Microwave reflection properties of planar anisotropy $Fe_{50}Ni_{50}$ powder/paraffin composites*

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(Received 14 July 2011; revised manuscript received 30 October 2011)

The reflection properties of planar anisotropy $Fe_{50}Ni_{50}$ powder/paraffin composites have been studied in the microwave frequency range. The permeability of $Fe_{50}Ni_{50}$ powder/paraffin composites is greatly enhanced by introducing the planar anisotropy, and can be further enhanced by using a rotational orientation method. The complex permeability can be considered as the superposition of two types of magnetic resonance. The resonance peak at high frequency is attributed to the natural resonance, while the peak at low frequency is attributed to the domain-wall resonance. The simulated results of the microwave reflectivity show that the matching thickness, peak frequency, permeability, and permittivity are closely related to the quarter wavelength matching condition. The $Fe_{50}Ni_{50}$ powder/paraffin composites can be attractive candidates for thinner microwave absorbers in the L-band (1–2 GHz).

Keywords: reflection property, complex permeability, microwave absorption

PACS: 76.50.+g, 75.30.Gw **DOI:** 10.1088/1674-1056/21/3/037601

1. Introduction

Recently, the electromagnetic wave absorption materials in the L-band of microwave frequency range (1–2 GHz) have attracted considerable attention due to serious electromagnetic interference.[1,2] The downsizing of the portable communication devices, such as cellular phones, demands a thinner microwave absorption layer. Complex permittivity and permeability of microwave absorbers play key roles in determining the reflection or absorption properties. The minimal reflection is obtained at given frequencies if the thickness of the absorber (t) satisfies[3–5]

$$t = \frac{n\lambda}{4} = \frac{n\lambda_0}{4\sqrt{(|\mu_r||\varepsilon_r|)}}, \quad (n = 1, 3, 5, \ldots), \qquad (1)$$

where λ_0 and λ are the wavelengths in vacuum and material, μ_r and ε_r are the complex permeability and permittivity of the material relative to vacuum, respectively. A thinner absorber layer can be achieved by loading material with high μ_r and ε_r.[3] The permittivity can be tuned conveniently by using the conductive polymer.[6] However, limited by the impedance matching condition that $Z_m = Z_0\sqrt{\mu_\mathrm{r}/\varepsilon_\mathrm{r}}$, increasing the permittivity unilaterally will lead to low reflection loss. Here, Z_0 is the impedance of air. Hence, high permeability and moderate permittivity are needed at the same time to achieve low reflection loss.[7,8]

The permeability of traditional magnetic materials at the quasi-microwave frequency range from 1 to 2 GHz is small due to Snoek's limit[9,10] as follows:

$$(\mu_\mathrm{i} - 1)f_\mathrm{r} = \frac{2}{3}\gamma' M_\mathrm{s}, \qquad (2)$$

where μ_i, f_r, γ', and M_s are the initial permeability, resonance frequency, gyromagnetic ratio and saturation magnetization of the material, respectively. However, if the magnetic moment of the material lies mainly in a plane (denoted as planar anisotropy), based on the Landau–Lifshitz–Gilbert (LLG) equation, Snoek's limit becomes[9]

$$(\mu_\mathrm{i} - 1)f_\mathrm{r} = \frac{2}{3}\gamma' M_\mathrm{s}\sqrt{\frac{H_\mathrm{ha}}{H_\mathrm{ea}}}, \qquad (3)$$

where H_ea and H_ha represent the in-plane and out-of-plane anisotropic fields, respectively. Planar anisotropy exists in many systems with a easy magnetization plane, such as soft magnetic thin films,[9] flake-like particles,[11] rare earth-3d intermetallics, and hexaferrite (Co_2Z).[12,13] The components of the magnetic moment mainly lie in the easy magnetization

*Project supported by the National Natural Science Foundation of China (Grant No. 10774061), the Defense Industrial Technology Development Program, China (Grant No. A1420080174), and the Fundamental Research Funds for the Central Universities.

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plane. For rare earth-3d intermetallics and the hexaferrites with a c-plane anisotropy,[13] the easy magnetization plane is introduced by the magnetocrystalline anisotropy. For soft magnetic thin films and the flake-like particles, the easy magnetization plane is introduced by shape anisotropy. $H_{\rm ha}$ approximates to the demagnetizing field ($H_{\rm ha} \approx N_z 4\pi M_{\rm s}$, where N_z is the demagnetization factor) due to the shape of the magnetic body. Here, $N_z \approx 1$ for the particles with a large aspect ratio.[14] Generally, $4\pi M_{\rm s}$ is much larger than the in-plane anisotropic field in the soft magnetic materials used as the microwave absorber. Hence, by introducing a simple planar anisotropy magnetization plane, the intrinsic high frequency property can be improved greatly compared with traditional materials constrained by Snoek's limit. Therefore, the flake-like metallic soft magnetic materials may have high permeability in the microwave band due to the high saturation magnetization and planar anisotropy.

This study provides a planar anisotropy $Fe_{50}Ni_{50}$ powder/paraffin composite with high permeability in the microwave frequency range. A method of rotating orientation is used to improve the permeability further. This composite can be used as a thinner microwave absorber in the L-Band.

2. Experimental details

The flake-like $Fe_{50}Ni_{50}$ powder was prepared by ball milling with commercially available $Fe_{50}Ni_{50}$ powder (10 μm) as the starting materials. The ball milling was carried out using a planetary ball mill for 16 hours. Both the ball and bowl were made of hardened steel. The weight ratio of ball to powder was fixed at 25:1. Heptane mixed with a 3% solution of oleic acid was used as the process control agent. X-ray diffraction (XRD) analysis was performed to investigate the phases of the ball-milled product. The morphology was examined by using a high-resolution scanning electron microscope (SEM). The Mössbauer spectrum was measured to confirm the magnetic moment distribution of flake-like particles.

The composite samples were prepared by uniformly mixing powders in paraffin with 30 vol.% $Fe_{50}Ni_{50}$ powder. The mixture was pressed to form a cylindrical/toroidal sample with an outer diameter of 7.0 mm and inner diameter of 3.04 mm. Its thickness was controlled to be as thin as about 1 mm to avoid the dimensional resonance of the electromagnetic wave in a coaxial line. The oriented sample was prepared in a copper mold by a rotational orientation method. During this process, the $Fe_{50}Ni_{50}$ powder/paraffin composite was first warmed up to 80 °C to melt the paraffin wax. Next, the container was mounted on a rotating motor and placed in an applied magnetic field of about 1.2 T with its rotated axis perpendicular to the field until the paraffin solidified, so that the powders could rotate freely in the melted paraffin. This process ensured that the easy magnetization plane of each particle was parallel to the plane of the toroidal samples. These toroidal samples were studied using a vector network analyzer (Agilent E8363B) in the range of 0.1–18 GHz. The complex permittivity ($\varepsilon_{\rm r} = \varepsilon' - {\rm j}\varepsilon''$) and complex permeability ($\mu_{\rm r} = \mu' - {\rm j}\mu''$) were determined by the measured reflection coefficient S_{11} and the transmission coefficient S_{21} in an APC-7 coaxial mode.

For convenience, the planar anisotropy $Fe_{50}Ni_{50}$ powder/paraffin composites with and without orientation are denoted as oriented and un-oriented samples, respectively.

3. Results and discussion

Figure 1(a) presents the XRD pattern of the milled product, which reveals a fcc FeNi alloy phase (Joint Committee on Powder Diffraction Standard No. 47-1405). The saturation magnetization $M_{\rm s}$ and coercivity $H_{\rm c}$ of the milled product are 143 emu/g (1 emu/g=1 $\rm A{\cdot}m^2{\cdot}kg^{-1}$) and 19.2 Oe (1 Oe=79.5775 $\rm A{\cdot}m^{-1}$), which are determined by a hysteresis loop at room temperature (see Fig. 1(b)). Due to the effective strain suppression of the surface-active substance, the coercive field of the milled product is much smaller than that of the other ball-milled FeNi alloys.[15] The high saturation magnetization and the small coercive field also indicate that it is a good soft magnetic material. The morphology of the milled product is presented in Fig. 1(c). The diameter of the flake-like particles is about 40 μm and their thickness is about 1 μm. The large aspect ratio of about 40:1 results in large shape anisotropy, and that the magnetic moments preferentially lie in the flake plane. To investigate the magnetic moment distribution of the flake-like particles, the measurement of Mössbauer spectra at the room temperature was performed on the oriented sample. The Mössbauer spectra are fitted with two sextets and one doublet using the MossWinn program, as shown in Fig. 1(d). The angle between the incident γ-ray and the magnetization direction (θ) can be described by the relative intensity ratio of magnetically split sextet as follows:

$$\frac{I_{2,5}}{I_{3,4}} = \frac{4\sin^2(\theta)}{1+\cos^2(\theta)}, \tag{4}$$

where $I_{2,5}$ ($I_{3,4}$) is the relative intensity ratio of the peaks 2 and 5 (3 and 4) in magnetically split sextet. From the fitted results, it is derived that $I_{2,5}/I_{3,4} = 3.3$, and $\theta = 72°$. Since $\sin(72°) = 0.95$, 95% of the magnetic moments distributes in the easy magnetization plane. After orientation, the alignment of crystal grains does not change. However, the magnetic moment mainly lies in the plane of the sample. These results reveal that the ball-milled particles have an easy magnetization plane (planar anisotropy), which is favorable for improving the microwave permeability according to Eq. (3).

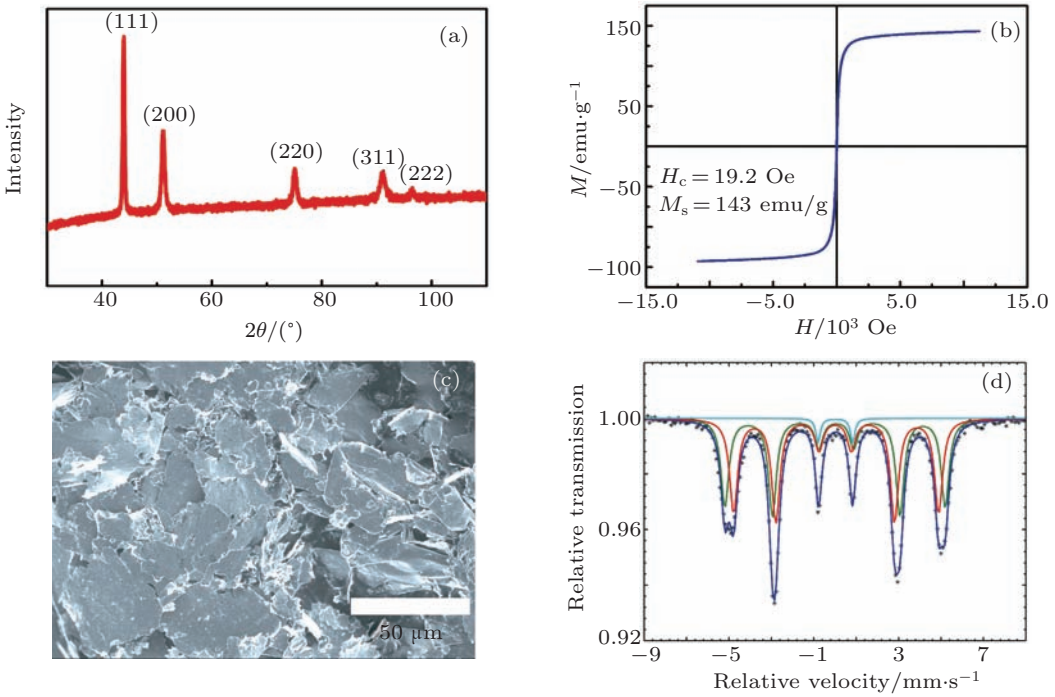


Fig. 1. (colour online) (a) XRD pattern, (b) hysteresis loop, (c) SEM image, and (d) Mössbauer spectrum of the ball-milled product.

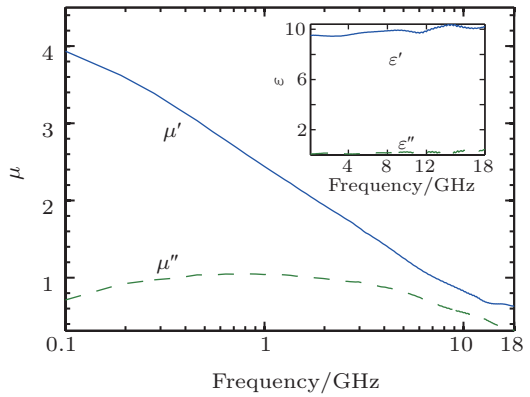


Fig. 2. (colour online) Frequency dependence of the complex permeability and permittivity (inset) spectra for original $Fe_{50}Ni_{50}$ powder/paraffin composite.

Figure 2 shows the frequency dependence of the complex permeability and complex permittivity for the original $Fe_{50}Ni_{50}$ powder/paraffin composite. The real part μ' is about 3.9 at 100 MHz, and decreases with the increasing frequency. The complex permittivity of the original $Fe_{50}Ni_{50}$ powder/paraffin composite is almost independent of the frequency within the measured frequency range.

The frequency dependence of the complex permittivity for the oriented and un-oriented samples are shown in Fig. 3. It is observed that the complex permittivity of the un-oriented sample is almost independent of the frequency within the measured frequency range, indicating that the sample is a non-percolation composite. The complex permittivity of the un-oriented sample is larger than that of the original one, which may be attributed to the flake-shape of the particles.[16] However, the oriented sample shows a significant increase in permittivity. After being oriented with an external magnetic field in the suspension, the magnetic particles agglomerate again, which can introduce more ohmic contact. Although the volume concentration of 30% is low, some limited region that is beyond the threshold of the percolation still exists in the sample. This results in high conductivity

and permittivity.[4,16,17]

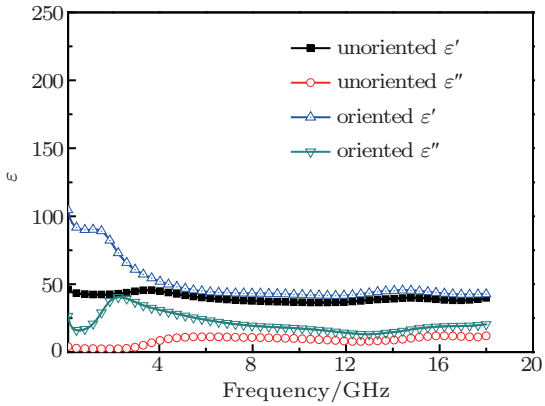


Fig. 3. (colour online) Frequency dependence of the complex permittivity spectra for a planar anisotropy $Fe_{50}Ni_{50}$ powder/paraffin composite without and with orientation.

Figure 4 shows the frequency dependence of the complex permeability for the oriented and un-oriented samples. The real part μ' is 12 and 10 at 100 MHz for the oriented and un-oriented samples respectively, and decreases with increasing frequency. μ' of the planar anisotropy $Fe_{50}Ni_{50}$ powder/paraffin composite is much larger than that of the original one. The permeability of the composite can be further enhanced after the composite is oriented, which is similar to the oriented hexagonal ferrite with planar anisotropy.[12,18] Generally, the frequency dispersion of the permeability can be characterized by the superposition of the domain-wall and natural resonances, which are induced by domain-wall motion and gyromagnetic spin rotation, respectively. The contribution of the domain-wall and natural resonances to the permeability spectra has been estimated in the spinel ferrite and the permalloy composite materials by fitting the permeability data to a formula containing the above-mentioned two components.[19–22] The numerical fitted results are shown in Fig. 4. For both of the samples with and without orientation, the resonance peak B at high frequency is attributed to the natural resonance, while the resonance peak A at low frequency is attributed to the domain-wall resonance, which is similar to Ref. [19]. The resonance frequency of the domain-wall resonance f_{rd} is equal to 0.25 GHz for two kinds of sample, and f_{rd} satisfies $2\pi f_{rd} = \alpha/\beta$ with α being the domain-wall energy, and β the damping factor. Because α and β are dependent on the intrinsic nature of materials, the resonance frequency of the domain-wall resonance for two kinds of sample is almost unchanged. However, the resonance frequency of the natural resonance f_r for the oriented sample is 3.5 GHz, which is higher than that of the un-oriented one (2.8 GHz). According to the LLG equation $f_r = \gamma'\sqrt{M_s H_{eff}}$, f_r is related to the saturation magnetization M_s and the effective anisotropic field H_{eff}. Since the raw materials of two kinds of samples are the same, M_s is the same. In the oriented sample, the flake planes of $Fe_{50}Ni_{50}$ particles are parallel to each other, introducing an additional effective anisotropic field due to the dipolar–dipolar interaction.[23] Therefore, the higher frequency of the natural resonance of the oriented sample is probably due to the special structure introduced by rotating the orientation.

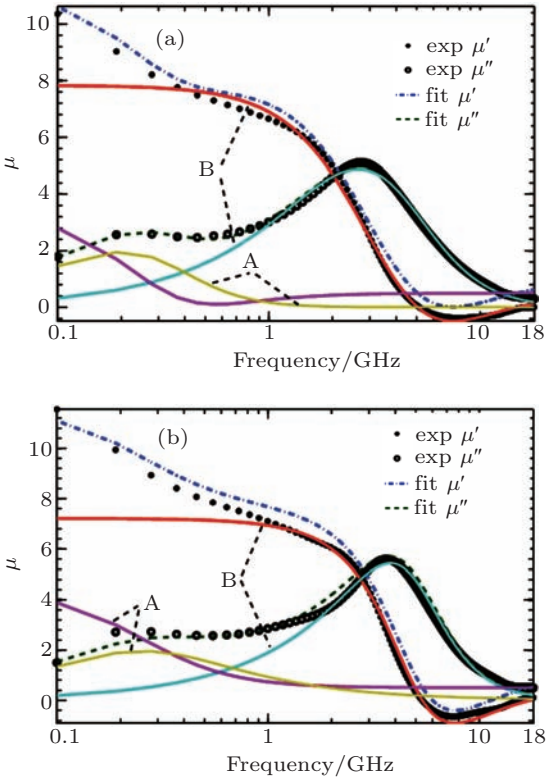


Fig. 4. (colour online) Frequency dependence of the measured and fitted complex permeability for the planar anisotropy $Fe_{50}Ni_{50}$ powder/paraffin composite (a) without and (b) with orientation.

Generally, good electromagnetic wave absorption results from efficient complementarity between the complex permittivity and permeability of the material.[7,8] To investigate the microwave absorption properties, the reflection loss (L_R) of the normal incident electromagnetic wave was calculated from the complex permeability and permittivity at a given frequency and absorber thickness with the following equations:

$$L_R = 20\lg|(Z_{in} - Z_0)/(Z_{in} + Z_0)|,$$
$$Z_{in} = Z_0\sqrt{\mu_r/\varepsilon_r}\tanh[\,j(2\pi ft/c)\sqrt{\mu_r\varepsilon_r}], \quad (5)$$

where f is the frequency of the electromagnetic wave, t is the thickness of the absorber, c is the velocity

of light, Z_0 is the impedance of air, and Z_{in} is the input impedance of the absorber. Figure 5 shows the frequency dependence of L_R for the two kinds of samples. L_R is found to be sensitive to the absorber thickness and the minimal L_R shifts to the low-frequency limit with the increasing thickness. A similar phenomenon has been reported elsewhere.[7,8,20] For the un-oriented sample, an optimal L_R of −49 dB is achieved at 1.3 GHz for a layer thickness of 3.3 mm, while an absorption exceeding −10 dB is obtained in the range of 0.8–2.8 GHz for a layer thickness of 4.0–1.5 mm. For the oriented sample, an optimal L_R of −16 dB is achieved at 0.8 GHz for a layer thickness of 3.5 mm. It is worth noting that the minimum L_R is −10 dB at 1.3 GHz for the oriented sample when the layer thickness is 2.0 mm, while the minimum L_R for the un-oriented one appears at a layer thickness greater than 3.0 mm. Hence, the un-oriented sample can serve as a broadband absorber in the L-band and the oriented sample can be used as a thin absorber in the microwave frequency range.

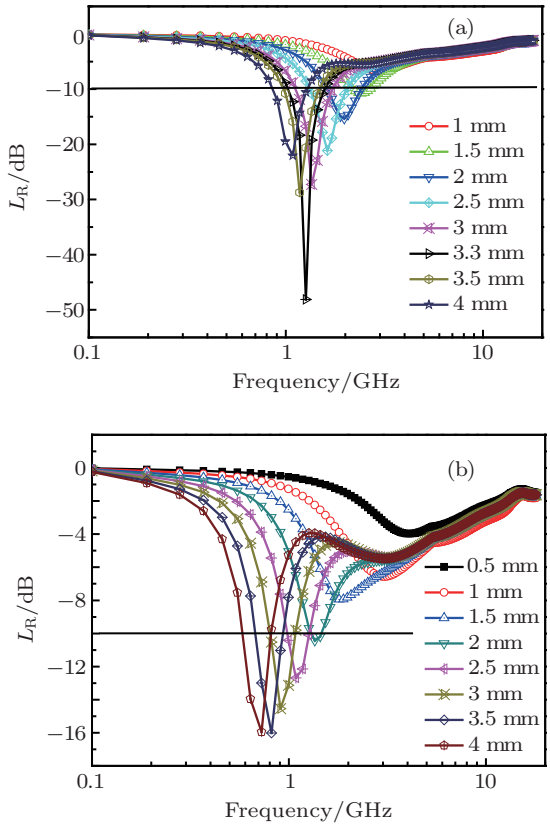


Fig. 5. (colour online) Frequency dependence of L_R for planar anisotropy $Fe_{50}Ni_{50}$ powder/paraffin composite (a) without and (b) with orientation.

When an electromagnetic wave shines on an absorber sample backed by a metal plate, it is partially reflected from air to the absorber interface and partially reflected from absorber to the metal interface. When the thickness of the absorber (given by Eq. (1)) is equal to a quarter wavelength in the absorber, these two reflective waves are out of phase (180°), and interfere destructively with each other.[24] The frequency dependence of $\lambda/4$ is shown in Fig. 6 for the oriented and un-oriented samples. Kim *et al.*[8] and Li *et al.*[20] defined the matching thickness (t_m^{sim}) as the thickness of the minimum dip that exceeds −30 dB among the curves of the reflection loss. Here, we take it as the perfect matching thickness. From Fig. 5, we can see that each curve of the reflection loss has a dip. The corresponding frequency and thickness of the dips are regarded as the peak frequency and matching thickness (including the perfect matching thickness), respectively. The matching thickness t_m^{sim} is also plotted in Fig. 6. The calculated thickness of $\lambda/4$ is well consistent with t_m^{sim}, not only for the perfect matching condition but also for the finite matching. This implies that the peak frequency, the matching thickness t_m^{sim}, μ, and ε are closely related to the quarter wavelength matching condition. It also indicates that a much smaller matching thickness of the oriented sample in the same frequency band is attributed to the enhancement of the permeability and permittivity. However, the relatively large permittivity leads to poor reflection loss due to the mismatch in impedance matching.

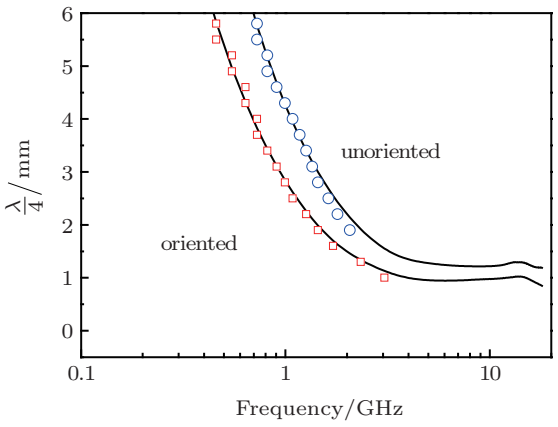


Fig. 6. (colour online) Frequency dependence of the matching thickness (open symbol) and $\lambda/4$ (solid line) for planar anisotropy $Fe_{50}Ni_{50}$/paraffin composite without and with orientation.

4. Conclusion

In summary, by introducing planar anisotropy, a $Fe_{50}Ni_{50}$ powder/paraffin composite is found to have fine reflection properties in the microwave frequency range. The resonance peak at high frequency is attributed to the natural resonance, while the peak at

low frequency is attributed to the domain-wall resonance. The permeability and permittivity of the flake-like $Fe_{50}Ni_{50}$ can be further enhanced by a rotational orientation method, resulting in a smaller matching thickness in L-band. The planar anisotropy $Fe_{50}Ni_{50}$ can be used as a L-band frequency absorber with thin layers.

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