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

S.T.Chui and Liangbin Hu

Bartol Research Institute, University of Delaware, Newark, Delaware, U.S.A

Abstract

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 which 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.

PACS numbers: 73.20.Mf, 41.20.Jb, 42.70.Qs
In classical electrodynamics, the response (typically frequency-dependent) of a material to electric and magnetic fields is characterized by two fundamental quantities, the permittivity $\epsilon$ and the permeability $\mu$. The permittivity relates the electric displacement field $\vec{D}$ to the electric field $\vec{E}$ through $\vec{D} = \epsilon \vec{E}$, and the permeability $\mu$ 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 $\epsilon$ and $\mu$ 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 $\epsilon$ and $\mu$ and cause losses, but for a qualitative picture, one can ignore losses and treat $\epsilon$ and $\mu$ as real numbers. Also, strictly speaking, $\epsilon$ and $\mu$ are second-rank tensors, but they reduce to scalars for isotropic materials). In a medium with $\epsilon$ and $\mu$ 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 $\epsilon$ and $\mu$ 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 10MHz in the ionosphere and falls at or above visible frequencies for most metals. At frequencies above the plasma frequency, $\epsilon$ is positive and electromagnetic waves are transmitted. For lower frequencies, $\epsilon$ 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 $\epsilon$ and positive $\mu$, from the theoretical point of view, in a medium with $\epsilon$ and $\mu$ 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 thirty years ago—when no material with simultaneously negative $\epsilon$ and $\mu$ 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 $\epsilon$ and $\mu$ 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 $\epsilon$ and $\mu$, including reversal of both the Doppler shift and the Cerenkov 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[1], Smith and coworkers 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 $\epsilon$ and negative permeability $\mu$ 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 rapid communication, we propose to investigate the possibility of preparing left-handed materials in another type of systems-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 achieve
a composite medium that has simultaneously negative $\epsilon$ and negative $\mu$ 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 $\omega_0$, 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 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 results of 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 particles are metallic ferromagnetic grains of radius $R_1$, the other type are non-magnetic dielectric (insulating) grains 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 non-magnetic dielectric grains are both scalars, and will be denoted as $\epsilon_1$ and $\mu_1$. The permittivity of metallic magnetic grains will be denoted as $\epsilon_2$ and will be taken to have a Drude form: $\epsilon_2 = 1 - \omega_p^2/\omega(\omega + i/\tau)$, where $\omega_p$ is the plasma frequency of the metal and $\tau$ is a relaxation time. Such a form of $\epsilon$ is representative of a variety of metal composites\cite{8}-\cite{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\cite{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]:

\[
\hat{\mu}_2 = \begin{bmatrix}
\mu_a & -i\mu' & 0 \\
 i\mu' & \mu_a & 0 \\
 0 & 0 & 1
\end{bmatrix}
\]  

(1)

where

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

(2)

\[
\mu' = \frac{\omega_m \omega}{\omega_0 + i\alpha\omega} - \omega^2,
\]

(3)

\( \omega_0 = \gamma H_0 \) 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; \( \omega_m = \gamma M_0 \), where \( \gamma \) is the gyromagnetic ratio, \( M_0 \) is the saturation magnetization of magnetic particles; \( \alpha \) is the magnetic damping coefficient; \( \omega \) 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 \( \lambda \), 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:

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

(4)

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

(5)

where \( \vec{E}_0^{(\pm)} = \hat{x} \mp i\hat{y} \), \( \vec{H}_0^{(\pm)} = \hat{x} \mp i\hat{y} \), \( k = \text{Real}[k_{eff}] \) is the effective wave number, \( \beta = \text{Im}[k_{eff}] \) is the effective damping coefficient caused by the eddy current, \( k_{eff} = \)
$k + i\beta$ is the effective propagation constant. In Eqs.(4)-(5) the signs of $k$ and $\beta$ 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)-(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 $\omega$, we can determine whether wave propagations in the composite is right-handed or left-handed through the relative sign changes of $k$ and $\beta$. In the following, we shall determine the effective propagation constant $k_{\text{eff}} = k + i\beta$ by means of the effective medium approximation. The details of various kinds of effective medium approximations have been discussed in a series of references [8]-[12], 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 $\epsilon_{\text{eff}}$ and an effective permeability $\mu_{\text{eff}}$, which satisfy the following relations

\[ \int \vec{D}(r,\omega) e^{ik_{\text{eff}}z} d\vec{r} = \epsilon_{\text{eff}} \int \vec{E}(r,\omega) e^{ik_{\text{eff}}z} d\vec{r}, \]  

\[ \int \vec{B}(r,\omega) e^{ik_{\text{eff}}z} d\vec{r} = \mu_{\text{eff}} \int \vec{H}(r,\omega) e^{ik_{\text{eff}}z} d\vec{r}, \]  

where $k_{\text{eff}}$ and $\omega$ are related by $k_{\text{eff}} = \omega [\epsilon_{\text{eff}} \mu_{\text{eff}}]^{1/2}$. Although these relations are simple and in principle exact, 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 \( \varepsilon_{\text{eff}} \) and magnetic permeability \( \mu_{\text{eff}} \). Consider, for example, the \( i \)th grain. Under the embedding assumption, the electric and magnetic fields incident on the grain are the form of Eqs.(4)-(5):

\[
\vec{E}_{\text{inc}} = \vec{E}_0^{(\pm)} e^{ik_{\text{eff}}z-i\omega t},
\]

\[
\vec{h}_{\text{inc}} = \vec{h}_0^{(\pm)} e^{ik_{\text{eff}}z-i\omega t},
\]

where \( \vec{E}_0^{(\pm)} = \hat{x} \mp i\hat{y} \) and \( \vec{h}_0^{(\pm)} = \hat{x} \mp i\hat{y} \), corresponding 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

\[
\vec{I}_i = \int_{v_i} \vec{E}_i(\vec{r},\omega)e^{ik_{\text{eff}}z}d\vec{r},
\]

\[
\vec{J}_i = \int_{v_i} \vec{h}_i(\vec{r},\omega)e^{ik_{\text{eff}}z}d\vec{r},
\]

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

\[
\vec{I}_i = (\hat{x} \mp i\hat{y})I_i,
\]

\[
\vec{J}_i = (\hat{x} \mp i\hat{y})J_i,
\]

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

\[
\varepsilon_{\text{eff}} = \frac{f_1\varepsilon_1 I_1 + f_2\varepsilon_2 I_2}{f_1 I_1 + f_2 I_2},
\]

\[
\mu_{\text{eff}} = \frac{f_1\mu_1 J_1 + f_2\mu_2^{(\pm)} J_2}{f_1 J_1 + f_2 J_2},
\]

where \( f_1 \) and \( f_2 \) are the volume fractions of the two types of grains, \( \mu_1 \) is the permeability of non-magnetic dielectric grains, \( \mu_2^{(+)} = \mu_a - \mu' \) and \( \mu_2^{(-)} = \mu_a + \mu' \) (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)-(15). Since this method is standard, we shall not present the details. In the final results, Eqs.(14)-(15) reduce to one self-consistent equation:

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

where \( R_i \) is the radius of the \( i \)th type of grains, and

\[
k_1 = \omega [\varepsilon_1 \mu_1]^{1/2},
\]

\[
k_2 = \omega [\varepsilon_2 \mu_2^{(\pm)}]^{1/2},
\]

\[
\psi_l(x) = x j_l(x),
\]

\[
\zeta_l(x) = x h_l^{(1)}(x),
\]

\( j_l(x) \) and \( h_l(x) \) are the usual spherical Bessel and Hankel functions. Eq.(16) determine the effective product of \((\varepsilon \mu)_{\text{eff}}\), or equivalently \( k_{\text{eff}} \), but not a single \( \varepsilon_{\text{eff}} \) and \( \mu_{\text{eff}} \). 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 \( \omega \neq 0 \) the electromagnetic properties of a composite cannot in general be specified by a single \( \varepsilon_{\text{eff}} \) and \( \mu_{\text{eff}} \). Since we can determine whether wave propagations through the composite is left-handed or right-handed by the calculation of the effective propagation constant \( k_{\text{eff}} \), Eq.(16) is enough for the problems we are discussing.

The numerical results for a metal volume fraction \( f_2 \) of 0.3 obtained from Eq.(16) are summarized in Fig.1-Fig.2. Fig.1(a) shows the frequency dependence of the real part of
the effective permeability $\mu^{(+)}$ of magnetic grains for positive circularly polarized plane waves. Fig.1(b) and (c) show the corresponding frequency dependences of the effective wave number $k$ and the effective damping coefficient $\beta$ in a composite consisting of metallic magnetic grains and dielectric grains. From Eqs.(1)-(3), we can see that if the magnetic damping coefficient $\alpha$ is zero, $\text{Re}[\mu^{(+)}]$ 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 $\alpha$ is nonzero but small enough, there can still be a frequency region near $\omega_0$ in which $\text{Re}[\mu^{(+)}]$ is negative. In this case, if the amplitude of the negative $\mu^{(+)}$ 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 the 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 $\alpha$ is not small enough, $\text{Re}[\mu^{(+)}]$ will be positive in the whole frequency region, or though $\text{Re}[\mu^{(+)}]$ is negative in a frequency region near $\omega_0$, the amplitude of the negative $\text{Re}[\mu^{(+)}]$ is not large enough, in this case $k$ will be positive in the whole frequency region, as was shown in Fig.1(b). In this case, the composite is right-handed for positive circularly polarized waves in the whole frequency region. 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 $\beta$ 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 $\beta$ will be much larger than the amplitude of wave number $k$ (not shown in the figure). In Fig.2(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.2(b) we show the corresponding frequency dependence of the effective
wave number $k$ in a composite consisting of the metallic magnetic grains and dielectric grains. We can see that for negative circularly polarized waves, $\text{Re}[\mu^(-)]$ is positive in the whole frequency region no matter how small $\alpha$ 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 $\alpha$ is.

In conclusion, we have discussed the possibility of preparing a left-handed material in metallic magnetic granular composites based on the effective medium approximation. Our model analysis shows 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, 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. Further theoretical investigations by other approximations such as first-principle numerical calculations may be needed to further confirm the possibility shown in this paper.

S. T. Chui was supported in part by the Office of Naval Research and by the Army Research Laboratory through the Center of Composite Materials at the University of Delaware. We thank John Xiao for helpful discussions.

References

[1] J.B.Pendry, A.J.Holden, W.J.Stewart, and I.Youngs, *Phys. Rev. Lett* 76, 4773 (1996)

[2] R.H.Ritchie and A.Howie, *Phil. Mag. A* 44, 931 (1981)

[3] T.L.Ferrell and P.M.Echenique, *Phys. Rev. Lett* 55, 1526 (1985)

[4] A.W.Howie and C.A.walsh, *Microsc.Microanal.Microstructure.* 2, 171 (1991)

[5] V.G.Veselago, *Sov.Phys.Usp.* 10, 509 (1968)
[6] D.R. Smith, W.J. Padilla, D.C. Vier, S.C. Nemet-Nasser, S. Schultz, *Phys. Rev. Lett.* 67, 3578 (2000)

[7] C.P. Slichter, *Principle of Magnetic Resonance*, Springer-Verlag Berlin, Heidelberg (1978)

[8] R. Burridge, S. Childress, and G. Papanicolaou (edition), *Macroscopic Properties of Disordered Media*, Springer-Verlag Berlin, Heidelberg (1982)

[9] J.C. Garland and D.B. Tanner (edition), *Electrical Transport and Optical Properties of Inhomogeneous Media*, American Institute of Physics, New York (1978)

[10] D. Stroud and F.P. Pan, *Phys. Rev. B* 20, 455 (1979)

[11] P. Sheng, *Phys. Rev. Lett* 45, 60 (1980)

[12] W. Lamb, D.M. Wood, and N.W. Ashcroft, *Phys. Rev. B* 21, 2248 (1980)

[13] H.C. Van de Hulst, *Light scattering by Small particles* (Chapter 9), Dover Publications Inc., New York (1981); J.D. Jackson, *Classical electrodynamics*, 2nd ed., Wiley, New York (1975)
Fig. 1. (a) The frequency dependencies 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 $\beta$ of the composite for positive circularly polarized waves propagating in the direction of magnetization. (The plasma frequency $\omega_p$ is usually in the visible or ultraviolet frequency region and the ferromagnetic resonance frequency $\omega_0$ is usually in the microwave frequency region. For simplicity, hereafter we will set $\omega_0/\omega_p = 10^{-5}$. The other parameters are: $\omega_m/\omega_0 = 4.0$, $\omega_pR/c = 0.2$, $f_2 = 0.3$, $\alpha$ is shown in the figures).

Fig. 2. (a) The frequency dependence 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 $\beta$ of the composite for negative circularly polarized waves propagating in the direction of magnetization. (The parameters are: $\omega_m/\omega_0 = 4.0$, $\omega_pR/c = 0.2$, $f_2 = 0.3$, $\alpha$ is shown in the figures.)
Fig. 1(a)
Fig. 1(c)
Fig. 2(a)
Fig. 2(b)