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Influence of shape anisotropy on microwave complex permeability in carbonyl iron flakes/epoxy resin composites*

Wen Fu-Sheng(温福昇)†, Qiao Liang(乔 亮), Zhou Dong(周 栋), Zuo Wen-Liang(左文亮), Yi Hai-Bo(伊海波), and Li Fa-Shen (李发伸)

Institute of Applied Magnetics, Key Laboratory of Magnetism and Magnetic Materials of Ministry of Education, Lanzhou University, Lanzhou 730000, *China*

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To explore the mechanism of carbonyl iron flake composites for microwave complex permeability, this paper investigates the feature of the flakes. The shape anisotropy was certified by the results of the magnetization hysteresis loops and the Mössbauer spectra. Furthermore, the shape anisotropy was used to explain the origin of composite microwave performance, and the calculated results agree with the experiment. It is believed that the shape anisotropy dominates microwave complex permeability, and the natural resonance plays main role in flake.

Keywords: carbonyl iron flake, complex permeability, shape anisotropy, Mössbauer spectroscopy
PACC: 7520E, 7540G, 7530G, 7680

1. Introduction

The complex permeability spectrum via frequency in gigahertz is one of typical measurements utilized to characterize microwave properties. The magnetic materials with high values of complex permeability are possible to design thinner absorbers.[1] Consequently, there is considerable interest in exploiting the large $4\pi M_{\rm s}$ of soft ferromagnetic alloys to fabricate magnetic objects for linear inductive sensor and magnetic effective media that operate at microwave frequencies, which is ascribed to their higher Snoek's limit.[2−5] Nevertheless, the high-frequency permeability of metallic magnetic materials may decrease due to the eddy current phenomenon induced by electromagnetic (EM) wave interference.[6] It is better to use smaller, thinner and isolated metallic particles with a size less than the skin-depth (1 μm for iron in the 1-5GHz range) for suppressing the eddy current phenomenon to enhance an effective incidence to EM wave absorbers.[6]

In this paper we are interested in flakes of magnetic elements such as Carbonyl iron,[7,8] Alnico,[9] Fe-Ni,[10] Fe-Co,[11] etc. In the past few years, several models of microwave resonance were built up by introducing magnetocrystalline anisotropy into dynamic magnetization in spherical particles, for instance spinel ferrites and hexaferrites.[12] However, magnetic resonance is well known to be very sensitive to the shape of material.[13] Shape anisotropy is considered to root with in each single flake for soft magnetic metal. Thus, magnetic resonance is expected to be sensitive to shape anisotropy field. Considering static magnetic, microscopic magnetic and structural properties, this work analyses the microwave complex permeability of flakes-resin composites. During this study, exploring the mechanism of microwave resonance, we pay attention to the characterization of carbonyl iron flakes-resin composites, especially the shape anisotropy of flake on the microwave properties.

2. Experimental

In this work, the raw material is purchased from Tianyi Superfine Metallic Powder Co. Ltd. Jiangsu province. The flakes were fabricated by milling for 16 h with 320 r/min using the high-energy planetary ball milling in hexane, and then dried. Aligned composites were prepared by mounting on a motor which rotates the cylindrical-shaped compact around its axis in a perpendicular magnetic field of about 0.6 T, until

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†E-mail: wenfsh03@126.com and lifs@lzu.edu.cn (Fashen Li)

the epoxy resin solidifies. This process ensures that the basal planes are aligned to each other and c-axis is also parallel to the axis of cylinder. The phase was examined by using x-ray diffraction (XRD) using Cu Kα radiation on Philips X'perts diffractometer. The morphology was analysed by scanning electron microscope (SEM) (HITACHI S-4800).

Magnetization hysteresis loops and Mössbauer spectra was performed on the aligned composites to determine the easy-magnetization direction. The magnetization hysteresis loops recorded by a vibrating sample magnetometer (Lakeshore 7304) parallel and perpendicular to the c-axis of the cylindrical-shaped composites at room temperature. The Mössbauer spectra are obtained at room temperature on a constant-acceleration spectrometer which utilized cobalt-57 source (Pd) and was calibrated at room temperature with α-iron foil. In the transmission geometry, the incident γ photon is parallel to the axis of cylinder.

The epoxy resin composites were prepared by epoxy resin with 40 vol.% flake composites and pressing into toroidal shape (φ_{out}: 7.00 mm, φ_{out}: 3.04 mm). The scattering parameters (S_{11}, S_{21}) were measured on the toroidal-shape samples by a network analyser (Agilent Technologies E8363B) in the range of 0.1–18 GHz. The relative complex permeability (μ_r) was determined from the scattering parameters.

3. Results and discussion

Figure 1(a) shows an XRD spectrum of random ball-milled carbonyl iron particles. The XRD pattern exhibits diffraction peaks from the (110), (200) and (211) plane of bcc Fe. There are well-defined α-Fe peaks and there is no detectable secondary phase for particles. The XRD of the aligned composites is shown in Fig.1(b) which exhibits peak ratios same as those of random powders. Mean crystallite size of the carbonyl iron particles were evaluated to be about 13 nm from the line broadening of the XRD peaks by using the Scherrer's formula. The pattern of achieved flake-shaped carbonyl iron particles is presented in Fig.2. The thickness of flakes is 0.4 μm, which is far less than the skin depth of iron (1 μm for iron in the 1–5 GHz range).[6]

Figure 3 shows the magnetization hysteresis loops of the cylindrical-shaped composites. It is obvious that it is easy to magnetize composites perpendicular to the c-axis, but hard to magnetize the composites parallel to the c-axis. Figure 4 shows the Mössbauer spectra of the cylindrical-shaped composites. The experimental data are fitted by a magnetic splitting sextet. The hyperfine field is 33.1 T, which is the same as the bulk of α-iron.

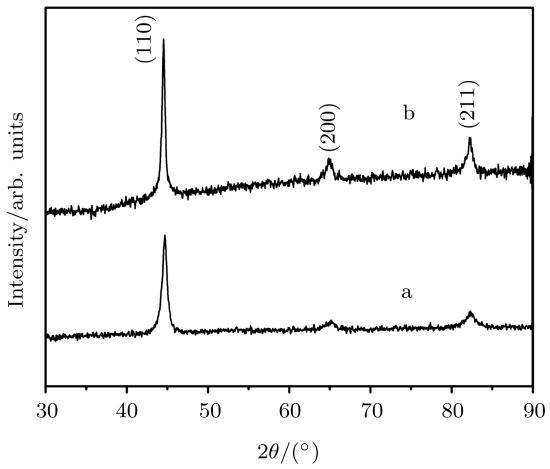


Fig.1. XRD spectrum of a random ball- milled particles and b the aligned composites.

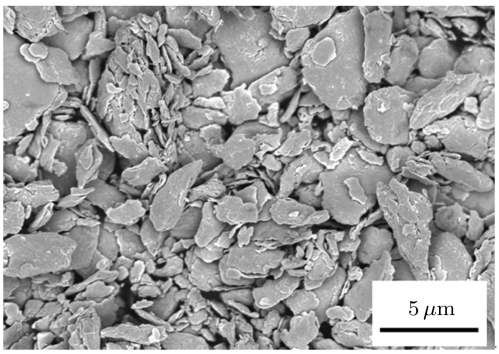


Fig.2. SEM image of carbonyl iron flakes.

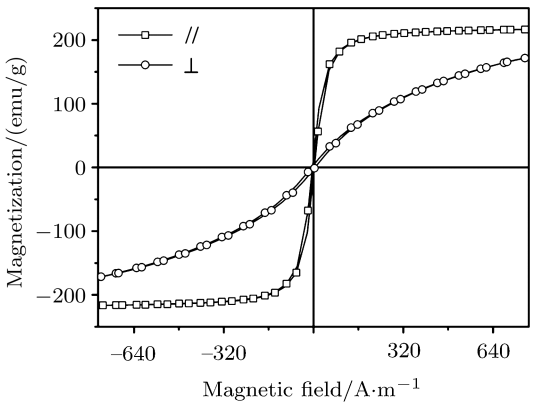


Fig.3. Magnetization hysteresis loops of the aligned composites at room temperature.

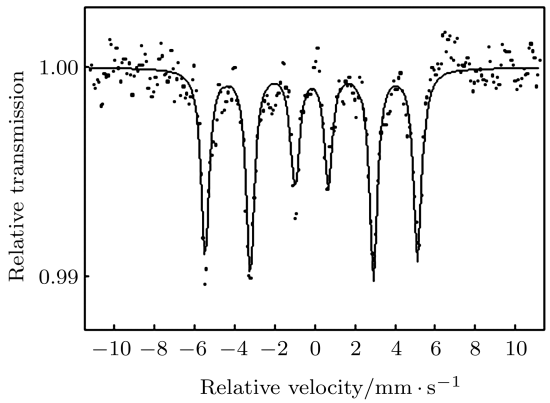


Fig.4. Mössbauer spectra of the aligned composites at room temperature.

In Mössbauer experiments, the angle (β) between the incident γ-photons and the magnetization direction can be assessed by the relative intensity ratio:[14]

$$\frac{I_{2,5}}{I_{1,6}} = \frac{4\sin^2\beta}{3(1+\cos^2\beta)}, \tag{1}$$

where $I_{2,5}$ and $I_{1,6}$ are the relative ratio of the 2, 5 and 1, 6 peaks in magnetic splitting sextet respectively. When the magnetic moments of the resonance atom ^{57}Fe, which can be detected, are arrayed randomly, the ratio between 2, 5 and 1, 6 peaks is 2/3.

Since the relative intensity ratio $I_{2,5}/I_{1,6}$ is 1.1, so the angle (β) is 72°. So the magnetic moments lean to the direction perpendicular to the c-axis of the composites. From Figs.3 and 4, we know that it is easy to magnetize the composites parallel to the plane of the flakes resulted from the demagnetization field of flaked shape. Therefore, the magnetic moments lean to the direction parallel to the plane of carbonyl iron flakes.

Generally speaking, magnetic materials with flaked shape have two types of shape anisotropy, namely c-axis anisotropy and basal plane anisotropy, respectively. For c-axis anisotropy, the magnetic field required for rotation of magnetization vector from basal plane to c-axis direction is defined as the anisotropy field. On the other hand, for basal plane anisotropy, an effective anisotropy field is introduced. When a magnetic field is applied, the magnetization vectors can be rotated from one to another easy magnetization direction in two ways: through the c axis or in the basal plane. The corresponding magnetic fields required are defined as anisotropy fields H_θ and H_ϕ, respectively, for the planar anisotropy. With the magnetic moments leaning to the direction parallel to the plane of carbonyl iron flake, c-axis anisotropy H_θ approximately equal to $4\pi M_s$ resulted from the demagnetization field.

Figure 5 shows the relative complex permeability for the epoxy resin composites with 40 vol.% carbonyl iron between 0.1 and 18 GHz. The real part of permeability μ_r' declined from 6.78 to around 1.00 with increasing the frequency in the 0.1–18 GHz range. The imaginary part of permeability μ_r'' firstly increased from about 0.00 to 2.47 at 4.1 GHz over the low frequency region, and decreased gradually in the higher frequency range.

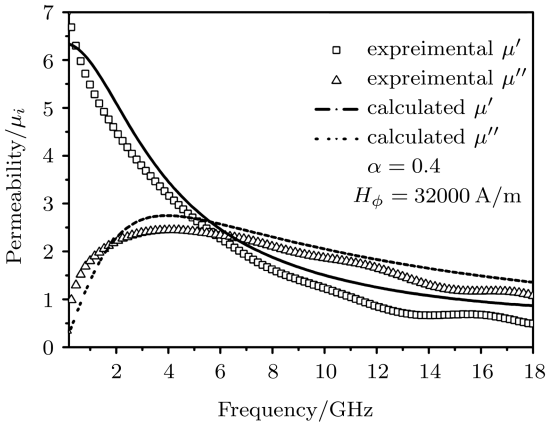


Fig.5. The experimental and calculated complex effective permeability of the composites with 40 vol.% of carbonyl iron flakes.

In general, the microwave magnetic loss of magnetic materials originates mainly from hysteresis, domain wall resonance, natural ferromagnetic resonance, and the eddy current loss. The hysteresis loss is mainly caused by the time lags of the magnetization vector behind the external electromagnetic field vector and is negligible in weak applied field. The domain wall resonance occurs usually only in the 1–100 MHz range and the thickness of carbonyl iron flakes is far less than the skin depth of iron 1 μm. The eddy current loss contribution to the imaginary part permeability is related to thickness (d) and the electric conductivity (σ):[15–17]

$$\mu'' = \frac{2}{3}\pi\mu_0(\mu')^2 d^2 f\sigma, \tag{2}$$

where μ_0 is the permeability of vacuum. Eq.(2) can transfer to

$$\mu''(\mu')^{-2} f^{-1} = \frac{2}{3}\pi\mu_0 d^2\sigma. \tag{3}$$

If the magnetic loss only results from eddy current loss, the values of left-hand side of Eq.(3) should be

constant when frequency is varied, we can call this the skin-effect criterion. Figure 6 shows the value of $\mu''(\mu')^{-2}f^{-1}$ for the carbonyl iron flake composites, as we can seen, the values of $\mu''(\mu')^{-2}f^{-1}$ for flakes markedly increase with increasing frequency at 2–18 GHz, and the changing extent is bigger than 0.17. Therefore, it means that the magnetic loss in flake particles is mainly caused by the natural resonance.

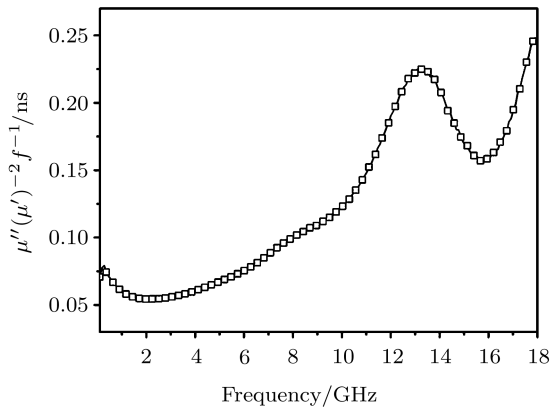


Fig.6. Values of $\mu''(\mu')^{-2}f^{-1}$ for the flake composites vs frequency.

Concerning magnetic metal, Refs.[8] and [18] reported the theoretical studies for behaviour of magnetization in microwave magnetic field. The magnetic flakes with a random spatial distribution of normal direction are considered to calculate the complex permeability of magnetic flake materials and its analytical model is derived from the Landau–Lifshitz–Gilbert equation and Bruggeman's effective medium theory.[8]

From the experimental data of magnetization hysteresis loops, the saturation magnetization $4\pi M_{\rm s}$ is 20 kGs. The theoretical fitting curves and experimental complex permeability curves are compared also in Fig.5. It is found that the experimental data are in good agreement with the theoretical line. The fitting result shows that $\alpha = 0.4$ and $H_\phi = 48000$ A/m. For bulk α-iron materials, the magnetocrystalline anisotropy field is 72000 A/m,[19] so the calculated effective basal plane anisotropy field is less than the magnetocrystalline anisotropy field. The calculated damping coefficient α is the same as the one of spherical carbonyl iron powders.[8] This demonstrates the validity of our idea in the above section that the magnetic loss in flake particles is mainly caused by the natural resonance. The effective permeability and the resonance frequency can be adjusted by the form of the basal plane and c-axis anisotropy field resulted from the shape of flake materials and polycrystalline isotropy frame. This result also indicates that the skin effect can be neglected which is caused by the thinner thickness of carbonyl iron flakes.

Comparing our results with those of the published ones in this field, we find that the obvious features are as follows.

(1) The flaked shape carbonyl iron particles have been presented in SEM image. Meanwhile, the magnetization hysteresis loops and the Mössbauer spectra prove that the magnetic moments lean to the direction parallel to plane of the flakes. The results demonstrate that the orientation of magnetic moments is caused by the demagnetization field from the flaked shape.

(2) For carbonyl iron flakes, good microwave magnetic properties have been obtained in the range of 0.1–18 GHz. The skin-effect criterion and the calculated results by using the combination of the LLG equation and Bruggeman's effective medium theory demonstrate that the magnetic loss mechanism is mainly caused by the natural resonance. The status that the magnetic moments lean to the direction parallel to plane of the flakes induces the shape anisotropy and the excellent microwave magnetic properties. The theoretical calculations agree with the experimental results.

4. Conclusions

In summary, we have investigated the microwave magnetic properties of carbonyl iron flakes embedded in epoxy resin. Our results reveal that the good agreement between calculated and experimental values indicates that the magnetic loss mechanism in flakes is mainly caused by the natural resonance. In our knowledge, this is the first study to demonstrate the origin of better microwave complex permeability for flake alloys which has shape anisotropy from experiment and calculation. The carbonyl iron flake with higher complex permeability might be used for thinner electromagnetic wave absorber.

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