are more important for yielding higher effective electrical conductivities of ferrogels than shape anisotropies, while magnetic fields can offer a correction.

In addition, we also discussed the effect of the volume fraction of particles (no figures shown here). It is shown that increasing



**Figure 2.** Case II: Same as Figure 1 but for a different depolarization factor,  $L_z$ .

the volume fraction of particles can cause the effective electrical conductivity to increase accordingly. This result is the same as that obtained by Xue.<sup>20</sup>

#### IV. Discussion and Conclusion

Here, some comments are in order. In this paper, we have presented a general model for a ferrogel in which both the particle shape and conductivity have a tensorial form, in an attempt to study the effect of the shape and/or material (conductivity) anisotropy on the effective electrical conductivity of the ferrogel, by taking into account the spatial orientation of the particles in the presence of an external magnetic field. Our theory reproduced several known results and provides a link between the particle properties and orientation distribution and the effective electrical conductivity of ferrogels.

For water, minerals such as sodium chloride are often dissolved in it, thus making it have a conductivity. More importantly, as one tries to make pure water by gradually removing electrolytes, its conductivity gradually decreases indeed. However, if all electrolytes are removed, its conductivity is still nonzero. The reason is that an infinitesimal part of the molecules of water (only about one in 500 million) is ionized as hydrogen ions ( $H^+$ ) and hydroxide ions ( $OH^-$ ). Theoretically, at this point, the conductivity becomes  $5.48 \times 10^{-6}$  S/m at 25 °C. In contrast, the conductivity of polymers could be  $\sim 10^{-15}$  S/m. For the ferrogel discussed in this paper, we see the ferromagnetic particles embedded in the host medium which is composed of polymers as well as water.

Since the structure and magnetization properties of a polydisperse ferrogel can differ from those of a monodisperse system,<sup>21–23</sup> it is instructive to extend the present work to the polydisperse case, so that one could investigate the polydisperse effect on the effective electrical conductivity of polydisperse ferrogels.

In this work, we derived an anisotropic Maxwell–Garnett formula (eq 11), which is nonsymmetrical and may thus be suitable for low concentrations. For a higher concentration of particles, we can derive an alternative anisotropic Bruggeman formula (see the Appendix), which is symmetrical. Similar results should be obtained by means of the anisotropic Brugge-

man formula. However, the Bruggeman formula can predict a percolation threshold.

It is also interesting to see what happens if one extends the present model to investigate the electro-optical effects,<sup>19</sup> non-linearity enhancement,<sup>24</sup> figure of merit,<sup>24</sup> and nonlinear ac responses<sup>25–27</sup> of ferrogels due to the presence of the (weak) nonlinearity inside the metallic particles. In doing so, the particle reorientation arising from external magnetic fields is expected to play a role as well. In addition, due to the analogy of the mathematical form between the conductivities and dielectric constants of composite materials, it is straightforward to apply the present theory to the effective dielectric constant of ferrogels.

To sum up, on the basis of our general model, it was shown that material (conductivity) anisotropies are more important for producing a high effective electrical conductivity of ferrogels than shape anisotropies, while magnetic fields can offer a correction due to the change in the orientation distribution of particles.

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### Appendix: Anisotropic Bruggeman Formula

To derive the anisotropic Bruggeman formula, let us start from the effective dipole factor of the two components in the system. For the particles, the dipole factor,  $\vec{b}_1$ , is

$$\vec{b}_1 = \begin{pmatrix} \frac{L_x(\sigma_{11} - \sigma_{e11})}{L_x\sigma_{11} + (1 - L_x)\sigma_{e11}} & 0 & 0 \\ 0 & \frac{L_y(\sigma_{22} - \sigma_{e22})}{L_y\sigma_{22} + (1 - L_y)\sigma_{e22}} & 0 \\ 0 & 0 & \frac{L_z(\sigma_{33} - \sigma_{e33})}{L_z\sigma_{33} + (1 - L_z)\sigma_{e33}} \end{pmatrix} \quad (15)$$

On the other hand, the host has a dipole factor,  $\vec{b}_2$ , such that

$$\vec{b}_2 = \frac{\sigma_2 \vec{I} - \vec{\sigma}_e}{\sigma_2 \vec{I} + 2\vec{\sigma}_e} \quad (16)$$

Now we can derive the Bruggeman formula by considering the fact that the effective dipole factor,  $\langle \vec{b} \rangle$ , of the whole system should be zero and then obtain

$$\langle \vec{b} \rangle \equiv p \langle \vec{b}_1 \rangle + (1 - p) \vec{b}_2 = 0 \quad (17)$$

where  $\langle \vec{b}_1 \rangle = \langle \vec{v} \vec{b}_1 \vec{v}^T \rangle$ . In the case of  $L_x = L_y = L_z$  and  $\sigma_{11} = \sigma_{22} = \sigma_{33}$  (scalar conductivity), eq 17 reduces to the well-known (isotropic) Bruggeman formula

$$p \frac{\sigma_1 - \sigma_e}{\sigma_1 + 2\sigma_e} + (1 - p) \frac{\sigma_2 - \sigma_e}{\sigma_2 + 2\sigma_e} = 0 \quad (18)$$

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