Theoretical investigation on the possibility of preparing left-handed materials in metallic magnetic granular composites

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We investigate the possibility of preparing left-handed materials in metallic magnetic granular composites. Based on the effective medium approximation, we show that by incorporating metallic magnetic nanoparticles into an appropriate insulating matrix and controlling the directions of magnetization of metallic magnetic components and their volume fraction, it may be possible to prepare a composite medium of low eddy current loss that is left-handed for electromagnetic waves propagating in some special direction and polarization in a frequency region near the ferromagnetic resonance frequency. This composite may be easier to make on an industrial scale. In addition, its physical properties may be easily tuned by rotating the magnetization locally.

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In classical electrodynamics, the response (typically frequency dependent) of a material to electric and magnetic fields is characterized by two fundamental quantities, the permittivity ϵ and the permeability μ. The permittivity relates the electric displacement field $\vec{D}$ to the electric field $\vec{E}$ through $\vec{D}=\epsilon\vec{E}$, and the permeability μ relates the magnetic field $\vec{B}$ and $\vec{H}$ by $\vec{B}=\mu\vec{H}$. If we do not take losses into account and treat ϵ and μ as real numbers, according to Maxwell's equations, electromagnetic waves can propagate through a material only if the index of refraction *n*, given by $(\epsilon\mu)^{1/2}$, is real. (Dissipation will add imaginary components to ϵ and μ and cause losses, but for a qualitative picture, one can ignore losses and treat ϵ and μ as real numbers. Also, strictly speaking, ϵ and μ are second-rank tensors, but they reduce to scalars for isotropic materials.) In a medium with ϵ and μ both positive, the index of refraction is real and electromagnetic waves can propagate. All our everyday transparent materials are such kind of media. In a medium where one of the ϵ and μ is negative but the other is positive, the index of refraction is imaginary and electromagnetic waves cannot propagate. Metals and Earth's ionosphere are such kind of media. Metals and the ionosphere have free electrons that have a natural frequency—the plasma frequency—which is on the order of 10 MHz in the ionosphere and falls at or above visible frequencies for most metals. At frequencies above the plasma frequency, ϵ is positive and electromagnetic waves are transmitted. For lower frequencies, ϵ becomes negative and the index of refraction is imaginary, and consequently electromagnetic waves cannot propagate through. In fact, the electromagnetic response of metals in the visible and near-ultraviolet regions is dominated by the negative epsilon concept.[1–4]

Although all our everyday transparent materials have both positive ϵ and positive μ, from the theoretical point of view, in a medium with ϵ and μ both negative, the index of refraction is also positive and electromagnetic waves can also propagate through. Moreover, if such media exist, the propagation of waves through them should give rise to several peculiar properties. This was first pointed out by Veselago over 30 years ago when no material with simultaneously negative ϵ and μ was known.[5] For example, the cross product of $\vec{E}$ and $\vec{H}$ for a plane wave in regular media gives the direction of both propagation and energy flow, and the electric field $\vec{E}$, the magnetic field $\vec{H}$, and the wave vector $\vec{k}$ form a right-handed triplet of vectors. In contrast, in a medium with ϵ and μ both negative, $\vec{E}\times\vec{H}$ for a plane wave still gives the direction of energy flow, but the wave itself (that is, the phase velocity) propagates in the opposite direction, i.e., wave vector $\vec{k}$ lies in the opposite direction of $\vec{E}\times\vec{H}$ for propagating waves. In this case, electric field $\vec{E}$, magnetic field $\vec{H}$, and wave vector $\vec{k}$ form a left-handed triplet of vectors. Such a medium is therefore termed left-handed medium.[5] In addition to this "left-handed" characteristic, there are a number of other dramatically different propagation characteristics stemming from a simultaneous change of the signs of ϵ and μ, including reversal of both the Doppler shift and the Čerenkov radiation, anomalous refraction, and even reversal of radiation pressure to radiation tension. However, although these counterintuitive properties follow directly from Maxwell's equations, which still hold in these unusual materials, such type of left-handed materials have never been found in nature and these peculiar propagation properties have never been demonstrated experimentally. If such media can be prepared artificially, they will offer exciting opportunities to explore new physics and potential applications in the area of radiation-material interactions. Recently, interesting progress has been achieved in preparing a "left-handed" material artificially. Following the suggestion of Pendry *et al.*[1] Smith and co-workers reported that a medium made up of an array of conducting nonmagnetic split ring resonators and continuous thin wires can have both an effective negative permittivity ϵ and negative permeability μ for electromagnetic waves propagating in some special direction and special polarization at microwave frequencies.[6] This is the first experimental realization of an artificial preparation of a left-handed material. Motivated by this progress, in this paper, we propose to investigate the possibility of preparing left-handed materials in another type of system—metallic magnetic granular composites. The idea is that, by incorporating metallic ferromagnetic nanoparticles into an appropriate insulating matrix and controlling the directions of magnetization of metallic magnetic particles and their volume fraction, it may be possible to prepare a composite medium that has simultaneously negative ϵ and negative μ and low eddy current loss. This idea was based on the fact that on the one hand, the permittivity of metallic particles is automatically negative at frequencies less than the plasma frequency, and on the other hand, the effective permeability of ferromagnetic materials for electromagnetic waves propagating in some particular direction and polarization can be negative at frequency in the vicinity of the ferromagnetic resonance frequency ω_0, which is usually in the frequency region of microwaves. So, if we can prepare a composite medium in which one component is both metallic and ferromagnetic and the other component insulating, and we can control the directions of magnetization of metallic magnetic particles and their volume fraction, it may be possible to achieve a left-handed composite medium of low eddy current losses for electromagnetic waves propagating in some special direction and polarization. This composite may be easier to make on an industrial scale. In addition, its physical properties may be easily tuned by rotating the magnetization locally.

To illustrate the above idea more clearly, in the following we present some results of our model calculations based on the effective medium theory. Let us consider an idealized metallic magnetic granular composite consisting of two types of spherical particles, in which one type of particle is a metallic ferromagnetic grain of radius R_1, and the other type is a nonmagnetic dielectric (insulating) grain of radius R_2. Each grain is assumed to be homogeneous. The directions of magnetization of all metallic magnetic grains are assumed to be in the same direction. In length scales much larger than the grain sizes, the composite can be considered as a homogeneous magnetic system. The permittivity and permeability of nonmagnetic dielectric grains are both scalars, and will be denoted as ϵ_1 and μ_1. The permittivity of metallic magnetic grains will be denoted as ϵ_2 and will be taken to have a Drude form: $\epsilon_2 = 1-\omega_p^2/\omega(\omega+i/\tau)$, where ω_p is the plasma frequency of the metal and τ is a relaxation time. Such a form of ϵ is representative of a variety of metal composites.[8,9] The permeability of metallic magnetic grains are second-rank tensors and will be denoted as $\hat{\mu}_2$, which can be derived from the Landau-Lifschitz equations.[7] Assuming that the directions of magnetization of all magnetic grains are in the direction of the z axis, $\hat{\mu}_2$ will have the following form:[7]

$$\hat{\mu}_2 = \begin{bmatrix} \mu_a & -i\mu' & 0 \\ i\mu' & \mu_a & 0 \\ 0 & 0 & 1 \end{bmatrix}, \tag{1}$$

where

$$\mu_a = 1 + \frac{\omega_m(\omega_0 + i\alpha\omega)}{(\omega_0+i\alpha\omega)^2-\omega^2}, \tag{2}$$

$$\mu' = -\frac{\omega_m\omega}{(\omega_0+i\alpha\omega)^2-\omega^2}, \tag{3}$$

where $\omega_0 = \gamma\vec{H}_0$ is the ferromagnetic resonance frequency; H_0 is the effective magnetic field in magnetic particles and may be a sum of the external magnetic field, the effective anisotropy field, and the demagnetization field; $\omega_m = \gamma\vec{M}_0$, where γ is the gyromagnetic ratio and M_0 is the saturation magnetization of magnetic particles; α is the magnetic damping coefficient; and ω is the frequency of incident electromagnetic waves. We shall only consider incident electromagnetic waves propagating in the direction of the magnetization. This is the most interesting case in the study of magneto-optical effects in ferromagnetic materials. We also assume that the grain sizes are much smaller compared with the characteristic wavelength λ, and consequently, electromagnetic waves in the composite can be treated as propagating in a homogeneous magnetic system. According to Maxwell's equations, electromagnetic waves propagating in the direction of magnetization in a homogeneous magnetic material is either positive or negative transverse circularly polarized. If the composite can truly be treated as a homogeneous magnetic system in the case of grain sizes much smaller than the characteristic wavelength, electric and magnetic fields in the composite should also be either positive or negative circularly polarized and can be expressed as

$$\vec{E}(\vec{r},t) = \vec{E}_0^{(\pm)} e^{ikz-\beta z-i\omega t}, \tag{4}$$

$$\vec{H}(\vec{r},t) = \vec{H}_0^{(\pm)} e^{ikz-\beta z-i\omega t}, \tag{5}$$

where $\vec{E}_0^{(\pm)} = \hat{x} \mp i\hat{y}$, $\vec{H}_0^{(\pm)} = \hat{x} \mp i\hat{y}$, $k = \mathrm{Re}[k_{\mathrm{eff}}]$ is the effective wave number, $\beta = \mathrm{Im}[k_{\mathrm{eff}}]$ is the effective damping coefficient caused by the eddy current, and $k_{\mathrm{eff}} = k + i\beta$ is the effective propagation constant. In Eqs. (4) and (5) the signs of k and β can both be positive or negative depending on the directions of the wave vector and the energy flow. For convenience we assume that the direction of energy flow is in the positive direction of the z axis, i.e., we assume $\beta > 0$ in Eqs. (4) and (5), but the sign of k still can be positive or negative. In this case, if $k > 0$, the phase velocity and energy flow are in the same directions, and from Maxwell's equation, one can see that the electric and magnetic field $\vec{E}$ and $\vec{H}$ and the wave vector $\vec{k}$ will form a right-handed triplet of vectors. This is the usual case for right-handed materials. In contrast, if $k < 0$, the phase velocity and energy flow are in opposite directions, and $\vec{E}$, $\vec{H}$, and $\vec{k}$ will form a left-handed triplet of vectors. This is just the peculiar case for left-handed materials. So, for incident waves of a given frequency ω, we can determine whether wave propagations in the composite is right handed or left handed through the relative sign changes of k and β.

In the following, we shall determine the effective propagation constant $k_{\mathrm{eff}} = k + i\beta$ by means of the effective medium approximation. In the study of the propagation of electromagnetic waves through composite media, the effective medium theories constitute the most prevalent approach to the problem.[8–12] Basically, the effective medium theories are based on the self-consistent embedding approximations, and the accuracy of various types of effective medium theories have been discussed in a series of references.[8–12] In many cases, the effective medium theories can give a qualitatively correct description on the propagation properties of composite media, but in some cases, due to the limitations of their accuracy, effective medium theories will produce incorrect results.

As a first step, in this paper, we will assume that the effective medium approximations are valid in our problem; further investigations by more accurate approximation schemes will be carried out in the future to further confirm the results obtained by the effective medium approximation. Since the details of the effective medium approximation have been discussed in many references, here we only list the main points. First, if the composite can truly be considered as a homogeneous magnetic system in the case of grain sizes much smaller than the characteristic wavelength, then for waves (positive or negative circularly polarized) propagating through the composite in the direction of magnetization, their propagations can be described by an effective permittivity ϵ_{eff} and an effective permeability μ_{eff}, which satisfy the following relations assuming that the response functions are local for all components in the composite:

$$\int \vec{D}(\vec{r},\omega)e^{ik_{\mathrm{eff}}z}d\vec{r}=\epsilon_{\mathrm{eff}}\int \vec{E}(\vec{r},\omega)e^{ik_{\mathrm{eff}}z}d\vec{r}, \tag{6}$$

$$\int \vec{B}(\vec{r},\omega)e^{ik_{\mathrm{eff}}z}d\vec{r}=\mu_{\mathrm{eff}}\int \vec{H}(\vec{r},\omega)e^{ik_{\mathrm{eff}}z}d\vec{r}, \tag{7}$$

where k_{eff} and ω are related by $k_{\mathrm{eff}}=\omega[\epsilon_{\mathrm{eff}}\mu_{\mathrm{eff}}]^{1/2}$. Equations (6) and (7) are exact in principle assuming that nonlocal effects can be neglected. This assumption is appropriate in many cases. But in some cases, nonlocal effects can be significant and cannot be neglected, as has been shown in the past. In such cases, Eqs. (6) and (7) shall be not exact. For simplicity, in this paper we assume that nonlocal effects can be neglected and hence Eqs. (6) and (7) shall be valid. Although the relations in Eqs. (6) and (7) are simple, it is very difficult to calculate the integrals in them because the fields in the composite are spatially varying in a random way. One therefore must resort to various types of approximations. The simplest approximation is the effective medium approximation. In this approximation, we calculate the fields in each grain as if the grain were embedded in an effective medium of dielectric constant ϵ_{eff} and magnetic permeability μ_{eff}. Consider, for example, the ith grain. Under the embedding assumption, the electric and magnetic fields incident on the grain are the form of Eqs. (4) and (5):

$$\vec{E}_{\mathrm{inc}}=\vec{E}_0^{(\pm)}e^{ik_{\mathrm{eff}}z-i\omega t}, \tag{8}$$

$$\vec{H}_{\mathrm{inc}}=\vec{H}_0^{(\pm)}e^{ik_{\mathrm{eff}}z-i\omega t}, \tag{9}$$

where $\vec{E}_0^{(\pm)}=\hat{x}\mp i\hat{y}$ and $\vec{H}_0^{(\pm)}=\hat{x}\mp i\hat{y}$ correspond to the positive $(+)$ or negative $(-)$ circularly polarized waves. If the fields inside the grain can be found, then the inside fields can be used to calculate the integral over the grain volume:

$$\vec{I}_i=\int_{v_i}\vec{E}_i(\vec{r},\omega)e^{ik_{\mathrm{eff}}z}d\vec{r}, \tag{10}$$

$$\vec{J}_i=\int_{v_i}\vec{h}_i(\vec{r},\omega)e^{ik_{\mathrm{eff}}z}d\vec{r}, \tag{11}$$

which is required to find the integral in Eqs. (6) and (7). For the positive or negative circularly polarized incident waves described by Eqs. (8) and (9), the integral $\vec{I}_i$ and $\vec{J}_i$ can be written as

$$\vec{I}_i=(\hat{x}\mp i\hat{y})I_i, \tag{12}$$

$$\vec{J}_i=(\hat{x}\mp i\hat{y})J_i, \tag{13}$$

where I_i and J_i are scalars. If I_i and J_i can be found, then from Eqs. (6) and (7), the effective permittivity ϵ_{eff} and effective permeability μ_{eff} can be calculated by

$$\epsilon_{\mathrm{eff}}=\frac{f_1\epsilon_1 I_1+f_2\epsilon_2 I_2}{f_1 I_1+f_2 I_2}, \tag{14}$$

$$\mu_{\mathrm{eff}}=\frac{f_1\mu_1 J_1+f_2\mu_2^{(\pm)}J_2}{f_1 J_1+f_2 J_2}, \tag{15}$$

where f_1 and f_2 are the volume fractions of the two types of grains, μ_1 is the permeability of nonmagnetic dielectric grains, and $\mu_2^{(+)}=\mu_a-\mu'$ and $\mu^{(-)}=\mu_a+\mu'$ [see Eqs. (1)–(3)] are the effective permeability of magnetic grains for positive and negative circularly polarized waves, respectively. As for the calculation of I_i and J_i, we can follow the method of expanding interior and exterior fields in a multipole series and matching the boundary conditions.[13] After the coefficients of the multipole expansion of interior and exterior fields are obtained by matching the boundary conditions, I_i and J_i can be found and subsequently be substituted into Eqs. (14) and (15). Since this method is standard, we shall not present the details. In the final results, Eqs. (14) and (15) reduce to single equation:

$$\sum_{i=1,2} f_i\sum_{l=1}^{\infty}(2l+1)\times\left[\frac{k_{\mathrm{eff}}\psi_l'(k_iR_i)\psi_l(k_{\mathrm{eff}}R_i)-k_i\psi_l(k_iR_i)\psi_l'(k_{\mathrm{eff}}R_i)}{k_{\mathrm{eff}}\psi_l'(k_iR_i)\zeta_l(k_{\mathrm{eff}}R_i)-k_i\psi_l(k_iR_i)\zeta_l'(k_{\mathrm{eff}}R_i)}+\frac{k_i\psi_l'(k_iR_i)\psi_l(k_{\mathrm{eff}}R_i)-k_{eff}\psi_l(k_iR_i)\psi_l'(k_{\mathrm{eff}}R_i)}{k_i\psi_l'(k_iR_i)\zeta_l(k_{\mathrm{eff}}R_i)-k_{\mathrm{eff}}\psi_l(k_iR_i)\zeta_l'(k_{\mathrm{eff}}R_i)}\right]=0, \tag{16}$$

where R_i is the radius of the ith type of grains, and

$$k_1=\omega[\epsilon_1\mu_1]^{1/2}, \tag{17}$$

$$k_2=\omega[\epsilon_2\mu_2^{(\pm)}]^{1/2}, \tag{18}$$

$$\psi_l(x)=xj_l(x), \tag{19}$$

$$\zeta_l(x)=xh_l^{(1)}(x), \tag{20}$$

where $j_l(x)$ and $h_l(x)$ are the usual spherical Bessel and Hankel functions. Equation (16) determines the effective

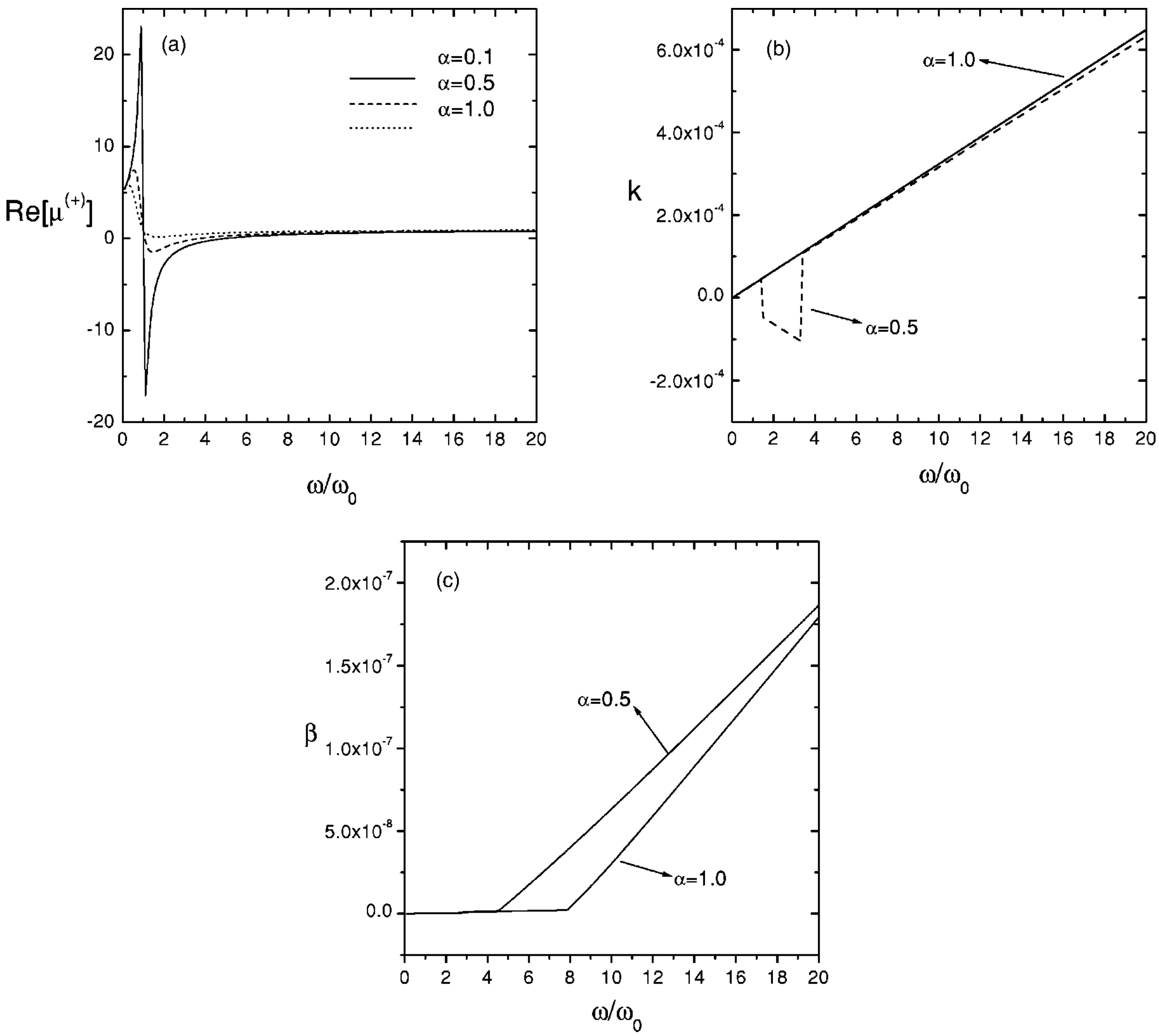

FIG. 1. (a) The frequency dependence of the real part of the effective permeability $\mu^{(+)}$ of magnetic grains and the corresponding frequency dependencies of (b) the effective wave number k and (c) the effective damping coefficient β of the composite for positive circularly polarized waves propagated in the direction of magnetization. (In all figures we use the units such that $\omega_p=1$ and $c=1$. For ferromagnetic metal grains such as Ni and Fe, ω_p is usually on the magnitude of 10^2–10^5 cm^{-1}, and the magnetic resonance frequency ω_0 is usually on the magnitude of 10^{-3}–$10^{-5}\omega_p$. For simplicity, hereafter we will set $\omega_0/\omega_p=10^{-5}$. The other parameters are $\omega_p\tau=100$, $\omega_m/\omega_0=4.0$, $\omega_pR_1/c=\omega_pR_2/c=0.2$, $f_2=0.3$; α is shown in the figures.)

product of $(\epsilon\mu)_{eff}$, or equivalently k_{eff}, but not a single ϵ_{eff} and μ_{eff}. It can describe the change of the phase of a plane wave across a slab of the composite, but it does not precisely describe wave propagations across a slab of the composite. This is due to the fact we make no attempt to rigorously solve the boundary-value problem for a slab of composite by matching the fields inside the slab and external fields outside the slab at the boundary. In fact, it is common in various types of effective medium theories that for $\omega\neq0$ the electromagnetic properties of a composite cannot in general be specified by a single ϵ_{eff} and μ_{eff}.

Since we can determine whether wave propagations through the composite is left handed or right handed by the calculation of the effective propagation constant k_{eff}, Eq. (16) is enough for the problems we are discussing. But it should be pointed out that Eq. (16) was derived by assuming no interaction between different particles. If the interaction between particles is strong, for example, if they are close together to each other, the effects of higher-order scattering will become important and Eq. (16) may break down. In this paper we are only interested in the case where the interaction between different particles is weak and hence Eq. (16) can be applied.

We have done the numerical calculation for a metal volume fraction f_2 of 0.3 based on Eq. (16). Since the main purpose of our model calculation is to illustrate the idea introduced in the beginning of this paper, and there have never been such left-handed composite media actually prepared in experiments, in addition, in real composite media the material parameters for each component in the composite are usually strongly dependent on many factors such as the method of preparing the sample, the particle sizes in the composite, and the external field, etc., so the choice of model parameters in our calculation will be somewhat arbitrary and not related

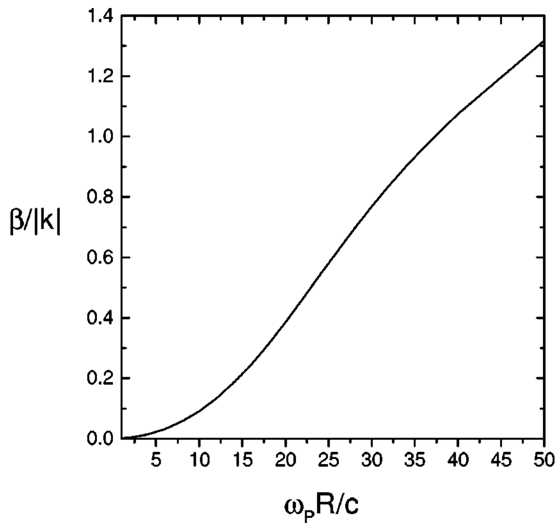

FIG. 2. The dependence of the damping coefficient $\beta/|k|$ of the composite on the grain sizes of the metallic component. (The parameters are $\omega_m/\omega_0=4.0$, $\omega_p\tau=100$, $\omega/\omega_0=1.2$, $\alpha=0.1$, $f=0.3$.)

to any real composite materials. From an experimental point of view, there are a few ferromagnetic metals such as Ni and Fe or their metallic magnetic compounds that are both metallic and ferromagnetic and have large negative permeability at frequency in the vicinity of the ferromagnetic resonance frequency, and may possibly be the candidates for the metallic components in preparing such left-handed composite media, and our calculations may be approximately valid for such composite materials. The numerical results obtained from Eq. (16) are summarized in Figs. 1–3. Figure 1(a) shows the frequency dependence of the real part of the effective permeability $\mu^{(+)}$ of magnetic grains for positive circularly polarized plane waves, Figures 1(b) and 1(c) show the corresponding frequency dependences of the effective wave number k and the effective damping coefficient β in a composite consisting of metallic magnetic grains and dielectric grains.

From Eqs. (1)–(3), we can see that if the magnetic damping coefficient α is zero, Re$[\mu^{(+)}]$ will be negative in the whole frequency region of $\omega>\omega_0$ (the magnetic resonance frequency). From Fig. 1(a), we can see that if α is nonzero but small enough, there can still be a frequency region near ω_0 in which Re$[\mu^{(+)}]$ is negative. In this case, if the amplitude of the negative Re$[\mu^{(+)}]$ is large enough, k will be negative in this frequency region as was shown in Fig. 1(b), and hence the phase velocity and energy flow will be in opposite directions in this frequency region, and $\vec{E}$, $\vec{H}$, and $\vec{k}$ will form a left-handed triplet of vectors, i.e., the composite will be left-handed in this frequency region for positive circularly polarized plane waves. But if α is not small enough, Re$[\mu^{(+)}]$ will be positive in the whole frequency region, or though Re$[\mu^{(+)}]$ is negative in a frequency region near ω_0, the amplitude of the negative Re$[\mu^{(+)}]$ is not large enough. In this case k will be positive in the whole frequency region, as was shown in Fig. 1(b). Here the composite is right-handed for positive circularly polarized waves in the whole frequency region.

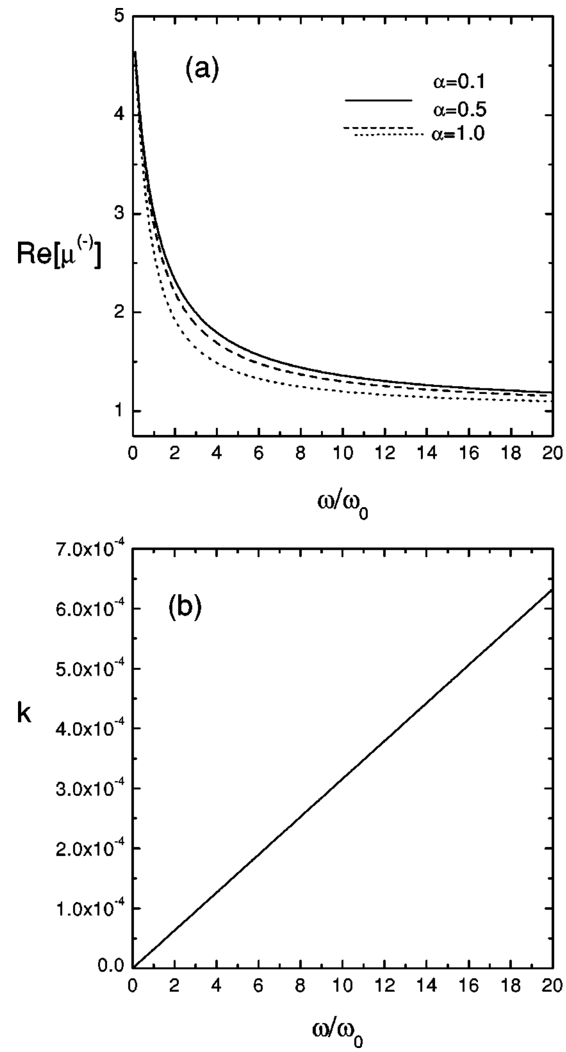

FIG. 3. (a) The frequency dependence of the real part of the effective permeability $\mu^{(-)}$ of magnetic grains and (b) the corresponding frequency dependencies of the effective wave number k of the composite for negative circularly polarized waves propagated in the direction of magnetization. (The parameters are $\omega_m/\omega_0=4.0$, $\omega_p\tau=100$, $\omega_pR_1/c=\omega_pR_2/c=0.2$, $f_2=0.3$, α is shown in the figures.)

The calculations also show that if the radius of metallic grains are small enough and the volume fraction of metal components is smaller than the threshold value of the insulator-metal transition, which is approximately 1/3 in our model, the losses caused by eddy current are very small and the composite is essentially an insulator. This can be seen from Fig. 1(c), in which the damping coefficient β is very small compared with the amplitude of the wave number k, i.e., the eddy current losses are very small in the cases shown in Fig. 1. If the volume fraction of metal components is larger than the threshold value, the composite will be essentially a metal, and the damping coefficient β will be much larger than the amplitude of wave number k (not shown in the figure). The dependence of the damping coefficient on the size of the metal grains is shown in Fig. 2. We can see that with the increase of the grain sizes of the metal component, the damping coefficient will increase substantially and will become very large even if the volume fraction of the metal component is below the threshold value of the metal-insulator transition.

Compared with the cases for the positive circularly polarized waves, in Fig. 3(a) we show the frequency dependence of the real part of the effective permeability $\mu^{(-)}$ of magnetic grains for negative circularly polarized waves and in Fig. 3(b) we show the corresponding frequency dependence of the effective wave number k in the composite consisting of the metallic magnetic grains and dielectric grains. We can see that for negative circularly polarized waves, $\mathrm{Re}[\mu^{(-)}]$ is positive in the whole frequency region no matter how small α is, and correspondingly, k is positive in the whole frequency region, i.e., the composite is right-handed in the whole frequency region for negative circularly polarized waves no matter how small α is.

In conclusion, we have investigated theoretically the possibility of preparing a left-handed material in metallic magnetic granular composites based on a simple model analysis. The numerical results show that, by incorporating metallic magnetic nanoparticles into an appropriated insulating matrix and controlling the directions of magnetization of metallic magnetic components and their volume fraction and the particle sizes, it may be possible to prepare a composite medium of low eddy current losses which is left-handed for electromagnetic waves propagating in some special direction and polarization in a frequency region near the magnetic resonance frequency. These interesting results are obtained based on the effective medium approximation, but as has been shown in the past, the accuracy of various types of effective medium approximations are limited by many factors, and different effective medium theories can produce significantly different predictions,[8–12] so further theoretical investigations by other approximation schemes such as first-principle numerical calculations may be needed in order to further confirm the interesting possibility shown in this paper.

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