Negative permeability of magnetic nanocomposite films for designing left-handed metamaterials

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Negative permeability of magnetic nanocomposite films for designing left-handed metamaterials

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We have theoretically evaluated magnetic permeability of Ni nanocomposite films, in which the dipolar interactions between the Ni particles are considered explicitly. A numerical model of the nanocomposite is constructed according to our recent experimental results and a micromagnetic calculation is conducted. We show that the value of permeability strongly depends on the volume fraction and randomness of the particles. Particularly, as the randomness of the particles increases, the negative value of permeability drastically shrinks. However, the region of applied field, where the permeability is negative, becomes broader. This is advantageous for the broadband left-handed metamaterials in microwave regions. © *2007 American Institute of Physics*.
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Metamaterials consisting of artificial units much smaller than the wavelength of electromagnetic (EM) waves are a rich and rapidly growing area in optics and photonics.[1–3] In particular, left-handed metamaterials (LHMs), which mimic negative electric permittivity (ϵ) and magnetic permeability (μ) simultaneously, are of great interest because of the intriguing EM responses, e.g., negative refraction and inverse Doppler shift.[4] The LHMs or negative index of refraction metamaterials have potential applications for subwavelength imaging[5] and compact optical components.[6] In order to realize negative μ, magnetic resonance is required. Structural magnetic resonance using Cu split-ring resonators is a well known method of obtaining negative μ in microwave regions.[7,8] In the present study, however, an alternative route using intrinsic magnetic resonance of magnetic metals, e.g., Fe, Co, and Ni, is used. This route was first proposed by Chui and Hu.[9]

It is known that negative μ is realized using electron magnetic resonance (EMR) of electron spins in magnetic metals.[10] In bulk metal, however, the eddy current loss is dominant; the effect of the negative value of the real part of μ (μ') cannot be observed due to the imaginary part (μ''). Since the eddy current loss is inversely scaled with the volume of magnetic metals,[10] miniaturization is a possible address to suppress the eddy current losses.[9] Therefore, nanocomposites consisting of magnetic metal nanoparticles embedded in nonmagnetic insulating matrices have been considered theoretically to realize LHMs in the microwave region. Nevertheless, detailed criteria for obtaining negative μ are still unclear.

Recently, we have prepared Ni nanocomposite films consisting of 8 nm diameter Ni particles with various volume fractions.[11–14] In this letter, we report a numerical simulation of μ' of the Ni nanocomposite films, in which the dipolar interactions between the Ni particles are considered explicitly. A numerical model of the nanocomposites is constructed according to our recent experimental results. Micromagnetic calculations are carried out and the μ' is evaluated. In particular, the influence of the magnetic nanoparticle volume fraction and the particle randomness is studied. The material design for the negative μ' and LHMs using the magnetic nanoparticle systems is discussed.

Let us suppose a Ni nanoparticle having a total magnetic moment $\boldsymbol{m}$, as illustrated in Fig. 1(a). The magnetization $\boldsymbol{m}_i$ is given by $\boldsymbol{m}_i = \mu_s 2\pi d_i^3 / 3a^3$ (emu), where d_i is the diameter of ith Ni particle, a is the lattice constant of bulk Ni, and μ_s is the magnetic moment of a Ni atom. When $d_i = 8$ nm, $\boldsymbol{m}_i$ is 1.3×10^{-16} emu. Under an external magnetic field $\boldsymbol{H}_{\text{ext}}$, the moment $\boldsymbol{m}_i$ precesses at an angular frequency ω_L (the Larmor frequency).[15]

To represent the nanocomposite films, we consider a periodic regular lattice of magnetization, as shown in Fig. 1(b). Ni particles are arranged in the $9 \times 9 \times 5$ lattice with the lattice spacing r_0.[16] This numerical model is treated by solving the motion equation of magnetization. The motion equation of magnetization in the ith Ni particle can be written as

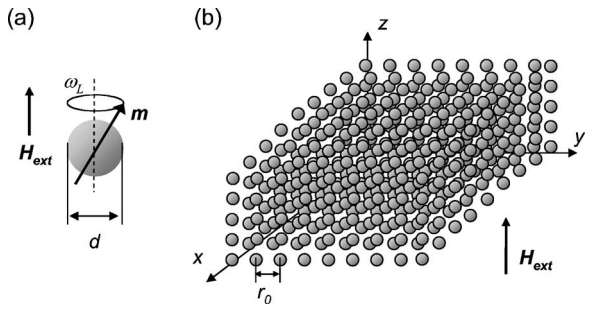

FIG. 1. Schematic illustrations of (a) a magnetically isolated spherical nanoparticle and (b) a thee-dimensional array of interacting nanoparticles.

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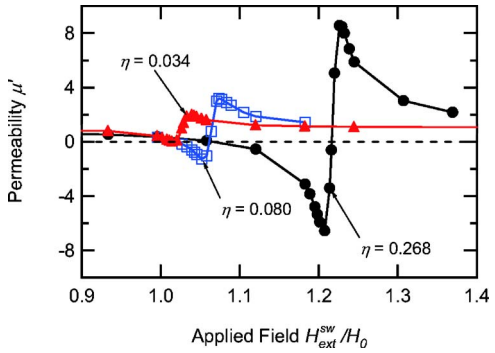

FIG. 2. (Color online) The permeability μ' of composite films with various volume fractions η of Ni nanoparticles. Filled circles correspond to η=0.268. Open squares correspond to η=0.080. Filled triangles correspond to η=0.034.

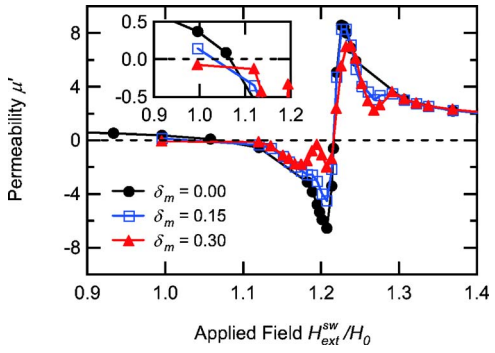

FIG. 3. (Color online) The permeability μ' of composite films consisting of 8 nm diameter Ni particles with the several distribution of particle diameter. The η is 0.268. The deviation of particle diameter is expressed as δ_m. Filled circles are assigned to δ_m=0.00. Open squares are assigned to δ_m=0.15. Filled triangles are assigned to δ_m=0.30. An inset exhibits magnification of the region where the permeability μ' changes the sign.

$$\frac{d\boldsymbol{m}_i}{dt} = -\gamma(\boldsymbol{m}_i \times \boldsymbol{H}_{\text{int}}^i) - \frac{\alpha\gamma}{\boldsymbol{m}_i}\left\{\boldsymbol{m}_i \times \left(\boldsymbol{m}_i \times \frac{d\boldsymbol{m}_i}{dt}\right)\right\}, \qquad (1)$$

where α is the Gilbert damping factor, which is phenomenologically introduced in the motion equation. α is fixed at 0.01 in the present calculations. The magnetic field $\boldsymbol{H}_{\text{int}}^i$ includes an applied external field $\boldsymbol{H}_{\text{ext}}$ and a dipole field $\boldsymbol{H}_{\text{dip}}^i$, which is given by the surrounding particles. In the simulation, $\boldsymbol{H}_{\text{ext}}$ is defined by $\boldsymbol{H}_{\text{ext}}=\boldsymbol{H}_{\text{ext}}^{\text{sw}}+\boldsymbol{H}_p \sin(2\pi\nu t)$. $\boldsymbol{H}_{\text{ext}}^{\text{sw}}$ is a dc sweeping magnetic field. $\boldsymbol{H}_p \sin(2\pi\nu t)$ is an ac irradiation field of microwaves for EMR. In the sine function, t is the time and ν is the frequency. $\boldsymbol{H}_{\text{ext}}^{\text{sw}}$ and $\boldsymbol{H}_p$ satisfy $\boldsymbol{H}_{\text{ext}}^{\text{sw}} \cdot \boldsymbol{H}_p = 0$. In the x and y directions in Fig. 1(b), the mirror boundary condition is applied to take account for the symmetry of the dipole field. On the other hand, the free boundary condition is provided in the z direction to consider the effect of the finite thickness of the composite film.

The numerical calculation of Eq. (1) is carried out by using the finite difference method with a forward Euler difference scheme.[17] The discretization scheme of Eq. (1) follows to a literature.[18] The forward Euler difference includes a problem of the instability of convergence. In order to stabilize the calculation, the time step Δt in a difference equation is defined by $\Delta t\nu = 9\times10^{-4}$. The dynamics of $\boldsymbol{m}_i$ as a function of time and the magnetic susceptibility of the particles are obtained from the calculation.

The nanocomposite films consist of Ni particles and nonmagnetic matrices. The real part of susceptibility χ' of the nanocomposite film in the equilibrium state of precession is thus averaged over total volume of the composite. In this work, the averaged χ' is simply defined using the volume fraction of Ni (η) and the saturation magnetization m_s of Ni in the composite films. The μ' of nanocomposite films is thus given by

$$\mu' = 1 + \chi' = 1 + \left\langle \eta m_s \frac{dm_{H_p}}{dH_p} \right\rangle, \qquad (2)$$

where m_{H_p} is a projection of a magnetization vector in the H_p direction in the equilibrium state of precession defined as $m_{H_p}=(\boldsymbol{m}\cdot\boldsymbol{H}_p)/(m_s H_p)$. The magnetization m_s in Eq. (2) is set to 6900 G to simulate Ni nanoparticles.

The μ' of nanocomposite films as a function of $H_{\text{ext}}^{\text{sw}}/H_0$ is calculated for various volume fractions η, as shown in Fig. 2. $\boldsymbol{H}_{\text{ext}}^{\text{sw}}$ is applied along the z axis. H_0 is a resonance field for the isolated single particle. The particle diameter is set to 8 nm.[11] The η=0.268, 0.080, and 0.034 correspond to r_0=10, 15, and 20 nm, respectively. We see that the apparent resonance field H_0' for composites shifts to a higher field with increasing η; sample with η=0.034, 0.080, and 0.268 shows $H_0'=1.029H_0$, $1.065H_0$, and $1.216H_0$, respectively. H_0' is given by $H_0'=H_0+H_{\text{dip}}$, where the H_{dip} is a dipole field generated from surrounding particles.[16] The shift is thus caused by an enhancement of dipole field H_{dip} with increasing η.

As the dc sweeping field $H_{\text{ext}}^{\text{sw}}$ increases, the μ' negatively increases and exhibits a negative peak. Then it positively increases and exhibits a positive peak. The μ' reaches a value of 1 at H_0'. The negative μ' can be obtained when χ' satisfies $\chi'<-1$. For η=0.268, the sample shows a negative μ'; the minimum value of permeability (μ_{min}) is −6.56. As the η decreases to η=0.080, μ_{min} decreases to −1.27. Moreover, μ_{min} decreases to 0.03 when η=0.034; the μ' of nanocomposite film is no longer negative. The reduction of μ_{min} is simply caused by a decrease in the magnetization in the composite films because the magnetization can be expressed as ηm_s. A relation between η and μ_{min} will be discussed later in detail.

In addition, as the η decreases, the region of applied field showing a negative μ' (ΔH_{neg}) becomes narrower. The ΔH_{neg} are $0.15H_0$ for η=0.268 and $0.04H_0$ for η=0.080. The bandwidth ΔH_{neg} finally disappears at η=0.034.

We see in Fig. 2 that μ_{min} depends on the η. We consider μ_{min} as a function of η. The curve fitting provides a linear function $\mu_{\text{min}}=k\eta+\mu_0'$, where k=−28.15 and μ_0'=0.9887. In the limit of η=0, μ_0' has to be equal to the permeability of vacuum μ_0=1. In this sense, the obtained function is accurate because of $\mu_0' \simeq \mu_0$. The parameter k is dominated by the effective damping factor and magnetization. This result indicates that, for nanocomposites containing Ni particles with d=8 nm, the negative μ' is achieved when the volume fraction of Ni particles satisfies $\eta>0.035$. Moreover, $\mu'=-1$ for perfect lens conditions[5] requires the $\eta>0.071$.

The negative μ' has been obtained in the case of an ideal model expressed in Fig. 1 where magnetic nanoparticle diameter is constant. Next, we consider the influence of the randomness of particle diameter. μ' of composites with various dispersions of particle diameter is calculated, as shown in Fig. 3. The volume fraction is fixed at η=0.268. The dispersion of particle diameter is defined by the deviation δ_m. The particle diameter d_i randomly distributes within a range

of $(1\pm\delta_m)^{1/3}d$, where d is the average diameter (d=8 nm in the present study). The deviation of magnetic dipole interaction affects the permeability through the change of magnetization $\boldsymbol{m}_i$. In the model calculation, the deviation δ_m is set to δ_m=0.00, 0.15, and 0.30. In Fig. 3, filled circles correspond to δ_m=0.00, open squares to δ_m=0.15, and filled triangles to δ_m=0.30.

Figure 3 shows that positive peaks of the permeability exhibit 7.02–8.56 even though the particle diameter is dispersive. Contrastingly, we notice here that the change of negative peak permeability $\mu_{\min}$ is more significant than that of the positive peak. The $\mu_{\min}$ shows −6.56, −4.51, and −2.00 for δ_m=0.00, 0.15, and 0.30, respectively. Therefore, the dispersion of particle diameter brings about the significant reduction of $\mu_{\min}$ in the composite films. Although the negative peak of permeability is smaller, ΔH_{neg} becomes broader when the deviation of particle diameter is increased (an inset in Fig. 3). For a composite with δ_m=0.00, ΔH_{neg} is $0.15H_0$. When δ_m increases to 0.15, ΔH_{neg} increases to $0.18H_0$. Finally if δ_m=0.30, ΔH_{neg} becomes $0.38H_0$. In a case using the magnetic resonance, a large μ'' at the resonant state is a concern for practical applications. An off-resonant state is thus useful to avoid the absorption and loss of microwaves. The broad ΔH_{neg} with dispersive sized nanoparticles is suitable to stabilize the negative μ' at the region of off-resonance.

We should mention here that the position of nanoparticles in nanocomposite films, experimentally prepared, deviates from the regular lattice, of Fig. 1. The deviation of interparticle spacing in low η samples does not strongly affect the resonance condition.[11] In case of a high η, however, the influence of disorder in the arrangement of the particles may become significant.[19] The effect of the position disorder for the negative μ' will be discussed in a separate paper.

In summary, numerical simulations of the magnetic resonance were performed to evaluate μ' of the nanocomposite films consisting of Ni nanoparticles. The positive μ' was obtained in the region $H_{\text{ext}}^{\text{sw}} > H_0'$. On the other hand, the below resonance condition $H_{\text{ext}}^{\text{sw}} < H_0'$ brought about the negative μ'. The value of μ' was strongly influenced by the volume fraction of Ni (η). As η increases, the negative peak of permeability $\mu_{\min}$ becomes small. The lower limit of volume fraction for the negative μ' was $\eta > 0.035$ for Ni particles having an average diameter of 8 nm. The negative permeability was also affected by the dispersion of a particle diameter. As the deviations of diameter (δ_m) increases, the negative peak of permeability drastically shrinks. In comparison with the positive peak of permeability, the negative permeability $\mu_{\min}$ was unstable. However, the region of applied field ΔH_{neg}, where μ' was negative, becomes broader with increasing δ_m. This may open the door to broadband left-handed metamaterials in microwave regions when the negative ϵ media such as metal wire grids[20] are combined.

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