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Effective magnetic permeability of granular ferromagnetic metals

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Abstract. The effective magnetic permeability μ and effective dielectric constant ϵ are two equally important quantities for the characterization of electromagnetic properties of composites. For granular ferromagnetic metals, however, the presence of permanent magnetic moments means that the Maxwell–Garnett theory is inapplicable to the calculation of effective μ. By extending the approach of Onsager, we derive a simple equation for μ whose numerical solution in the case of granular Ni shows significant deviation from unity in far-infrared and microwave frequencies. The behaviour of μ is predicted to be a sensitive function of particle size and temperature, thus making the material potentially usable as an absorber of far-infrared or microwave radiation.

1. Introduction

When the scale of inhomogeneities in a composite material is much less than the wavelength of the probing electromagnetic radiation, lack of scattering makes the composite appear as a homogeneous medium characterized by effective material properties. The function of an effective medium theory is to relate the effective material properties to those of the components and their microstructures. In the case of the effective dielectric constant of metallic particles dispersed in an insulating matrix, the well known Maxwell–Garnett theory has been successful in achieving this role (Garnett 1904). Theoretically, the effective magnetic permeability is a quantity exactly analogous to the dielectric constant and should therefore be calculable by the same approach (Lamb *et al* 1980). However, to the extent that the Maxwell–Garnett theory is based on the Clausius–Mossotti (CM) relation, it cannot describe the case in which one component of the composite possesses permanent moments. This is due to the problem of polarization catastrophe where the CM relation predicts a polarization divergence for all materials possessing permanent moments, but this is rarely seen. To resolve this problem, Onsager (1936) presented a new, albeit slightly more complicated theory where the role of permanent moments is properly taken into account. It is the purpose of this paper to present a simple extension of the Onsager theory for the calculation of the effective magnetic permeability of granular ferromagnetic metals. The theory takes into account both the induced magnetic moment of a conducting particle as well as the orientational alignment of ferromagnetic moments.

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2. Formulation

Consider a composite consisting of metallic ferromagnetic particles of size 50–200 Å dispersed in an insulating matrix. The composite is assumed to be at a temperature much lower than the ferromagnetic transition temperature so that saturation magnetization is achieved in each particle. Furthermore, the grains are assumed to be sufficiently small so that each grain has only one magnetic domain. Since the wavelength of electromagnetic radiation in the optical to far-infrared range is orders of magnitude larger than either the grain size or the average inter-grain separation, it follows that one can use the quasi-static approximation for the treatment of the electromagnetic field with regard to its interaction with the metallic ferromagnetic particles.

In a quasi-static magnetic field, the magnetization of the composite can result from two sources: (i) the orientational alignment of the ferromagnetic moments, and (ii) the induced magnetic moments due to the eddy currents. To proceed with the formulation of the problem, let us first focus our attention on a single particle of radius a. The magnetization is given by

$$\boldsymbol{m} = m_0\hat{\boldsymbol{u}} + \alpha \boldsymbol{F} \tag{1}$$

where m_0 is the ferromagnetic moment of the particle, $\hat{\boldsymbol{u}}$ is a unit vector along the direction of the ferromagnetic moment, α is the magnetic polarizability of the particle, and $\boldsymbol{F}$ is the local magnetic field. The quantity α has been calculated in the literature (Landau and Lifshitz 1960) and is given by

$$\alpha = (4\pi/3)a^3(\tilde{\alpha}' + \mathrm{i}\tilde{\alpha}'') \tag{2a}$$

$$\tilde{\alpha}' = -\frac{3}{8\pi}\left(1 - \frac{3}{2}\frac{1}{x}\frac{\sinh(2x) - \sin(2x)}{\cosh(2x) - \cos(2x)}\right) \tag{2b}$$

$$\tilde{\alpha}'' = -\frac{9}{16\pi x^2}\left(1 - x\frac{\sinh(2x) + \sin(2x)}{\cosh(2x) - \cos(2x)}\right) \tag{2c}$$

$x = a/\delta$, $\delta = c/\sqrt{(2\pi\sigma\omega)}$ is the skin depth, c denotes the speed of light, σ denotes the conductivity of the particle, and we have taken the relative magnetic permeability of pure metal to be 1. Onsager's insightful contribution (Onsager 1936) lies in his analysis of the local field $\boldsymbol{F}$ as composed of two components: the cavity-field component $\boldsymbol{G}$ and the reaction-field component $\boldsymbol{R}$. Both components can be obtained by solving well defined boundary-value problems. The results, when combined together, give

$$\boldsymbol{F} = \boldsymbol{G} + \boldsymbol{R} = [3\mu/(2\mu + 1)]\boldsymbol{H} + [2(\mu - 1)/(2\mu + 1)a^3]\boldsymbol{m}. \tag{3}$$

Here $\boldsymbol{H}$ is the externally applied magnetic field and μ is the effective magnetic permeability. Substitution of equation (3) into equation (1) gives

$$\boldsymbol{m} = m_0\left(1 - \frac{8\pi(\mu - 1)\tilde{\alpha}}{3(2\mu + 1)}\right)^{-1}\hat{\boldsymbol{u}} + \frac{3\mu}{2\mu + 1}\alpha\left(1 - \frac{8\pi(\mu - 1)\tilde{\alpha}}{3(2\mu + 1)}\right)^{-1}\boldsymbol{H}. \tag{4}$$

The coefficient of $\hat{\boldsymbol{u}}$ in equation (4) is seen to differ from m_0 mainly due to the reaction-field effect, which renormalizes m_0. If the material is made completely

of ferromagnetic metallic particles, then $M = N\langle m\rangle$, where N is the number of particles per unit volume, and $\langle\ \rangle$ denotes thermodynamic averaging. By definition

$$M = [(\mu - 1)/4\pi]H$$

which means

$$\frac{\mu - 1}{4\pi}H = \frac{\overline{m_0}\langle\hat{u}\rangle 3(2\mu + 1) + 9\mu\tilde{\alpha}H}{3(2\mu + 1) - 8\pi(\mu - 1)\tilde{\alpha}}. \tag{5a}$$

Here $\overline{m_0}$ is the saturation magnetization, or magnetic moment per unit volume. The quantity $\langle\hat{u}\rangle$ is calculated by following Onsager's approach. We obtain

$$\langle\hat{u}\rangle = \frac{\mu\overline{m_0}4\pi a^3 H}{kT[3(2\mu + 1) - 8\pi(\mu - 1)\tilde{\alpha}]} \tag{5b}$$

for a DC applied field. Here k is Boltzmann's constant, and T is the temperature. The main point to be noted in the derivation of equation (5*b*) is that only the cavity-field component G has any effect on the moment. The reaction field R is always parallel to m and therefore exerts no torque on the particle. At finite frequencies, $\langle\hat{u}\rangle$ has to be multiplied by a factor $g(\omega)$ due to the finite orientational relaxation time of the magnetic moments, where

$$g(\omega) = (1 - i\omega\tau)^{-1} \tag{5c}$$

represents the Debye relaxation response function, and

$$\tau = \tau_0 \exp(KV/kT) \tag{5d}$$

is the relaxation time for the orientational alignment of the ferromagnetic moments (Xiao *et al* 1986). In equation (5*d*), K represents the anisotropy energy per unit volume, and $V = 4\pi a^3/3$ is the volume of the particle. By substituting equation (5*b*), with the additional $g(\omega)$ factor, into equation (5*a*), a self-consistent equation for μ is obtained:

$$\frac{\mu - 1}{4\pi} = \frac{9\mu(2\mu + 1)\,(\overline{m_0^2}4\pi a^3/3kT)}{[3(2\mu + 1) - 8\pi(\mu - 1)\tilde{\alpha}]^2} + \frac{9\mu\tilde{\alpha}}{3(2\mu + 1) - 8\pi(\mu - 1)\tilde{\alpha}}.$$

At this point we would like to make the transition to the case of a composite. It is noted that whereas α is the induced magnetic polarizability, $m_0^2/3kT$ represents the polarizability of permanent magnetic moments through orientational alignment. Both have the dimension of volume. This means that in a composite where the ferromagnetic particles occupy only a fraction of the total volume, the dimensionless quantities $\tilde{\alpha}$ and $(4\pi a^3\overline{m_0^2}/9kT)$, which are normalized per unit volume, should be written as $f\tilde{\alpha}$ and $f(4\pi a^3\overline{m_0^2}/9kT)$, respectively. Therefore, the equation for the effective μ of the composite is

$$\frac{\mu - 1}{4\pi} = \frac{9f\mu(2\mu + 1)\,(4\pi a^3\overline{m_0^2}/3kT)}{[3(2\mu + 1) - 8\pi(\mu - 1)f\tilde{\alpha}]^2} + \frac{9f\mu\tilde{\alpha}}{3(2\mu + 1) - 8\pi(\mu - 1)\tilde{\alpha}f}. \tag{6}$$

Equation (6) can be rearranged to give a third-order polynomial equation for μ with complex coefficients:

$$A\mu^3 + B\mu^2 + C\mu + D = 0 \tag{7a}$$

$$A = 4\left(9 + 16\pi^2 f^2 \tilde{\alpha}^2 - 24\pi f \tilde{\alpha}\right) \tag{7b}$$

$$B = -24\pi f\left[\tilde{\alpha}\left(3 - 4\pi f \tilde{\alpha}\right) + 3Rg(\omega)\right] \tag{7c}$$

$$C = -3\left[32\pi^2 \tilde{\alpha}^2 f^2 + 12\pi f\left(Rg(\omega) + 3\tilde{\alpha}\right) + 9\right] \tag{7d}$$

$$D = -\left[9 + 16\pi\tilde{\alpha} f\left(3 + 4\pi\tilde{\alpha} f\right)\right] \tag{7e}$$

and $R = (4\pi a^3 \tilde{m}_0^2/3kT)$. Since there are three solutions to equation (7), the physical one is selected by the condition that $\mathrm{Re}\mu \geqslant 0$ and $\mathrm{Im}\mu \geqslant 0$. If one sets $R = 0$, it is easy to verify that μ satisfies the Maxwell–Garnett theory, or the CM relation, $(\mu - 1)/(\mu + 2) = 4\pi\tilde{\alpha} f/3$, as expected.

3. Numerical solution for granular Ni

We consider granular Ni with particle size of the order of 100 Å ($a = 50$ Å). For Ni, $\sigma = 1.43 \times 10^5\ (\Omega\ \mathrm{cm})^{-1}$, $\overline{m_0} \simeq 480$ G so that $R \simeq 3$ at room temperature, and $\tau \approx 48 \times 10^{-13}$ s is obtained from equation (5*d*) with $\tau_0 \simeq 10^{-13}$ s (Xiao *et al* 1986), $K \simeq 5 \times 10^4$ erg cm^{-3} (Bozorth, 1951) and $T = 300$ K. In figure 1(*a*) we show the real and imaginary parts of μ as a function of f for $\lambda = 500$ μm. For comparison, we plot in figure 1(*b*) the μ obtained from the CM relation with $\tilde{\alpha}$ replaced by $(\tilde{\alpha} + Rg/3)$. The difference in predicted behaviours is striking. For the CM case, the real part even changes sign as a function of f. This is unphysical, demonstrating the inapplicability of the CM relation in the present case.

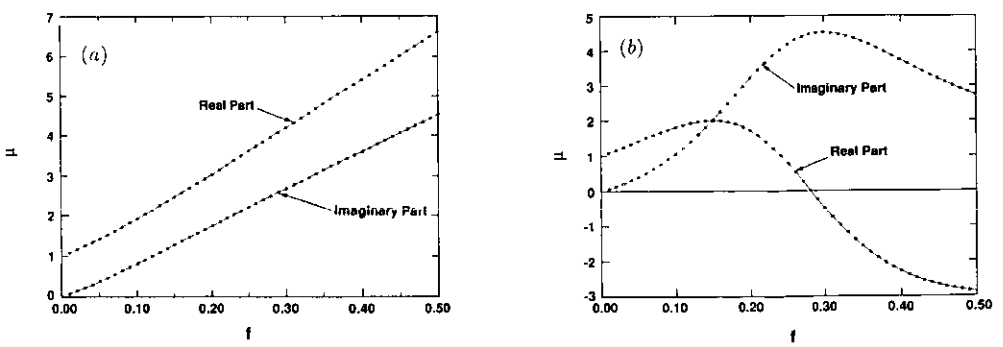

Figure 1. The effective magnetic permeability μ at 500 μm wavelength, plotted as a function of ferromagnetic metal volume fraction f. (*a*) Real and imaginary parts of μ calculated from the present theory with Ni parameters given in the text. (*b*) Real and imaginary parts of μ calculated from the direct extension of the Clausius–Mossotti relation.

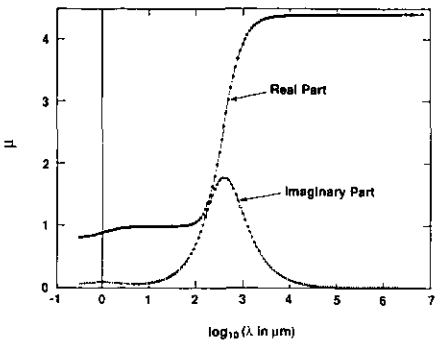

Figure 2. The effective magnetic permeability μ calculated at $f = 0.2$ as a function of wavelength, with parameters pertinent to 100 Å Ni particles at room temperature.

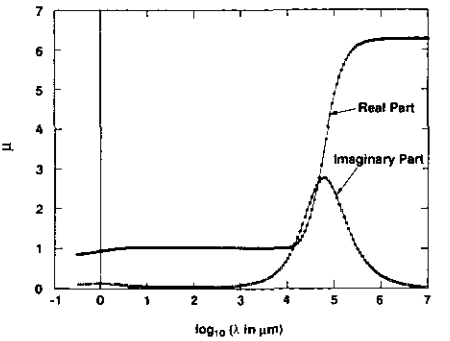

Figure 3. The effective magnetic permeability μ calculated at $f = 0.2$ as a function of wavelength, with parameters pertinent to 100 Å Ni particles at $T = 200$ K.

In figure 2, the frequency dependence of μ is shown for $f = 0.2$. At $\lambda \simeq 1$ μm the real part of μ is seen to be less than 1, with an attendant increase in $\mathrm{Im}\,\mu$. This is due to the induced magnetic moment associated with the eddy currents. Since the effect is diamagnetic, it accounts for the slight decrease in $\mathrm{Re}\,\mu$ from unity. The most significant feature, however, is the large deviation from unity at far-infrared frequencies, which is due to the orientational alignment of the ferromagnetic moments. Since τ and R are both functions of temperature T and particle size a, both the magnitude and frequency dependence of μ at this frequency regime can be tuned by varying these parameters. For example, by lowering the temperature, one expects the peak of $\mathrm{Im}\,\mu$ to shift to microwave frequencies. At the same time, lowering the temperature also has the effect of increasing R. One thus expects the magnitude of the $\mathrm{Im}\,\mu$ peak to increase as T decreases. Figure 3 illustrates this temperature effect through the frequency dependence of μ at $f = 0.2$ and $T = 200$ K, where $\tau = 3 \times 10^{-11}$ s is obtained from equation (5*d*) with $\tau_0 \simeq 10^{-13}$ s and $K \simeq 3 \times 10^5$ erg cm^{-3} at this temperature (Bozorth, 1951). The value of R is increased to 4.5. One sees directly that the magnetic absorption peak is now in the microwave regime, and the effect is larger than that at 300 K. These results thus provide direct predictions on the behaviour of ferromagnetic granular metals that could be experimentally tested.

4. Concluding remarks

By following Onsager's approach, we have derived an equation for the effective magnetic permeability of granular ferromagnetic metal whose predictions differ significantly from those of the Maxwell–Garnett theory, or the CM relation. Numerical calculation shows granular ferromagnetic metals to be a temperature-tunable absorber of far-infrared and microwave radiations. Experimental verification of these predictions is presently underway.

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