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The permeability tensor of composite consisting of magnetic particles

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The demagnetizing energy of spheroidal magnetic particles dispersed in cubic lattices has been calculated. The demagnetizing energy contains contributions from the demagnetizing self-energy and magnetostatic coupling energy between particles. The total demagnetizing energy can be conveniently expressed in terms of three demagnetizing factors whose sum equals unity. For low volume loading of particles the self-energy contribution is the dominant one, which is insensitive to particle coordinations and can be approximated by that of an isolated particle. However, when particle loading increases appreciably, significant differences arise for different particle lattices (sc, bcc, and fcc). One advantage of our formulation is that the permeability tensor of the composite can then be calculated for either a magnetic nonsaturated or saturated state of the particle. In contrast to conductivity calculations at the percolation limit there is no divergence of the permeability.

INTRODUCTION

The study of composite materials has been the subject of considerable interest in recent years.¹ In this paper we consider microwave applications of composite materials consisting of magnetic particles embedded in a binder matrix. Besides the permittivity of the composite, the magnetic permeability is of crucial importance regarding the usefulness of materials, in general, for microwave applications. We assume that the particles are arranged in the matrix in an idealized manner such that the particles form a cubic lattice. Generally, there are two ways to calculate the properties of the composite. One way is to expand the static field in multipoles surrounding the particles such that the multipole coefficients are determined by matching the field at the particle surfaces. This approach was developed by Rayleigh² and Doyle³ in calculation of the permittivity for metallic particles embedded in a dielectric binder. The second approach proposed by us is to describe the interaction between particles in terms of the free energy of the system. By adding up all the free-energy terms, the motion of the magnetization vector within the magnetic particles can be determined which gives rise to the permeability tensor of the composite. A similar approach was used by Artman and Charap⁴ and Vittoria and Rachford⁵ in describing the microwave properties of magnetic bubble materials. Among the two methods of calculations, the latter one proves to be more general, since other interactions and fields, e.g., elastic coupling and anisotropy field, can be readily added into the free energy formulation. However, both methods are applicable only in the long wavelength regime where particles in close proximity are treated identically.

CALCULATIONS

Let z be the revolution axis of the spheroidal particles, and μ_1 and 1 be the permeabilities of the particle and the binder, respectively. All the lengths are normalized in terms of the length of the conventional unit cell of the particle lattice. Denote a and b as the semimajor and semiminor axes. The analysis below utilizes a formalism

which is generalized from Ref. 6 and Gaussian units are used throughout. Let the magnetization vector be $\mathbf{M}(x, y, z)$. Assume the magnetization distribution possesses inversion symmetry, $\mathbf{M}(\mathbf{r}) = \mathbf{M}(-\mathbf{r})$. Similar to results obtained in Ref. 6, the demagnetizing energy density associated with $\mathbf{M}(\mathbf{r})$ can be written as

$$E_D = 2\pi \sum_{\alpha=1}^3 \sum_{\mathbf{k}}' [C_{\mathbf{k}}^{(\alpha)} k_{\alpha}/k]^2, \quad (1)$$

where $C_{\mathbf{k}}^{(\alpha)}$ denotes Fourier transform of M_{α} (over one unit cell) and $\mathbf{k} = 2\pi(p\hat{x} + q\hat{y} + r\hat{z})$ is the wave vector with $p, q,$ and r being integers from $-\infty$ to ∞ . Here Σ' denotes summation over all the \mathbf{k} vectors except the term $\mathbf{k} = 0$. For uniform magnetization of the particles the magnetization vector $\mathbf{M}(\mathbf{r})$ has the form $\mathbf{M}(\mathbf{r}) = M_{0x}\hat{x} + M_{0y}\hat{y} + M_{0z}\hat{z}$, for \mathbf{r} located within the particles, and $\mathbf{M} = 0$, otherwise. It turns out that

$$C_{\mathbf{k}}^{(\alpha)} = M_{0\alpha} I_{\mathbf{k}} F_{\mathbf{k}} \quad (2)$$

with the form factor $F_{\mathbf{k}}$ given by $1, 1 + (-1)^{p+q+r}$, and $1 + (-1)^{p+q} + (-1)^{q+r} + (-1)^{r+p}$ for sc, bcc, and fcc lattices, respectively. $I_{\mathbf{k}}$ is related to the Fourier integral of a single particle as

$$I_{\mathbf{k}} = \int \int \int_{\text{single particle}} d\tau e^{i\mathbf{k}\cdot\mathbf{r}}$$

For a prolate $I_{\mathbf{k}}$ can be calculated as

$$I_{\mathbf{k}} = \int_c^a ds \left[\left[s^2 + \left(-\frac{k_z^2 s^2 + 1}{Z^2} + \frac{3k_z^2 s^2}{Z^4} \right) c^2 \right] \frac{\sin Z}{Z} + \left(\frac{1}{Z^2} - \frac{3k_z^2 s^2}{Z^4} \right) c^2 \cos Z \right], \quad (3)$$

where c is the focal length defined by $c = (a^2 - b^2)^{1/2}$ and $Z = [k^2 s^2 - (k_x^2 + k_y^2) c^2]^{1/2}$. For an oblate the above integral is changed by replacing c with $-ic$ and the integration limits " c to a " with " 0 to b ." Denote v as the volume fraction of the particles and n as the number of particles per unit cell ($n=1, 2,$ and 4 for sc, bcc, and fcc lattices, respectively). We define the demagnetizing factor N_{α} to be

$$N_\alpha = \frac{n^2}{v(1-v)} \sum_k \left| \frac{I_k F_k k_\alpha}{k} \right|^2 \quad (4)$$

It can be verified explicitly that $N_x + N_y + N_z = 1$. In terms of N_α the demagnetizing energy density E_D in Eq. (1) can be rewritten as

$$E_D = \frac{2\pi v(1-v)}{n^2} \sum_{\alpha=1}^3 N_\alpha M_{0\alpha}^2 \quad (5)$$

In the following an expression for the permeability tensor of the particle composite is derived utilizing Eq. (5). Assume the total free energy density of the system is composed of the Zeeman energy, the anisotropy energy, and the demagnetizing energy terms as

$$F = -vH_0M_x + vK(M_x^2 + M_y^2)/M_s^2 + E_D, \quad (6)$$

where M_s , K , and H_0 are, respectively, the saturation magnetization, the anisotropy constant, and the external field applied in the x direction. Using the same calculational method as in Ref. 5, it can be shown that the permeability tensor is of the following form:

$$\mu = \begin{pmatrix} \mu_1 & ik & 0 \\ -ik & \mu_2 & 0 \\ 0 & 0 & 1 \end{pmatrix}, \quad (7)$$

where $\mu_1 = 1 + 4\pi v M_s A / (\Omega^2 + AB)$, $\mu_2 = 1 + 4\pi v M_s B / (\Omega^2 + AB)$, $ik = 4\pi v M_s \Omega / (\Omega^2 + AB)$, $\Omega = i\omega/\gamma$, $A = H_0/\sin\theta_0 - i\Delta H/2$, $B = |H_A + H_N - H_0 \sin\theta_0| - i\Delta H/2$, and the linewidth ΔH is given by $\Delta H = 2\lambda\omega/\gamma^2 M_s$ with λ and γ being the Landau-Lifshitz damping constant and gyromagnetic ratio, respectively. Here the spherical coordinates θ_0 and ϕ_0 define the equilibrium configuration of the magnetization as

$$\phi_0 = 0, \quad (8a)$$

$$\theta_0 = \sin^{-1} [H_0 / (H_A + H_N)] \text{ for } H_0 < H_A + H_N \\ = \pi/2 \text{ otherwise,} \quad (8b)$$

and H_A and H_N are the effective fields associated with anisotropy and demagnetization given by $H_A = 2K/M_s$ and $H_N = 4\pi(1-v)(N_x - N_z)M_s/n^2$. For $H_0 < H_A + H_N$ the magnetization is not aligned along the applied field, and for $H_0 > H_A + H_N$ the magnetization is saturated and along H_0 . For a composite consisting of oblate particles it is possible to have $H_A + H_N < 0$. In this case the magnetization aligned in the \hat{z} axis is metastable; an infinitesimal application of H_0 will rotate the magnetization into the field direction. The permeability tensor of Eq. (7) is defined with respect to a coordinate system in which the z axis is parallel to the static magnetization defined by θ_0 and ϕ_0 . Note that μ is Hermitian if and only if A and B are real, i.e., the particle medium is lossless.

CALCULATIONAL RESULTS

For large k values the summation over k in Eq. (4) converges very slowly. If p , q , and r are taken from $-N$ to $+N$ in the sum, convergence within 90% was achieved when N was taken to be 100. However, we notice that the

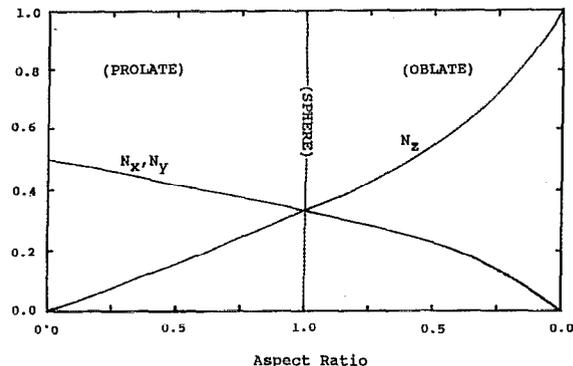


FIG. 1. Demagnetizing factors as functions of the aspect ratio.

convergence is inversely proportional to N , and, when the sums have been extrapolated by $N=50$ and $N=100$, the demagnetizing factors N_x , N_y , and N_z can be evaluated with errors less than a few parts in 1000. Errors can be estimated by comparing the sum of N_x , N_y , and N_z with unity. Figure 1 shows the dependences of N_x , N_y , and N_z on the aspect ratio $A=b/a$ for simple cubic coordination. In Fig. 1 a is fixed to be 0.3 and b varies from 0 to 0.3 (the prolate case) and then from 0.3 to 0 (the oblate case). Since the loading of particles in Fig. 1 is quite low, we may approximate the curves in Fig. 1 by those associated with an isolated spheroid⁷:

$$N_z = (1 - e^2)(\tanh^{-1} e - e)/e^3 \text{ for a prolate,} \quad (9a)$$

$$= (1 + e^2)(e - \tan^{-1} e)/e^3 \text{ for an oblate,} \quad (9b)$$

and $N_x = N_y = (1 - N_z)/2$. Here e is the eccentricity of the particle defined as c/a and c/b for a prolate and an oblate, respectively. Almost indistinguishable results are obtained for the same parameters used in Fig. 1 if bcc and fcc coordinations are considered, as we might expect.

Figure 2 shows the dependence of N_x , N_y , and N_z on the packing density of particles in the sc, bcc, and fcc structures for the prolate case. Here b is taken as $a/2$ for the constant aspect ratio. In the dilute situations where a is small, the demagnetizing factors are almost independent of the particle concentration and coordination. When a in-

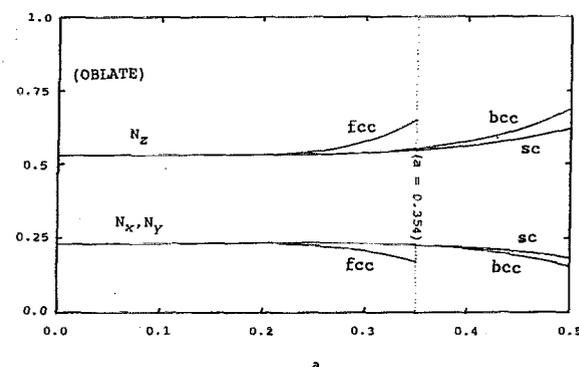


FIG. 2. Demagnetizing factors as functions of a for $b=a/2$: oblate case.

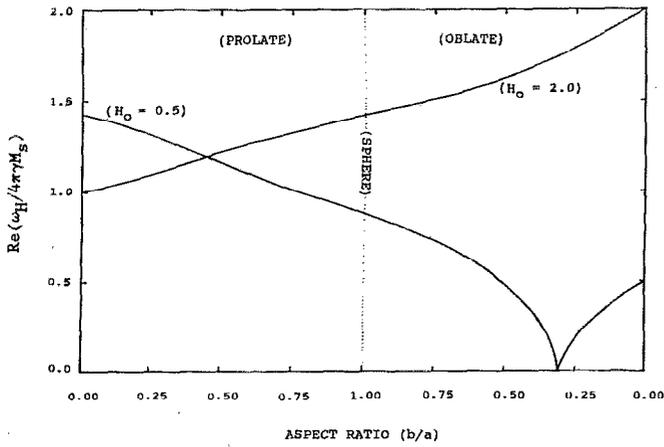


FIG. 3. Resonant frequency as functions of the particle aspect ratio.

creases, the curves gradually depart from their dilute values until the packing limits are finally reached. For prolates, the packing limits are $a=0.5$, 0.5 , and 0.447 for the sc, bcc, and fcc lattices, respectively. Unlike the dielectric-metallic transition found in the percolation limit in a metal-particle composite, there are no catastrophic changes in the values of N_x , N_y , and N_z when the same particle loading limit is reached. However, when the percolation limit is passed over, Eq. (4) still provides values for N_x , N_y , and N_z except that their sum is no longer equal to unity.

Figure 3 shows the resonant frequency as a function of the particle's aspect ratio $A=b/a$ for a fixed to be 0.3 . b is taken from 0 to 0.3 (prolate case) and 0.3 to 0 (the oblate case). The resonant frequency is defined as the zeroing condition of the denominators in μ_1 , μ_2 , and ik , i.e., $\Omega^2 + AB = 0$. In Fig. 3 (as well as Fig. 4) we take $4\pi M_s \approx H_A \approx 3000$ G, and $\Delta H \approx 100$ Oe (at 9 GHz), which correspond to substituted hexagonal barium ferrite particles. The external field H_0 is normalized with respect to $4\pi\gamma M_s$. The particle coordination of Fig. 3 (as well as Fig.

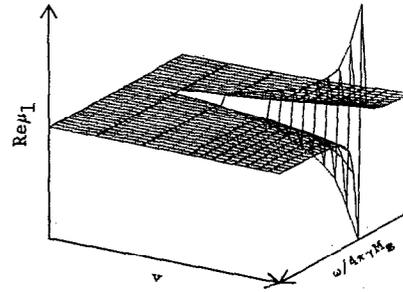


FIG. 4. Real part of μ_1 as a function of v and $\omega/4\pi\gamma M_s$.

4) is taken to be simple cubic. Two H_0 values ($H_0 = 0.5$ and 2) are used in Fig. 3. For $H_0 = 2$ the particles are saturated for all the values of aspect ratio and the resonant frequency dictates a smooth function over A . For $H_0 = 0.5$ the particle's magnetization is nonsaturated for prolates. It can be saturated for disk-like oblates as discussed previously following Eqs. (8a) and (8b). This is shown in Fig. 3 as a cusp ($A=0.305$) which separates the $H_0 = 0.5$ curve in two part: nonsaturated (left) and saturated (right). Figure 4 shows the real part of μ_1 as a function of particle volume fraction, v , and frequency, $\omega/4\pi\gamma M_s$. The particles are prolates with $b=a/2$, and the external field H_0 is taken to be 0.5 . In Fig. 4 v is taken from 0 to 0.13 (maximum particle loading in this case), and $\omega/4\pi\gamma M_s$ from 0 to 0.2 . From Fig. 4 it is seen that resonance is most pronounced when the particle loading is high, and the permeability values have little dependence on v as v is small, again, the situation of dilute particle loading.

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