Derivation of nondiagonal effective dielectric-permeability tensors for magnetized granular composites

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The effective dielectric-permeability tensors, including off-diagonal terms, for magnetized composites are derived. Based on Bragg and Pippard's average field approximation, the effective tensor is derived for a composite containing an ensemble of oriented ellipsoidal particles embedded in a host medium, which is magnetized along an arbitrary direction. The effective tensor elements are given by the average of the tensor elements of particles and the host medium weighted by ''virtual volume fractions.'' The average electric field at the particle is shown to be a local Lorentz field generalized to ellipsoids. Based on Bruggeman's symmetrized effective-medium theory, the effective permeability tensor is derived self-consistently for the magnetized composite involving n types of ensembles of randomly oriented ellipsoidal particles. The diagonal effective tensor element $\hat{\epsilon}$ is obtained by solving the equation for $\hat{\epsilon}$ of order $2n$, independently of the off-diagonal effective tensor element $\hat{\Gamma}$, while $\hat{\Gamma}$ is given as the average of the off-diagonal permeabilities of the constituents weighted by ''symmetrized virtual volume fractions.'' Bruggeman's effective permeability tensor, including off-diagonal terms, is calculated for Fe-SiO_2 cermet, which falls between the theoretical upper and lower bounds derived by Hashin and Shtrikman.

I. INTRODUCTION

Light wave propagates in a granular composite as if it were a continuous medium, provided the extension of the inhomogeneity is much smaller than the wavelength of the light. The optical and magneto-optical properties of such a composite can be characterized by the effective dielectric permeability, which is the space average of the dielectric permeability over all components of the composite.

Effective dielectric permeability has been long known to exist. In the beginning of this century Maxwell-Garnett[1] derived an effective dielectric constant for metal glasses in which metal fine aggregates spherical in shape are dispersed. He generalized the Clausius-Mossotti equation[2] for spherical atoms to spherical metal particles by approximating the local field acting on the particles by the local Lorentz field. The Maxwell-Garnett effective permeability has been used frequently to describe the optical properties of a wide variety of aggregated systems.[3]

Cohen *et al.*,[3] by direct inspection, generalized the Maxwell-Garnett effective dielectric constant for aggregates of spherical particles to oriented ellipsoidal particles by substitution of the appropriate depolarization factor. The Maxwell-Garnett effective dielectric constant was generalized to randomly oriented ellipsoids by Polder and Van Santen[4] and Hayashi, Nakamori, and Kanamori;[5] they applied averaged electric polarizability to the random ensemble of the ellipsoids.

The effective permeability has also been derived in a way different from that used by Maxwell-Garnett. Approximating the local field acting on the particles by the average field in the medium surrounding the particles, Bragg and Pippard[6] derived an effective dielectric permeability for an ensemble of ellipsoidal particles. They, and later Landauer,[7] pointed out that the average field approximation is equivalent to the Lorentz field approximation. The theory derived by Maxwell-Garnett, or Bragg and Pippard, includes interactions between the particles only through the Lorentz field. This limits its applicability to situations in which the particles are sparsely dispersed, or the volume fractions occupied by the particles is small. When the volume fractions of constituent components in a two-component composite become of the same order of magnitude, the roles of host and inclusions become ambiguous; we will have two different values of the Maxwell-Garnett effective permeability by interchanging the roles of host and inclusions, even if the respective volume fractions are kept constant.

A better description of the effective dielectric permeability can be achieved within a self-consistent theory, which was originally put forward by Bruggeman[8] and has since been rediscovered by Landauer.[7,9] In the self-consistent theory, inclusion and host are treated symmetrically; both are considered particles, and a particle of either inclusion or host is embedded in an effective medium (involving the two components) whose effective dielectric permeability is to be determined self-consistently. The self-consistency requirement is derived by claiming that the deviation of the electric field vanishes when averaged over the total volume of the composite. This symmetrical approximation theory is called Bruggeman's effective-medium theory, which has a close analogy with the coherent potential approximation for alloys.

Based on Bruggeman's symmetrical approximation, Granqvist and Hunderi[10,11] derived the self-consistent effective dielectric permeability for composites involving randomly oriented ellipsoidal granules. They analyzed the optical transmittance in metal-rich Ag-SiO_2 cermet films containing randomly oriented ellipsoidal SiO_2 granules in an Ag matrix in terms of the effective permeability derived from Bruggeman's self-consistent theory. In their model the host was assumed to be composed of spherical grains, which

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were later generalized to randomly oriented ellipsoidal grains by Norris, Sheng, and Callegari[12] and Pecharroman and Iglesias.[13]

The effective-medium theory was combined with scattering theories to explain light absorption in composite films[14] and poorly crystallized films,[15] birefringence in phase-separated glasses,[16] and microwave loss in granular $YBa_2Cu_3O_{\eta-\delta}$ superconductors.[17] The effective-medium theory was also combined with percolation scaling theory to account for the dielectric response in porous media.[18] The effective-medium approximation was further applied successfully to explain various effects in composites, which include electrical resistivity and its percolation,[19] the Hall effect,[20] and the nonlinear optical effect.[21]

The effective dielectric permeability has also been calculated by the Fourier expansion method for a periodic array of spheres,[22] and by various scattering theories for periodic and random arrays of spheres.[23]

Most physical effects thus far studied on composites are expressed in terms of diagonal tensors; exceptions are the Hall effect and the magneto-optical effect which are characterized by the off-diagonal terms of the conductivity and dielectric permeability tensors, respectively. However, studies of them, especially of the latter, are small in number.

To our knowledge there have been only two theoretical studies on the nondiagonal effective dielectric-permeability tensors for granular composites. Lissberger and Saunders[24] extended the Maxwell-Garnett effective dielectric constant to tensor form, including off-diagonal terms, for a composite containing magnetized spherical particles embedded in a dielectric matrix. The present author[25] has derived the effective permeability tensor for an array of magnetized ellipsoids dispersed in a dielectric host medium, extending Bragg and Pippard's permeability. However, there was an error in the approximation in our previous work, as will be shown below.

Recently a monograph dealing with an extended range of optical properties of metal cluster composites was written by Kreibig and Vollmer.[26] They did not, however, deal with the off-diagonal effective dielectric tensor.

This paper is concerned with the off-diagonal dielectric tensor for magnetized composites. We will first generalize Bragg and Pippard's effective dielectric permeability to a nondiagonal tensor for a magnetized composite in which oriented ellipsoidal granules are dispersed in a host medium. Next we will generalize Bruggeman's self-consistent effective permeability to a nondiagonal tensor for a magnetized composite containing multiple ensembles of randomly oriented ellipsoidal granules.

In Sec. II we will solve the quasistatic potential boundary problem for the electric field induced in a magnetized ellipsoid, and the electric polarizability of the ellipsoid will be derived. The result is used in Sec. III to derive the effective dielectric permeability tensors for magnetized composites based on Bragg and Pippard's approximation and Bruggeman's self-consistent approximation. We will also show that the self-consistent permeability tensor calculated for a $Fe-SiO_2$ granular composite falls between the upper and lower bounds derived by Hashin and Shtrikman.[27]

II. POLARIZABILITY OF MAGNETIZED ELLIPSOIDS

A. Potential boundary problem

Here we solve the potential boundary problem for the electric field induced in a magnetized ellipsoid in order to obtain the electric polarizability for the ellipsoid, based on a quasistatic approximation, keeping the time but not the spatial dependence of the electromagnetic field.

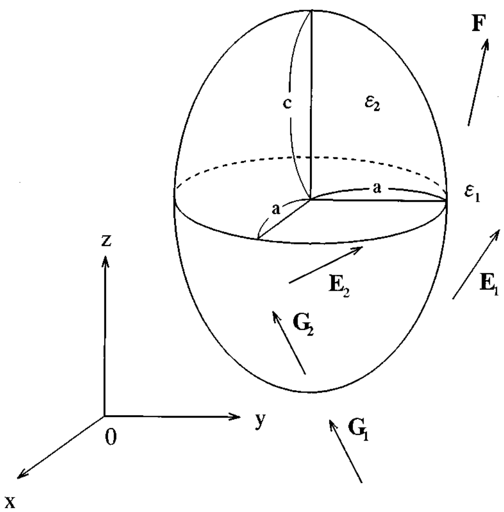

FIG. 1. Electric fields outside and inside a rotational ellipsoid of dielectric tensor $[\boldsymbol{\epsilon}_2]$ embedded in host material of $[\boldsymbol{\epsilon}_1]$, with the rotational axis parallel to the z direction. Both the host material and ellipsoid are magnetized along an arbitrary direction, having gyration vectors $\mathbf{G}_1$ and $\mathbf{G}_2$, respectively, which are parallel to the magnetization direction.

Consider, as shown in Fig. 1, that a uniform, isotropic medium with a wavelength-dependent dielectric constant ϵ_1 has in it a uniform, or quasistatic electric field

$$\mathbf{F}=\begin{pmatrix} F_x \\ F_y \\ F_z \end{pmatrix}. \tag{2.1}$$

Let a rotational ellipsoid, uniform and isotropic with a wavelength-dependent dielectric constant ϵ_2, be immersed in the medium. We assume that both the ellipsoid and the host medium are magnetized along the same, arbitrary direction. The dielectric permeability tensor is expressed to the first order of magnetization

$$[\boldsymbol{\epsilon}_1]=\begin{pmatrix} \epsilon_1 & \gamma_1 & -\delta_1 \\ -\gamma_1 & \epsilon_1 & \zeta_1 \\ \delta_1 & -\zeta_1 & \epsilon_1 \end{pmatrix} \tag{2.2}$$

for the medium and

$$[\boldsymbol{\epsilon}_2]=\begin{pmatrix} \epsilon_2 & \gamma_2 & -\delta_2 \\ -\gamma_2 & \epsilon_2 & \zeta_2 \\ \delta_2 & -\zeta_2 & \epsilon_2 \end{pmatrix}, \tag{2.3}$$

for the ellipsoid, differences among the diagonal terms being neglected. The gyration vectors $\mathbf{G}_1(\zeta_1,\delta_1,\gamma_1)$ and $\mathbf{G}_2(\zeta_2,\delta_2,\gamma_2)$ are parallel to each other and to the direction of the magnetization in the particle and the surrounding medium.

Inside the ellipsoid the field $\mathbf{F}$ induces an electric field

$$\mathbf{E}_2 = \begin{pmatrix} E_2^x \\ E_2^y \\ E_2^z \end{pmatrix}, \tag{2.4}$$

and an electric polarization **P**. Due to the depolarizing effect of **P**, $\mathbf{E}_2$ is depressed below **F**, while the field

$$\mathbf{E}_1 = \begin{pmatrix} E_1^x \\ E_1^y \\ E_1^z \end{pmatrix}, \tag{2.5}$$

induced outside the ellipsoid, exceeds **F** due to the dipole field from **P**. Since the dipole field reduces with the distance, the field $\mathbf{E}_1$ converges to the field **F** at infinity from the ellipsoid.

The electric fields $\mathbf{E}_1$ and $\mathbf{E}_2$ induce electric flux density fields

$$\mathbf{D}_1 = \begin{pmatrix} D_1^x \\ D_1^y \\ D_1^z \end{pmatrix} \tag{2.6}$$

and

$$\mathbf{D}_2 = \begin{pmatrix} D_2^x \\ D_2^y \\ D_2^z \end{pmatrix} \tag{2.7}$$

in the medium and ellipsoid, respectively, which satisfy the relations

$$\mathbf{D}_1 = [\boldsymbol{\epsilon}_1]\mathbf{E}_1, \tag{2.8}$$

$$\mathbf{D}_2 = [\boldsymbol{\epsilon}_2]\mathbf{E}_2. \tag{2.9}$$

The electric fields **F**, $\mathbf{E}_1$, and $\mathbf{E}_2$ are described in terms of potentials ϕ_0, ϕ_1, and ϕ_2 as

$$\mathbf{F} = -\nabla\phi_0, \tag{2.10}$$

$$\mathbf{E}_1 = -\nabla\phi_1, \tag{2.11}$$

$$\mathbf{E}_2 = -\nabla\phi_2. \tag{2.12}$$

Let the principal radii of the ellipsoid perpendicular and parallel to the rotational axis be a and c, respectively, and introduce spheroidal coordinates ξ, η, and ϕ, relating to the Cartesian coordinates as follows:[28]

$$x = \{(\xi + a^2)(\eta + a^2)/(a^2 - c^2)\}^{1/2}\cos\phi, \tag{2.13a}$$

$$y = \{(\xi + a^2)(\eta + a^2)/(a^2 - c^2)\}^{1/2}\sin\phi, \tag{2.13b}$$

$$z = \{(\xi + c^2)(\eta + c^2)/(c^2 - a^2)\}^{1/2}. \tag{2.13c}$$

The coordinates lie in the range

$$0 \leqslant \phi \leqslant 2\pi, \tag{2.14a}$$

$$-c^2(\text{or } a^2) < \xi, \tag{2.14b}$$

$$-a^2(\text{or } c^2) < \eta < -c^2(\text{or } a^2), \tag{2.14c}$$

where quantities outside and inside the parentheses correspond to oblate $(a > c)$ and prolate $(a < c)$ spheroidal coordinates, respectively. When $c = a$, the coordinates reduce to spherical coordinates.

In the spheroidal coordinate system the electric fields are expressed as

$$\mathbf{F} = \begin{pmatrix} F^\xi \\ F^\eta \\ F^\phi \end{pmatrix} = \begin{pmatrix} -h_1^{-1}\partial\phi_0/\partial\xi \\ -h_2^{-1}\partial\phi_0/\partial\eta \\ -h_3^{-1}\partial\phi_0/\partial\phi \end{pmatrix}, \tag{2.15}$$

$$\mathbf{E}_1 = \begin{pmatrix} E_1^\xi \\ E_1^\eta \\ E_1^\phi \end{pmatrix} = \begin{pmatrix} -h_1^{-1}\partial\phi_1/\partial\xi \\ -h_2^{-1}\partial\phi_1/\partial\eta \\ -h_3^{-1}\partial\phi_1/\partial\phi \end{pmatrix}, \tag{2.16}$$

$$\mathbf{E}_2 = \begin{pmatrix} E_2^\xi \\ E_2^\eta \\ E_2^\phi \end{pmatrix} = \begin{pmatrix} -h_1^{-1}\partial\phi_2/\partial\xi \\ -h_2^{-1}\partial\phi_2/\partial\eta \\ -h_3^{-1}\partial\phi_2/\partial\phi \end{pmatrix}, \tag{2.17}$$

where h_1, h_2, and h_3 are metrical coefficients given by Eq. (A3) in Appendix A.

On the surface of the spheroid, where $\xi = 0$, the induced fields must satisfy boundary conditions,[29] the continuity of the tangential component of the **E** vector

$$(E_1^\eta)_0 = (E_2^\eta)_0, \tag{2.18}$$

$$(E_1^\phi)_0 = (E_2^\phi)_0, \tag{2.19}$$

and the continuity of the normal component of the **D** vector

$$(D_1^\xi)_0 = (D_2^\xi)_0, \tag{2.20}$$

where 0 on the parentheses means $\xi = 0$. Using Eqs. (2.16) and (2.17), Eqs. (2.18) and (2.19) are rewritten in terms of the field potential as

$$\left(\frac{\partial\phi_1}{\partial\eta}\right)_0 = \left(\frac{\partial\phi_2}{\partial\eta}\right)_0, \tag{2.21}$$

$$\left(\frac{\partial\phi_1}{\partial\phi}\right)_0 = \left(\frac{\partial\phi_2}{\partial\phi}\right)_0, \tag{2.22}$$

and the left-hand side of Eq. (2.20), multiplied by $-(h_1)_0$, is expressed by the field potential as

$$\begin{aligned} -(h_1)_0(D_1^\xi)_0 = \epsilon_1\left(\frac{\partial\phi_1}{\partial\xi}\right)_0 + \left(\frac{h_1}{h_2h_3}\right)_0\Bigg[\gamma_1\Bigg\{&\left(\frac{\partial z}{\partial\phi}\right)_0\left(\frac{\partial\phi_1}{\partial\eta}\right)_0 \\ &- \left(\frac{\partial z}{\partial\eta}\right)_0\left(\frac{\partial\phi_1}{\partial\phi}\right)_0\Bigg\} + \delta_1\Bigg\{\left(\frac{\partial y}{\partial\phi}\right)_0\left(\frac{\partial\phi_1}{\partial\eta}\right)_0 \\ &- \left(\frac{\partial y}{\partial\eta}\right)_0\left(\frac{\partial\phi_1}{\partial\phi}\right)_0\Bigg\} + \zeta_1\Bigg\{\left(\frac{\partial x}{\partial\phi}\right)_0\left(\frac{\partial\phi_1}{\partial\eta}\right)_0 \\ &- \left(\frac{\partial x}{\partial\eta}\right)_0\left(\frac{\partial\phi_1}{\partial\phi}\right)_0\Bigg\}\Bigg], \end{aligned} \tag{2.23}$$

as shown in Appendix A.

Now we assume that the field potentials ϕ_1 and ϕ_2 are expressed in a similar way as expressed when both the particle and matrix are nonmagnetic (i.e., $\zeta_1=\delta_1=\gamma_1=\zeta_2=\delta_2=\gamma_2=0$),[28,29]

$$\phi_1=-\{F^x+c^xA(\xi)\}x-\{F^y+c^yA(\xi)\}y-\{F^z+c^2A'(\xi)\}z, \tag{2.24}$$

$$\phi_2=-\{E_2^xx+E_2^yy+E_2^zz\}, \tag{2.25}$$

where c^x, c^y, and c^z are constants, and

$$A(\xi)=\int_\xi^\infty \frac{ds}{(s+a^2)^2(s+c^2)^{1/2}}, \tag{2.26a}$$

$$A'(\xi)=\int_\xi^\infty \frac{ds}{(s+c^2)^2(s+a^2)^{1/2}}. \tag{2.26b}$$

That is, we assume that inside the ellipsoid a uniform electric field is induced, while outside the ellipsoid the electric dipole field due to the polarization of the ellipsoid superimposes on the applied external field. Our task is to express E_2^x, E_2^y, and E_2^z in terms of F^x, F^y, and F^z by eliminating c^x, c^y, and c^z using the boundary conditions of Eqs. (2.20)–(2.22). In the case when both the ellipsoid and host medium are not magnetized, one component, e.g., E_2^x, is solved independent of components E_2^y and E_2^z. In our case, where the off-diagonal dielectric permeabilities correlate the x component with the y and z components, we must solve simultaneous equations for E_2^x, E_2^y, and E_2^z.

A calculation shown in Appendix B yields the results

$$\{\epsilon_1+N(\epsilon_2-\epsilon_1)\}E_2^x+N(\gamma_2-\gamma_1)E_2^y-N'(\delta_2-\delta_1)E_2^z =\epsilon_1F^x, \tag{2.27a}$$

$$-N(\gamma_2-\gamma_1)E_2^x+\{\epsilon_1+N(\epsilon_2-\epsilon_1)\}E_2^y+N'(\zeta_2-\zeta_1)E_2^z =\epsilon_1F^y, \tag{2.27b}$$

$$N(\delta_2-\delta_1)E_2^x-N(\zeta_2-\zeta_1)E_2^y+\{\epsilon_1+N'(\epsilon_2-\epsilon_1)\}E_2^z=\epsilon_1F^z. \tag{2.27c}$$

Here we put

$$N=a^2cA(0)/2, \tag{2.28a}$$

$$N'=ac^2A'(0)/2, \tag{2.28b}$$

which are depolarization factors of the ellipsoid along x (or y) and z directions, respectively, satisfying the relation

$$2N+N'=1. \tag{2.29}$$

Equation (2.27) is expressed in a vector form

$$\mathbf{E}_2=\{[\mathbf{1}]+[\mathbf{N}]([\boldsymbol{\epsilon}_2]-[\boldsymbol{\epsilon}_1])\epsilon_1^{-1}\}^{-1}\mathbf{F}, \tag{2.30}$$

where $[\mathbf{1}]$ is an identical tensor and

$$[\mathbf{N}]=\begin{pmatrix} N & 0 & 0\\ 0 & N & 0\\ 0 & 0 & N'\end{pmatrix} \tag{2.31}$$

is the depolarization factor tensor.

B. Polarizability of magnetized ellipsoid

The polarization $\mathbf{P}$ induced by $\mathbf{F}$, in the ellipsoid having a permeability $[\boldsymbol{\epsilon}_2]$ with respect to the host medium having a permeability $[\boldsymbol{\epsilon}_1]$, is given by the equation

$$[\boldsymbol{\epsilon}_2]\mathbf{E}_2=[\boldsymbol{\epsilon}_1]\mathbf{E}_2+\mathbf{P}, \tag{2.32}$$

which is obtained by replacing $[\boldsymbol{\epsilon}_1]$ for ϵ_0 (dielectric permeability of vacuum) in ordinary definition of $\mathbf{P}$. The polarizability tensor $[\boldsymbol{\alpha}]$ of the ellipsoid with respect to the surrounding medium defined by

$$\mathbf{P}=[\boldsymbol{\alpha}]\mathbf{F} \tag{2.33}$$

is calculated from Eqs. (2.30), (2.32), and (2.33) as

$$[\boldsymbol{\alpha}]=([\boldsymbol{\epsilon}_2]-[\boldsymbol{\epsilon}_1])\{\mathbf{1}+[\mathbf{N}]([\boldsymbol{\epsilon}_2]-[\boldsymbol{\epsilon}_1])\epsilon_1^{-1}\}^{-1}. \tag{2.34}$$

For a nonmagnetized ellipsoid embedded in a nonmagnetized host medium, the polarizability tensor has been obtained as[28,29]

$$[\boldsymbol{\alpha}]=(\epsilon_2-\epsilon_1)\{[\mathbf{1}]+[\mathbf{N}](\epsilon_2-\epsilon_1)\epsilon_1^{-1}\}^{-1}. \tag{2.35}$$

Comparing Eqs. (2.34) and (2.35), we notice that generalizing the nonmagnetized ellipsoid and matrix to magnetized ones changes $(\epsilon_2-\epsilon_1)$ to tensor form, $[\boldsymbol{\epsilon}_2]-[\boldsymbol{\epsilon}_1]$, but keeps ϵ_1^{-1} of the scalar form.

Since $[\boldsymbol{\epsilon}_1]$ and $[\boldsymbol{\epsilon}_2]$ are approximated to the first order of magnetization as expressed by Eqs. (2.2) and (2.3), the following relation holds:

$$|\epsilon_i|\gg|\zeta_i|,\quad |\delta_i|,\quad \text{and}\quad |\gamma_i|\qquad (i=1,2). \tag{2.36}$$

Consequently, we can neglect the second and higher terms of ζ_i, δ_i, and γ_i, and Eq. (2.34) is calculated as

$$[\boldsymbol{\alpha}]=\begin{pmatrix} (\epsilon_2-\epsilon_1)/\beta & (\gamma_2-\gamma_1)/\beta^2 & -(\delta_2-\delta_1)/(\beta\beta')\\ -(\gamma_2-\gamma_1)/\beta^2 & (\epsilon_2-\epsilon_1)/\beta & (\zeta_2-\zeta_1)/(\beta\beta')\\ (\delta_2-\delta_1)/(\beta\beta') & -(\zeta_2-\zeta_1)/(\beta\beta') & (\epsilon_2-\epsilon_1)/\beta'\end{pmatrix}, \tag{2.37}$$

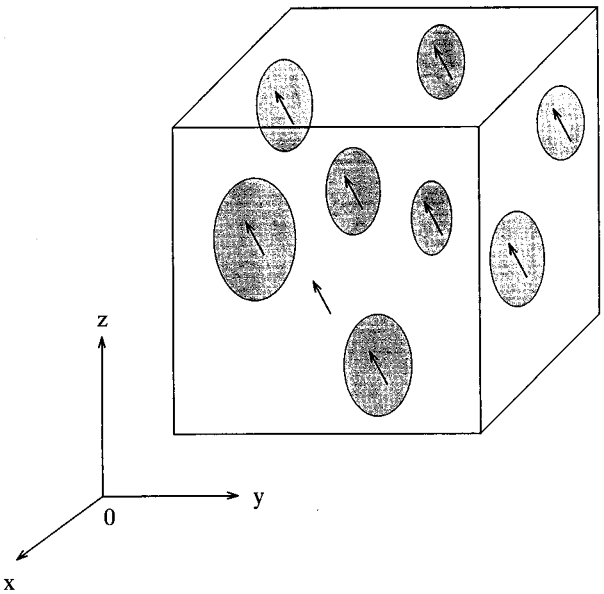

FIG. 2. A magnetized composite containing an oriented, spatially random, array of rotational ellipsoidal particles embedded in a host material. Arrows show magnetization (or gyration) vectors, which are directed along an arbitrary direction.

where we put

$$\beta = 1 + N(\epsilon_2 - \epsilon_1)/\epsilon_1, \tag{2.38a}$$

$$\beta' = 1 + N'(\epsilon_2 - \epsilon_1)/\epsilon_1. \tag{2.38b}$$

III. EFFECTIVE DIELECTRIC PERMEABILITY TENSOR

A. Average field approximation

In this section we first derive the effective permeability tensor based on the average field approximation theory proposed by Bragg and Pippard.[6] Let us consider, as shown in Fig. 2, a composite containing rotational-ellipsoidal particles, which are the same in shape but not necessarily in size, dispersed in a host medium. The particles are oriented with their rotational axes parallel to the z axis. They occupy a fraction f of the total volume of the composite. The host medium and the particles are magnetized along an arbitrary direction, having permeabilities $[\boldsymbol{\epsilon}_1]$ and $[\boldsymbol{\epsilon}_2]$ as expressed by Eqs. (2.2) and (2.3).

Now we assume that an external electric field, or light wave field $\mathbf{E}_0$, is applied to the composite. Due to the electric dipole interaction between the ellipsoids, the field $\mathbf{F}$ acting on the ellipsoids is not equal to the external field. Following Bragg and Pippard, we approximate the local field $\mathbf{F}$ acting on the ellipsoids by the average field in the host medium, and also approximate the average field in the ellipsoids by $\mathbf{E}_2$ which is induced by $\mathbf{F}$ in the ellipsoid following Eq. (2.30); here the applied external field $\mathbf{E}_0$ must be the same as the average field over the whole space inside and outside the ellipsoids:

$$\mathbf{E}_0 = f\mathbf{E}_2 + (1-f)\mathbf{F}. \tag{3.1}$$

The total electric flux density field $\hat{\mathbf{D}}$ of the composite is expressed in terms of the permeability $[\boldsymbol{\epsilon}_1]$ of the host and the polarizability $[\boldsymbol{\alpha}]$ of the inclusions with respect to the host as

$$\hat{\mathbf{D}} = [\boldsymbol{\epsilon}_1]\mathbf{E}_0 + f[\boldsymbol{\alpha}]\mathbf{F}. \tag{3.2}$$

The effective dielectric-permeability tensor $[\hat{\boldsymbol{\epsilon}}]$ relates $\hat{\mathbf{D}}$ to $\mathbf{E}_0$ as

$$\hat{\mathbf{D}} = [\hat{\boldsymbol{\epsilon}}]\mathbf{E}_0. \tag{3.3}$$

Substituting Eq. (2.30) into Eq. (3.1) we obtain

$$\mathbf{F} = [[\mathbf{1}] + [\mathbf{N}]\{([\boldsymbol{\epsilon}_2] - [\boldsymbol{\epsilon}_1])\epsilon_1^{-1}\}][[\mathbf{1}] + (1-f)[\mathbf{N}]\{([\boldsymbol{\epsilon}_2] - [\boldsymbol{\epsilon}_1])\epsilon_1^{-1}\}^{-1}]\mathbf{E}_0, \tag{3.4}$$

which is further substituted into Eq. (3.2) to yield

$$[\hat{\boldsymbol{\epsilon}}] = [\boldsymbol{\epsilon}_1] + f([\boldsymbol{\epsilon}_2] - [\boldsymbol{\epsilon}_1])\{[\mathbf{1}] + (1-f)[\mathbf{N}]([\boldsymbol{\epsilon}_2] - [\boldsymbol{\epsilon}_1])\epsilon_1^{-1}\}^{-1} \tag{3.5}$$

on referring to Eq. (3.3). Neglecting the second and higher terms of the off-diagonal permeabilities, the second term in the right-hand side of Eq. (3.5) is calculated as

$$f\begin{pmatrix} (\epsilon_2-\epsilon_1)/A & (\gamma_2-\gamma_1)/A^2 & -(\delta_2-\delta_1)/(AA') \\ -(\gamma_2-\gamma_1)/A^2 & (\epsilon_2-\epsilon_1)/A & (\zeta_2-\zeta_1)/(AA') \\ (\delta_2-\delta_1)/(AA') & -(\zeta_2-\zeta_1)/(AA') & (\epsilon_2-\epsilon_1)/A' \end{pmatrix} \tag{3.6a}$$

where we put

$$A = 1 + (1-f)N(\epsilon_2-\epsilon_1)/\epsilon_1, \tag{3.6b}$$

$$A' = 1 + (1-f)N'(\epsilon_2-\epsilon_1)/\epsilon_1. \tag{3.6c}$$

Thus the effective tensor elements $\hat{\epsilon}_{ij}(i,j=x,y,z)$ are expressed

$$\hat{\epsilon}_{xx} = \hat{\epsilon}_{yy} = (1-g)\epsilon_1 + g\epsilon_2, \tag{3.7a}$$

$$\hat{\epsilon}_{zz} = (1-g')\epsilon_1 + g'\epsilon_2, \tag{3.7b}$$

$$\hat{\epsilon}_{xy} = -\hat{\epsilon}_{yx} = (1-h)\gamma_1 + h\gamma_2, \tag{3.7c}$$

$$\hat{\epsilon}_{yz} = -\hat{\epsilon}_{zy} = (1-h')\zeta_1 + h'\zeta_2, \tag{3.7d}$$

$$\hat{\epsilon}_{zx} = -\hat{\epsilon}_{xz} = (1-h')\delta_1 + h'\delta_2, \tag{3.7e}$$

where we replaced

$$g = f/A, \tag{3.8a}$$

$$g' = f/A', \tag{3.8b}$$

$$h = f/A^2, \tag{3.8c}$$

$$h' = f/(AA'). \tag{3.8d}$$

We call g, g', h, and h', virtual volume fractions, with which the effective dielectric tensor elements are proportionally allotted between those of the particle and the matrix. In other words, the effective dielectric tensor elements are the average of the dielectric tensor elements of a fictitious composite having virtual volume fractions, though their definition differs for different elements of the tensor. The virtual volume fractions are given by the true fractions divided by A or A', or their products, which express the effect of the Lorentz field correction for the dipole interactions between particles as well as the depolarizing effect in the ellipsoid.

We want to call attention to an error in our previous work,[25] where the ellipsoids were magnetized along the rotational axis, or the z axis ($\zeta_1 = \zeta_2 = \delta_1 = \delta_2 = 0$). We incorrectly neglected terms of γ_2 in calculating $\hat{\epsilon}_{xy}$ which is of the same order as γ_2. This error leads to $h = f/A$ rather than the definition in Eq. (3.8c) above.

B. Effective-medium approximation

Next we derive the effective dielectric tensor based on Bruggeman's effective-medium theory. As mentioned in Sec. I, the self-consistency requirement in Bruggeman's theory is derived by requiring that the deviation of electric field is space averaged to zero. Since electric polarization causes the deviation of electric field, the self-consistency requirement is met when the electric dipole moment averaged over all elements of the composite vanishes. Therefore, for a composite involving multiple types of components, $1,2,\ldots,n$ which occupy fractions, $\Delta_1, \Delta_2, \ldots, \Delta_n$, respectively, of total volume of the composite ($\Sigma_{i=1}^{n}\Delta_i = 1$), the self-consistency requirement is expressed as follows:[7]

$$\sum_{i=1}^{n} \{\Delta_i [\hat{\boldsymbol{\alpha}}_i]\} = 0. \tag{3.9}$$

Here $[\hat{\boldsymbol{\alpha}}_i]$ is the symmetrized polarizability tensor for the component of type i $(=1,2,\ldots,n)$, which is expressed in terms not only of $[\boldsymbol{\epsilon}_i]$, the permeability of the ith component, but also of $[\hat{\boldsymbol{\epsilon}}]$, the effective permeability tensor to be determined self-consistently. We assume that ellipsoids belonging to the ensemble of type i are characterized by the depolarization factor N_i (and $N_i' = 1 - 2N_i$), which are the same in shape but not necessarily in size, as shown in Fig. 3.

In order to calculate the symmetrized polarizability $[\hat{\boldsymbol{\alpha}}_i]$ for the ellipsoid ensemble, we assign, as shown in Fig. 3, the x,y,z coordinates to one of the ellipsoids with the z axis parallel to the rotational axis, as before. We also assign the x',y',z' coordinates with the z' axis parallel to the magnetization or gyration vector. In the x',y',z' system, the permeability tensor of the ellipsoid belonging to the ith ensemble is expressed as

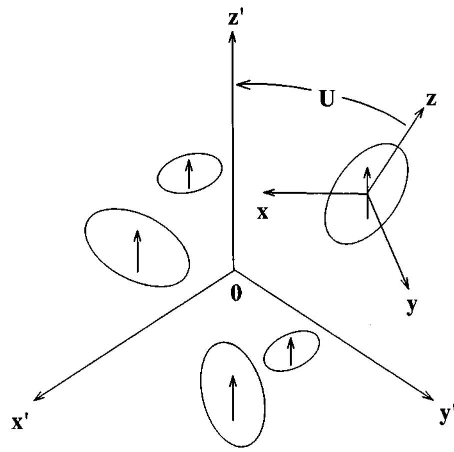

FIG. 3. An ensemble of randomly oriented rotational ellipsoids of similar shapes embedded in a host medium, magnetized along the z' direction. The x, y, and z coordinate system, which is assigned to one of the ellipsoids with its rotational axis parallel to the z axis, is transformed by $\mathbf{U}$ to the x',y',z' system with the z' axis parallel to the magnetization (or gyration) vector.

$$[\boldsymbol{\epsilon}_i'] = \begin{pmatrix} \epsilon_i & \Gamma_i & 0 \\ -\Gamma_i & \epsilon_i & 0 \\ 0 & 0 & \epsilon_i \end{pmatrix}, \tag{3.10}$$

and the symmetrized effective permeability tensor as

$$[\hat{\boldsymbol{\epsilon}}'] = \begin{pmatrix} \hat{\epsilon} & \hat{\Gamma} & 0 \\ -\hat{\Gamma} & \hat{\epsilon} & 0 \\ 0 & 0 & \hat{\epsilon} \end{pmatrix}. \tag{3.11}$$

Now we introduce a unitary matrix

$$\mathbf{U} = \begin{pmatrix} U_{11} & U_{12} & U_{13} \\ U_{21} & U_{22} & U_{23} \\ U_{31} & U_{32} & U_{33} \end{pmatrix}, \tag{3.12}$$

which transforms the x,y,z coordinate system to the x',y',z' system. The average polarizability for the ith ensemble of the ellipsoids is obtained by averaging over all unitary transformations which transform the rotational axis of all the ellipsoids belonging to the ith ensemble to the z' axis, as described in Appendix C. The result is

$$[\hat{\boldsymbol{\alpha}}_i] = \begin{pmatrix} (\epsilon_i - \hat{\epsilon})/\bar{\beta}_i & (\Gamma_i - \hat{\Gamma})/\overline{\beta_i^2} & 0 \\ -(\Gamma_i - \hat{\Gamma})/\overline{\beta_i^2} & (\epsilon_i - \hat{\epsilon})/\bar{\beta}_i & 0 \\ 0 & 0 & (\epsilon_i - \hat{\epsilon})/\bar{\beta}_i \end{pmatrix}, \tag{3.13a}$$

where

$$1/\bar{\beta}_i = \tfrac{2}{3}\{1 + N_i(\epsilon_i - \hat{\epsilon})/\hat{\epsilon}\}^{-1} + \tfrac{1}{3}\{1 + N_i'(\epsilon_i - \hat{\epsilon})/\hat{\epsilon}\}^{-1}, \tag{3.13b}$$

$$1/\overline{\beta_i^2} = \tfrac{1}{3}\{1 + N_i(\epsilon_i - \hat{\epsilon})/\hat{\epsilon}\}^{-2} + \tfrac{2}{3}\{1 + N_i(\epsilon_i - \hat{\epsilon})/\hat{\epsilon}\}^{-1} \times \{1 + N_i'(\epsilon_i - \hat{\epsilon})/\hat{\epsilon}\}^{-1}. \tag{3.13c}$$

Substituting Eq. (3.13) into the tensorial equation, Eq. (3.9), from the diagonal terms we obtain

$$\sum_{i=1}^{n}\{\Delta_i(\epsilon_i-\hat{\epsilon})/\overline{\beta}_i\}=0, \tag{3.14}$$

and, from the off-diagonal terms,

$$\sum_{i=1}^{n}\{\Delta_i(\Gamma_i-\hat{\Gamma})/\overline{\beta}_i^2\}=0. \tag{3.15}$$

From Eq. (3.15) we obtain

$$\hat{\Gamma}=\frac{\sum_{i=1}^{n}\{\Gamma_i\Delta_i/\overline{\beta}_i^2\}}{\sum_{i=1}^{n}\{\Delta_i/\overline{\beta}_i^2\}}=\sum_{i=1}^{n}\{\Gamma_i\overline{\Delta_i}\}, \tag{3.16}$$

where we put

$$\overline{\Delta_i}=\frac{\Delta_i/\overline{\beta}_i^2}{\sum_{j=1}^{n}\{\Delta_j/\overline{\beta}_j^2\}}, \tag{3.17}$$

which we call symmetrized virtual volume fractions for the ith ensemble of the ellipsoids. Equation (3.14) is an equation for $\hat{\epsilon}$ of order $2n$, which does not contain $\hat{\Gamma}$ or Γ_i. Thus we obtain $\hat{\epsilon}$ independently of $\hat{\Gamma}$ by solving the equation of order $2n$. From Eq. (3.16) we obtain $\hat{\Gamma}$ by averaging Γ_i, weighted by $\overline{\Delta_i}$.

Setting $n=2$, Eq. (3.14) for the diagonal term agrees with Eqs. (2) and (8) given in Refs. 12 and 13, respectively, for the composite composed of two randomly oriented kinds of ellipsoidal grains.

When the composite has a host composed of spherical grains, the average polarizability for the host is expressed

$$[\hat{\boldsymbol{\alpha}}_1]=\begin{pmatrix}(\epsilon_1-\hat{\epsilon})/\beta_1 & (\Gamma_1-\hat{\Gamma})/\beta_1^2 & 0\\ -(\Gamma_1-\hat{\Gamma})/\beta_1^2 & (\epsilon_1-\hat{\epsilon})/\beta_1 & 0\\ 0 & 0 & (\epsilon_1-\hat{\epsilon})/\beta_1\end{pmatrix}, \tag{3.18a}$$

$$\beta_1=(\epsilon_1+2\hat{\epsilon})/(3\hat{\epsilon}), \tag{3.18b}$$

by putting $i=1$ and $N_1=N'=\frac{1}{3}$ in Eq. (3.13). Substituting Eq. (3.18b) into Eq. (3.15), after some manipulation we obtain

$$\hat{\epsilon}=\epsilon_1\frac{\Delta_1+\frac{1}{3}\sum_{i=2}^{n}\Delta_i\overline{\beta}_i}{\Delta_1-\frac{2}{3}\sum_{i=2}^{n}\Delta_i\overline{\beta}_i}. \tag{3.19}$$

This agrees with the formula derived by Granqvist and Hunderi [Eq. (10) in Ref. 10] for $n-1$ ensembles of the randomly oriented ellipsoids embedded in a spherical granular host. When $n=2$, or the composite has only one ensemble of the randomly oriented ellipsoids in the spherical grain host, Eq. (3.14) is expressed

$$\Delta_1\frac{\hat{\epsilon}-\epsilon_1}{2\hat{\epsilon}+\epsilon_1}+\Delta_2\frac{(\hat{\epsilon}-\epsilon_2)\{(1+3N_2)\hat{\epsilon}+(2-3N_2)\epsilon_2\}}{9\{(1-N_2)\hat{\epsilon}+N_2\epsilon_2\}\{(1-N_2')\hat{\epsilon}+N_2'\epsilon_2\}}=0. \tag{3.20}$$

This is a cubic equation of $\hat{\epsilon}$, which agrees with Eq. (9) in Ref. 13. When the inclusions are also spheres ($N_2=N_2'=\frac{1}{3}$), Eq. (3.20) reduces to the following quadratic equation for $\hat{\epsilon}$,

$$\Delta_1(\hat{\epsilon}-\epsilon_1)(2\hat{\epsilon}+\epsilon_2)+\Delta_2(\hat{\epsilon}-\epsilon_2)(2\hat{\epsilon}+\epsilon_1)=0, \tag{3.21}$$

and the off-diagonal term, Eq. (3.16) is expressed as

$$\hat{\Gamma}=\frac{\Delta_1\Gamma_1(2\hat{\epsilon}+\epsilon_2)^2+\Delta_2\Gamma_2(2\hat{\epsilon}+\epsilon_1)^2}{\Delta_1(2\hat{\epsilon}+\epsilon_2)^2+\Delta_2(2\hat{\epsilon}+\epsilon_1)^2}. \tag{3.22}$$

Equation (3.21) agrees with that given by Landauer.[7,9]

C. Effective permeability and bounds calculated for cermet

As an example, Bruggeman's effective permeability is calculated at a wavelength of 0.8 μm for Fe-SiO_2 cermet, assuming both Fe and SiO_2 are isotropically characterized by the spherical depolarization factor ($N=\frac{1}{3}$). As Fig. 4 shows, the diagonal term $\hat{\epsilon}$, or the off-diagonal term $\hat{\Gamma}$, falls between the two values, or theoretical bounds,[27] calculated from the Maxwell-Garnett theory assuming that Fe and SiO_2 are playing the role of inclusions and host medium, respectively, and vice versa.

IV. DISCUSSION

Solving the potential boundary problem, we derived the electric field $\mathbf{E}_2$ induced in a magnetized ellipsoid exposed to electric field $\mathbf{F}$. The result is given by Eq. (2.30), which is rewritten, by combining Eqs. (2.33) and (2.34) with Eq. (2.30), as

$$\mathbf{E}_2=\mathbf{F}-[\mathbf{N}]\mathbf{P}/\epsilon_1, \tag{4.1}$$

where $\mathbf{P}$ is the polarization induced in the ellipsoid. Therefore, a uniform depolarizing field expressed as $[\mathbf{N}]\mathbf{P}/\epsilon_1$ is working in the magnetized ellipsoid as well as in a nonmagnetized ellipsoid.[28,29] It should be noted that this is derived on approximating the permeability tensor to the first order of magnetization, or setting $\epsilon_{xx}=\epsilon_{yy}=\epsilon_{zz}$ in Eqs. (2.2) and (2.3).

Based on the average field approximation proposed by Bragg and Pippard,[6] we derived a nondiagonal effective permeability tensor for a two-component composite containing oriented ellipsoidal particles in a host medium, which is magnetized along an arbitrary direction. The average field $\mathbf{F}$ acting on the particles is rewritten as

$$\mathbf{F}=\mathbf{E}_0+[\mathbf{N}]\mathbf{p}/\epsilon_1 \tag{4.2}$$

by combining Eqs. (2.33) and (2.34) with Eq. (3.4) and setting

$$\mathbf{p}=f\mathbf{P}. \tag{4.3}$$

Since $\mathbf{p}$ represents the polarization induced with respect to the host medium of the dielectric constant $[\boldsymbol{\epsilon}_1]$ per unit volume of the total composite, Eq. (4.2) reduces to the local

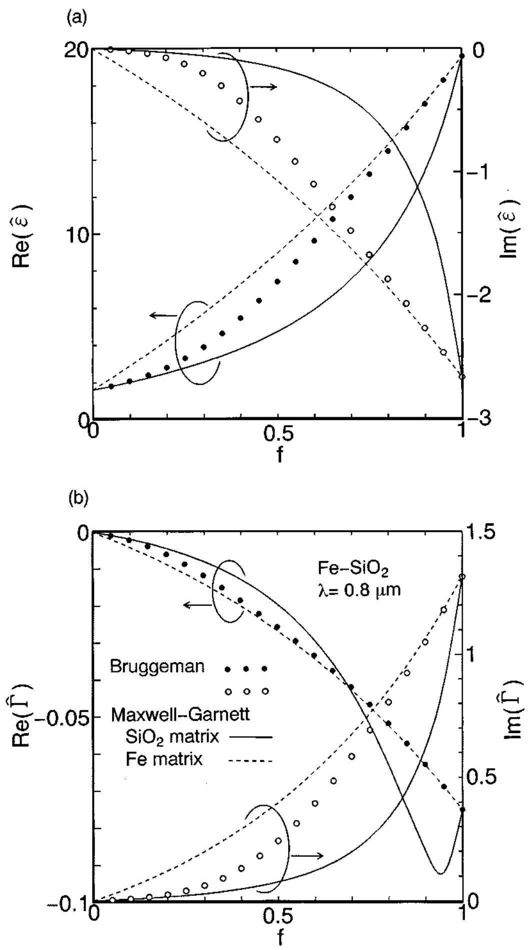

FIG. 4. Real and imaginary parts of diagonal ($\hat{\epsilon}$) and off-diagonal ($\hat{\Gamma}$) effective dielectric permeabilities calculated at $\lambda = 0.8$ μm for Fe-SiO$_2$ cermet as a function of Fe volume fraction, assuming the spherical depolarization factor ($N=\frac{1}{3}$) for both Fe and SiO$_2$. Circles: Bruggeman's permeability. Solid lines: Maxwell-Garnett's permeability for Fe particles in a SiO$_2$ matrix. Dashed lines: the same for SiO$_2$ particles in an Fe matrix. Diagonal and off-diagonal permeabilities of Fe are taken from Refs. 30 and 31, respectively.

Lorentz field if we put $[\mathbf{N}]=[\mathbf{1}]/3$. This reveals that Bragg and Pippard's approximation is a generalization of Maxwell-Garnett's or Clausius-Mossotti's approximation, as has been suggested previously,[6,7] though not by formula.

Our Maxwell-Garnett effective dielectric permeability extended to the magnetized composite is expressed by a simple formula, Eq. (3.7), using virtual volume fractions. However, we found that the effective tensor cannot be expressed by a simple formula when the composite involves many components (i.e., $n \geqslant 3$) and/or randomly oriented ellipsoidal particles. Conversely, Bruggeman's effective permeability tensor for a magnetized composite is derived from a simple formula [Eqs. (3.14) and (3.16)] even when it contains many ($n \geqslant 3$) ensembles of randomly oriented ellipsoidal particles.

V. CONCLUSION

The main results of this study are summarized as follows.

(1) Using the dielectric permeability tensor approximated to the first order of magnetization, we solved the quasistatic potential boundary problem for electric field induced inside a magnetized ellipsoid which is exposed to a uniform external electric field. A uniform depolarizing field $[\mathbf{N}]\mathbf{P}/\epsilon_1$ is superimposing the external field in the magnetized ellipsoid, similar as in a nonmagnetized ellipsoid.

(2) Bragg and Pippard's effective dielectric permeability for the composite containing oriented ellipsoidal particles embedded in a matrix was generalized to the magnetized composite. The off-diagonal, as well as diagonal, terms of the effective permeability are given by the average of those for the particles and the matrix, weighted by the virtual volume fractions. The field acting on the particles was revealed to be a local Lorentz field, $\mathbf{E}_0+[\mathbf{N}]\mathbf{p}/\epsilon_1$, generalized to the ellipsoidal particles.

(3) Bruggeman's self-consistent effective permeability was generalized to the magnetized composite which contains n different ensembles of randomly oriented ellipsoidal particles. The diagonal effective permeability $\hat{\epsilon}$ is obtained by solving the equation of order $2n$, independently of the off-diagonal permeability $\hat{\Gamma}$, while $\hat{\Gamma}$ is given by averaging the off-diagonal permeabilities Γ_i of the components weighted by the symmetrized virtual volume fractions $\overline{\Delta}_i$. When the composite contains only two ensembles of spherical particles, the equation of $\hat{\epsilon}$ reduces to a quadratic one.

(4) Bruggeman's effective permeability tensor calculated for Fe-SiO$_2$ cermet falls between theoretical upper and lower bounds derived by Hashin and Shtrikman. An experimental study is in progress to describe the magneto-optical properties of metal granular composite in terms of the effective permeability tensors.

ACKNOWLEDGMENT

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APPENDIX A: FIELD POTENTIAL EXPRESSION OF $\mathbf{D}^\xi$

Let us derive Eq. (2.23), the normal component of $\mathbf{D}$ vector expressed in terms of field potentials. The normal, or ξ, component of $\mathbf{D}$ vector is written as

$$D_1^\xi=\mathbf{n}_\xi\cdot\mathbf{D}_1=n_\xi^x D_1^x+n_\xi^y D_1^y+n_\xi^z D_1^z. \tag{A1}$$

Here $\mathbf{n}_\xi$ is one of the following unitary vectors along the ξ, η, and ϕ coordinates,

$$\mathbf{n}_\xi=\begin{pmatrix} n_\xi^x \\ n_\xi^y \\ n_\xi^z \end{pmatrix}=\frac{1}{h_1}\begin{pmatrix} \partial x/\partial\xi \\ \partial y/\partial\xi \\ \partial z/\partial\xi \end{pmatrix}=h_1\begin{pmatrix} \partial\xi/\partial x \\ \partial\xi/\partial y \\ \partial\xi/\partial z \end{pmatrix}, \tag{A2a}$$

$$\mathbf{n}_\eta=\begin{pmatrix} n_\eta^x \\ n_\eta^y \\ n_\eta^z \end{pmatrix}=\frac{1}{h_2}\begin{pmatrix} \partial x/\partial\eta \\ \partial y/\partial\eta \\ \partial z/\partial\eta \end{pmatrix}=h_2\begin{pmatrix} \partial\eta/\partial x \\ \partial\eta/\partial y \\ \partial\eta/\partial z \end{pmatrix}, \tag{A2b}$$

$$\mathbf{n}_\phi = \begin{pmatrix} n_\phi^x \\ n_\phi^y \\ n_\phi^z \end{pmatrix} = \frac{1}{h_3}\begin{pmatrix} \partial x/\partial\phi \\ \partial y/\partial\phi \\ \partial z/\partial\phi \end{pmatrix} = h_3 \begin{pmatrix} \partial\phi/\partial x \\ \partial\phi/\partial y \\ \partial\phi/\partial z \end{pmatrix}, \quad \text{(A2c)}$$

where h_1, h_2, and h_3 are metrical coefficients given by[29]

$$h_1 = \left\{ \left(\frac{\partial x}{\partial \xi}\right)^2 + \left(\frac{\partial y}{\partial \xi}\right)^2 + \left(\frac{\partial z}{\partial \xi}\right)^2 \right\}^{1/2}, \quad \text{(A3a)}$$

$$h_2 = \left\{ \left(\frac{\partial x}{\partial \eta}\right)^2 + \left(\frac{\partial y}{\partial \eta}\right)^2 + \left(\frac{\partial z}{\partial \eta}\right)^2 \right\}^{1/2}, \quad \text{(A3b)}$$

$$h_3 = \left\{ \left(\frac{\partial x}{\partial \phi}\right)^2 + \left(\frac{\partial y}{\partial \phi}\right)^2 + \left(\frac{\partial z}{\partial \phi}\right)^2 \right\}^{1/2}. \quad \text{(A3c)}$$

From Eqs. (2.2), (2.8), and (2.11), we obtain

$$-D_1^x = \epsilon_1 \frac{\partial \phi_1}{\partial x} + \gamma_1 \frac{\partial \phi_1}{\partial y} - \delta_1 \frac{\partial \phi_1}{\partial z}, \quad \text{(A4a)}$$

$$-D_1^y = -\gamma_1 \frac{\partial \phi_1}{\partial x} + \epsilon_1 \frac{\partial \phi_1}{\partial y} + \zeta_1 \frac{\partial \phi_1}{\partial z}, \quad \text{(A4b)}$$

$$-D_1^z = \delta_1 \frac{\partial \phi_1}{\partial x} - \zeta_1 \frac{\partial \phi_1}{\partial y} + \epsilon_1 \frac{\partial \phi_1}{\partial z}. \quad \text{(A4c)}$$

Using Eq. (A2), the partial derivatives in the right-hand side of Eq. (A4) are given as

$$\begin{aligned} \frac{\partial \phi_1}{\partial x} &= \frac{\partial \xi}{\partial x}\frac{\partial \phi_1}{\partial \xi} + \frac{\partial \eta}{\partial x}\frac{\partial \phi_1}{\partial \eta} + \frac{\partial \phi}{\partial x}\frac{\partial \phi_1}{\partial \phi} \\ &= \frac{n_\xi^x}{h_1}\frac{\partial \phi_1}{\partial \xi} + \frac{n_\eta^x}{h_2}\frac{\partial \phi_1}{\partial \eta} + \frac{n_\phi^x}{h_3}\frac{\partial \phi_1}{\partial \phi}, \end{aligned} \quad \text{(A5a)}$$

$$\frac{\partial \phi_1}{\partial y} = \frac{n_\xi^y}{h_1}\frac{\partial \phi_1}{\partial \xi} + \frac{n_\eta^y}{h_2}\frac{\partial \phi_1}{\partial \eta} + \frac{n_\phi^y}{h_3}\frac{\partial \phi_1}{\partial \phi}, \quad \text{(A5b)}$$

$$\frac{\partial \phi_1}{\partial z} = \frac{n_\xi^z}{h_1}\frac{\partial \phi_1}{\partial \xi} + \frac{n_\eta^z}{h_2}\frac{\partial \phi_1}{\partial \eta} + \frac{n_\phi^z}{h_3}\frac{\partial \phi_1}{\partial \phi}. \quad \text{(A5c)}$$

Equation (A5) is substituted into Eq. (A4), which is further substituted into Eq. (A1) to yield after some manipulation

$$\begin{aligned} -D_1^\xi = {} & \frac{\epsilon_1}{h_1}(\mathbf{n}_\xi \cdot \mathbf{n}_\xi)\frac{\partial \phi_1}{\partial \xi} + \gamma_1 \left\{ \frac{(\mathbf{n}_\xi \times \mathbf{n}_\eta)^z}{h_2}\frac{\partial \phi_1}{\partial \eta} \right. \\ & \left. + \frac{(\mathbf{n}_\xi \times \mathbf{n}_\phi)^z}{h_3}\frac{\partial \phi_1}{\partial \phi} \right\} - \delta_1 \left\{ \frac{(\mathbf{n}_\eta \times \mathbf{n}_\xi)^y}{h_2}\frac{\partial \phi_1}{\partial \eta} \right. \\ & \left. + \frac{(\mathbf{n}_\phi \times \mathbf{n}_\xi)^y}{h_3}\frac{\partial \phi_1}{\partial \phi} \right\} + \zeta_1 \left\{ \frac{(\mathbf{n}_\xi \times \mathbf{n}_\eta)^x}{h_2}\frac{\partial \phi_1}{\partial \eta} \right. \\ & \left. + \frac{(\mathbf{n}_\xi \times \mathbf{n}_\phi)^x}{h_3}\frac{\partial \phi_1}{\partial \phi} \right\}. \end{aligned} \quad \text{(A6)}$$

Here superscripts x, y, and z mean respective components of the vector products. The unitary vectors satisfy

$$\mathbf{n}_\xi \cdot \mathbf{n}_\xi = 1, \quad \text{(A7a)}$$

$$\mathbf{n}_\xi \times \mathbf{n}_\eta = \mathbf{n}_\phi, \quad \mathbf{n}_\eta \times \mathbf{n}_\phi = \mathbf{n}_\xi, \quad \mathbf{n}_\phi \times \mathbf{n}_\xi = \mathbf{n}_\eta. \quad \text{(A7b)}$$

On the surface of the ellipsoid, where $\xi = 0$, we have from Eqs. (2.13) and (A3)

$$(h_1)_0 = \frac{\sqrt{-\eta}}{2ac}, \quad \text{(A8a)}$$

$$(h_2)_0 = \frac{1}{2}\left(\frac{-\eta}{(\eta + a^2)(\eta + c^2)}\right)^{1/2}, \quad \text{(A8b)}$$

$$(h_3)_0 = a\left(\frac{\eta + a^2}{a^2 - c^2}\right)^{1/2}. \quad \text{(A8c)}$$

Substituting Eqs. (A7) and (A8) into Eq. (A6) yields Eq. (2.23).

APPENDIX B: DERIVATION OF ELECTRIC FIELD INDUCED IN MAGNETIZED ELLIPSOID

Let us derive Eq. (2.27), starting from Eq. (2.23).

The partial differential coefficient appearing in the first term of the right-hand side of Eq. (2.23) is calculated from Eq. (2.24) as

$$\begin{aligned} \left(\frac{\partial \phi_1}{\partial \xi}\right)_0 = {} & -\{F^x + c^x A(0)\}\left(\frac{\partial x}{\partial \xi}\right)_0 - c^x \left(\frac{\partial A}{\partial \xi}\right)_0 (x)_0 \\ & -\{F^y + c^y A(0)\}\left(\frac{\partial y}{\partial \xi}\right)_0 - c^y \left(\frac{\partial A}{\partial \xi}\right)_0 (y)_0 \\ & -\{F^z + c^z A'(0)\}\left(\frac{\partial z}{\partial \xi}\right)_0 - c^z \left(\frac{\partial A'}{\partial \xi}\right)_0 (z)_0 . \end{aligned} \quad \text{(B1)}$$

Calculating partial derivatives appearing in the right-hand side of Eq. (B1) from Eqs. (2.13) and (2.26), we obtain

$$\left(\frac{\partial \phi_1}{\partial \xi}\right)_0 = -XK\cos\phi - YK\sin\phi - ZL, \quad \text{(B2)}$$

where we replaced

$$X = F^x + c^x\left\{A(0) - \frac{2}{a^2 c}\right\}, \quad \text{(B3a)}$$

$$Y = F^y + c^y\left\{A(0) - \frac{2}{a^2 c}\right\}, \quad \text{(B3b)}$$

$$Z = F^z + c^z\left\{A'(0) - \frac{2}{c^2 a}\right\}, \quad \text{(B3c)}$$

$$K = \frac{1}{2a}\left(\frac{\eta + a^2}{a^2 - c^2}\right)^{1/2}, \quad \text{(B4a)}$$

$$L = \frac{1}{2c}\left(\frac{\eta + c^2}{c^2 - a^2}\right)^{1/2}. \quad \text{(B4b)}$$

In a similar way, from Eqs. (2.24) and (2.13) we obtain

$$\left(\frac{\partial\phi_1}{\partial\eta}\right)_0 = -X'\left(\frac{\partial x}{\partial\phi}\right)_0 - Y'\left(\frac{\partial y}{\partial\phi}\right)_0 = X'2a^2K\sin\phi - Y'2a^2K\cos\phi, \quad \text{(B5)}$$

$$\left(\frac{\partial\phi_1}{\partial\phi}\right)_0 = -X'\left(\frac{\partial x}{\partial\eta}\right)_0 - Y'\left(\frac{\partial y}{\partial\eta}\right)_0 - Z'\left(\frac{\partial z}{\partial\phi}\right)_0 = -X'a^2(\eta+a^2)^{-1}K\cos\phi - Y'a^2(\eta+a^2)^{-1}K\sin\phi - Z'c^2(\eta+c^2)^{-1}L, \quad \text{(B6)}$$

where we replaced

$$X' = F^x + c^xA(0), \quad Y' = F^y + c^yA(0), \quad Z' = F^z + c^zA'(0). \quad \text{(B7)}$$

Substituting Eqs. (B2), (B5), and (B6) into Eq. (2.23), we obtain

$$(h_1D_1^{\xi})_0 = (\epsilon_1X + \gamma_1Y' - \delta_1Z')K\cos\phi + (-\gamma_1X' + \epsilon_1Y + \zeta_1Z')K\sin\phi + (\delta_1X' - \zeta_1Y' + \epsilon_1Z)L \quad \text{(B8)}$$

after some manipulation.

On comparing Eqs. (2.25) and (2.24), we notice that an equation similar to Eq. (B8) also holds for $\mathbf{D}_2$ as follows:

$$(h_1D_2^{\xi})_0 = (\epsilon_2E_2^x + \gamma_2E_2^y - \delta_2E_2^z)K\cos\phi + (-\gamma_2E_2^x + \epsilon_2E_2^y + \zeta_2E_2^z)K\sin\phi + (\delta_2E_2^x - \zeta_2E_2^y + \epsilon_2E_2^z)L. \quad \text{(B9)}$$

In order to equate Eqs. (B9) and (B8) for all values of η and ϕ, we must have

$$\epsilon_1X + \gamma_1Y' - \delta_1Z' = \epsilon_2E_2^x + \gamma_2E_2^y - \delta_2E_2^z, \quad \text{(B10a)}$$

$$-\gamma_1X' + \epsilon_1Y + \zeta_1Z' = -\gamma_2E_2^x + \epsilon_2E_2^y + \zeta_2E_2^z, \quad \text{(B10b)}$$

$$\delta_1X' - \zeta_1Y' + \epsilon_1Z = \delta_2E_2^x - \zeta_2E_2^y + \epsilon_2E_2^z. \quad \text{(B10c)}$$

In a similar way, the boundary condition of Eq. (2.21) is transformed into

$$-X'a^2K\cos\phi - Y'a^2K\sin\phi - Z'acL = -E_2^xa^2K\cos\phi - E_2^ya^2K\sin\phi - E_2^zacL \quad \text{(B11)}$$

for which to hold for all values of ϕ, we must have

$$X' = E_2^x, \quad Y' = E_2^y, \quad Z' = E_2^z. \quad \text{(B12)}$$

The remaining boundary condition of Eq. (2.22) yields Eq. (B12), as well.

Eliminating c^x, c^y, and c^z by combining Eqs. (B3), (B7), and (B12) with Eq. (B10), we obtain Eq. (2.27).

APPENDIX C: AVERAGE POLARIZABILITY FOR RANDOMLY ORIENTED ELLIPSOIDS

Here we derive Eq. (3.13), the average polarizability for the ith ensemble of the randomly oriented ellipsoids. On referring to Eq. (2.37), the symmetrized polarizability $[\boldsymbol{\alpha}_i]$ for the ith ensemble is expressed in the xyz system as

$$[\boldsymbol{\alpha}_i] = \begin{pmatrix} (\epsilon_i-\hat{\epsilon})/\beta_i & (\gamma_i-\hat{\gamma})/\beta_i^2 & -(\delta_i-\hat{\delta})/(\beta_i\beta_i') \\ -(\gamma_i-\hat{\gamma})/\beta_i^2 & (\epsilon_i-\hat{\epsilon})/\beta_i & (\zeta_i-\hat{\zeta})/(\beta_i\beta_i') \\ (\delta_i-\hat{\delta})/(\beta_i\beta_i') & -(\zeta_i-\hat{\zeta})/(\beta_i\beta_i') & (\epsilon_i-\hat{\epsilon})/\beta_i' \end{pmatrix}. \quad \text{(C1a)}$$

Here

$$\beta_i = 1 + N_i(\epsilon_i - \hat{\epsilon})/\hat{\epsilon}, \quad \text{(C1b)}$$

$$\beta_i' = 1 + N_i'(\epsilon_i - \hat{\epsilon})/\hat{\epsilon}, \quad \text{(C1c)}$$

and $\hat{\zeta}$, $\hat{\delta}$, and $\hat{\gamma}$ are the off-diagonal terms of the symmetrized effective tensor (to be determined) expressed in the x,y,z system. Since the off-diagonal polarizabilities given by Eq. (C1a) are antisymmetric ($\alpha_{mj} = -\alpha_{jm}$, $m \neq j$), they are transformed by $\mathbf{U}$ to be expressed in the x',y',z' system as

$$\alpha'_{mj} = \sum_{k,l=1}^{3} U_{mk}U_{jl}\alpha_{kl} = \sum_{k=1}^{3} U_{mk}U_{jk}\alpha_{kk} + \sum_{k\neq l}{}' (U_{mk}U_{jl} - U_{ml}U_{jk})\alpha_{kl}, \quad \text{(C2)}$$

where we omitted the suffix i for simplicity and $\Sigma'_{k\neq l}$ means summing only for $k,l = 1,2$; 2,3; and 3,1. Equation (C2) is rewritten as

$$\alpha'_{mj} = \sum_{k=1}^{3} U_{mk}U_{jk}\alpha_{kk} + \sum_{k\neq l}{}' (\mathbf{U}_m \times \mathbf{U}_j)_{S(k,l)}\alpha_{kl}, \quad \text{(C3)}$$

where $\mathbf{U}_j (j = 1,2,3)$ is the jth unitary vector, and subscript $S(k,l)$ on the parentheses designates the component of the vector product as follows:

$$S(k,l) = \begin{Bmatrix} 3 & (k,l=1,2 \text{ and } 2,1) \\ 1 & (k,l=2,3 \text{ and } 3,2) \\ 2 & (k,l=3,1 \text{ and } 1,3) \end{Bmatrix}. \quad \text{(C4)}$$

Since the unitary vectors satisfy the relation

$$\mathbf{U}_m \times \mathbf{U}_j = \pm \mathbf{U}_{S(m,j)}, \quad \text{(C5)}$$

where the complex sign takes + for $m,j = 1,2$; 2,3; and 3,1; and − for $m,j = 2,1$; 3,2; and 1,3, Eq. (C3) is transformed to

$$\alpha'_{mj} = \sum_{k=1}^{3} U_{mk}U_{jk}\alpha_{kk} \pm \sum_{k\neq l}{}' U_{S(m,j)S(k,l)}\alpha_{kl}. \quad \text{(C6)}$$

Substituting α_{kl} given by Eq. (C1) into Eq. (C6), we obtain

$$\begin{aligned}\alpha'_{mj}=&(\epsilon_i-\hat{\epsilon})(U_{m1}U_{j1}/\beta_i+U_{m2}U_{j2}/\beta_i+U_{m3}U_{j3}/\beta'_i)\\&\pm\{U_{S(m,j)3}(\gamma_i-\hat{\gamma})/\beta_i^2+U_{S(m,j)2}(\delta_i-\hat{\delta})/(\beta_i\beta'_i)\\&+U_{S(m,j)1}(\zeta_i-\hat{\zeta})/(\beta_i\beta'_i)\}.\end{aligned}\tag{C7}$$

Because $\hat{\zeta}$, $\hat{\delta}$, and $\hat{\gamma}$ (or ζ_i, δ_i, and γ_i) are projection elements of the gyration vector (which is directed along the z axis) to the x', y', and z' axes, respectively, we have the following relations:

$$U_{31}\hat{\Gamma}=\hat{\zeta},\quad U_{32}\hat{\Gamma}=\hat{\delta},\quad U_{33}\hat{\Gamma}=\hat{\gamma}.\tag{C8a}$$

$$U_{31}\Gamma_i=\zeta_i,\quad U_{32}\Gamma_i=\delta_i,\quad U_{33}\Gamma_i=\gamma_i.\tag{C8b}$$

Substituting Eqs. (C8) into Eq. (C7) yields

$$\begin{aligned}\alpha'_{mj}=&(\epsilon_i-\hat{\epsilon})(U_{m1}U_{j1}/\beta_i+U_{m2}U_{j2}/\beta_i+U_{m3}U_{j3}/\beta'_i)\\&\pm(\Gamma_i-\hat{\Gamma})\{U^2_{S(m,j)3}/\beta_i^2+U^2_{S(m,j)2}/(\beta_i\beta'_i)\\&+U^2_{S(m,j)1}/(\beta_i\beta'_i)\}.\end{aligned}\tag{C9}$$

Averaging the polarizability over all ellipsoids is equivalent to averaging over all unitary transformations which transform rotational axes of all ellipsoids to the z' direction. We now assume that the randomness of the orientation of the ellipsoids in the ensemble is so complete that averaging over all unitary transformations is equivalent to averaging U_{mj} in Eq. (C9) over all directions or solid angles. Then we have the averages (denoted by $\langle\rangle$) of the polarizability elements as

$$\begin{aligned}[\hat{\alpha}_i]_{mj}=&\langle\alpha'_{mj}\rangle\\=&(\epsilon_i-\hat{\epsilon})\{\langle U_{m1}U_{j1}\rangle/\beta_i+\langle U_{m2}U_{j2}\rangle/\beta_i\\&+\langle U_{m3}U_{j3}\rangle/\beta'_i\}\pm(\Gamma_i-\hat{\Gamma})\{\langle U^2_{S(m,j)3}\rangle/\beta_i^2\\&+\langle U^2_{S(m,j)2}\rangle/(\beta_i\beta'_i)+\langle U^2_{S(m,j)1}\rangle/(\beta_i\beta'_i)\}.\end{aligned}\tag{C10}$$

Since U_{mj} is the mth component of the jth unitary vector, we can write $\mathbf{U}_j=(x_1/r,x_2/r,x_3/r)$, where $r=(x_1^2+x_2^2+x_3^2)^{1/2}$. Then we have

$$\langle U_{mk}U_{jk}\rangle=\int(x_mx_j/r^2)d\Omega\Bigg/\int d\Omega=\delta_{mj}/3,\tag{C11}$$

where integration is performed over the total range of solid angle Ω, and δ_{mj} is Kronecker's delta. Substituting Eq. (C11) into Eq. (C10) yields Eq. (3.13).

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