Magnetic and microwave properties of glass-coated amorphous ferromagnetic microwires

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Abstract: Glass-coated amorphous FeCuNbSiB microwires were prepared by Taylor-Ulitovsky technique. X-ray diffractometry and scanning electron microscopy were used to investigate the microstructure and morphology of the glass-coated microwires respectively. The vibrating sample magnetometer and vector network analyzer were used to study the magnetostatic and microwave properties of glass-coated microwires. The experimental results show that the effective anisotropy of an array of 150 microwires of 10 mm in length is large than that of one microwire of 10 mm in diameter and an array of 150 microwires of 1 mm in diameter. The natural ferromagnetic resonance takes place as the microwave magnetic component is perpendicular to the microwires axis, and the electric dipole resonance takes place as the microwire is long or the short microwire concentration is moderate. The natural ferromagnetic resonance shifts to higher frequency with the larger microwire concentration. The electric dipole resonance is governed by the microwires length and concentration. The glass-coated FeCuNbSiB microwires can be used to design EMI filters and microwave absorbing materials.

Key words: glass-coated amorphous microwires; magnetic property; magnetic microwires-dielectric composite; natural ferromagnetic resonance; electric dipole resonance

1 Introduction

The magnetic properties of glass-coated microwires and microwave characteristics of magnetic wires-dielectric composites have received considerable attention due to their much application as magnetic sensors and potential applications as microwave materials and radio absorbing materials in recent years [1−3]. The amorphous magnetic alloy has outstanding magnetic properties[4−6].

The investigation showed that there are length effect[7] and dipolar effect[8] for the magnetic microwires. Other studies showed that the effective permittivity or permeability of microwires-dielectric composites exhibits characteristics of resonance or relaxation when the composites were excited by the electromagnetic wave[9], but the effect of various microwires length and concentration on the microwave properties was not considered. In the study of microwave properties of wires-dielectric composites[10], no coupling between the wires was taken into account.

To design microwave absorbing materials, glass-coated alloy microwires of various lengths and concentrations need to be selected to meet the requirements of particular applications. The objective of this work is to investigate the magnetostatic and microwave properties of glass-coated FeCuNbSiB microwires. The effect of microwires concentration and arrange on the microwave properties is analyzed. The mechanisms of natural ferromagnetic resonance(NFMR) and electric dipole resonance(EDR) of the glass-coated FeCuNbSiB microwires-dielectric composites are discussed.

2 Experimental

The glass-coated $Fe_{73.5}Cu_{1.0}Nb_{3.0}Si_{13.5}B_{9.0}$ microwires were prepared by the Taylor-Ulitovsky technique [11]. The microwires were quenched by tap water with a

cooling rate of 10^5–10^6 K/s during the preparation. The structures of the microwires were characterized by X-ray diffractometry(XRD, XPert PRO, Cu K$_\alpha$, 40 kV, 40 mA) and scanning electron microscopy (SEM, Philips Quanta 200, 200 kV).

The magnetostatic properties were measured by Vibrating Sample Magnetometer (VSM) of Model 3472-70 GMW. The studied number of 10 mm-long microwire was 1 and 150. The length of an array of 150 measured microwires placed side by side with their axis parallel to each other was 10 mm and 1 mm, respectively. In these hysteresis figures, "//" refers to the longitudinal magnetization with the external magnetic field applied along the microwires axis, and "⊥" refers to the transverse magnetization with the external magnetic field applied perpendicularly to the axis of the microwires.

The investigated composites having the form of cylindrical toroidal rings were prepared by bonding the microwires with rubber dissolved by acetone. The dimension of cylindrical toroidal composites is 3.04 mm in inner diameter, 7 mm in outer diameter and 3–4 mm in thickness. Two groups of composites were prepared [9,12–13]: the A-type composites A1, A2, and A3 were made by dispersing short microwires of 1–2 mm long randomly to rubber with microwires mass fractions of 1%, 15% and 25%, respectively; the B-type composites with a multithread coil-like structure of 50–80 mm microwires have the form of cylindrical toroidal rings with microwires mass fractions of 5%, 15% and 25%, respectively (Fig.1).

The T/R coaxial line method was used to determine the relative complex permeability $\mu=\mu'-\mathrm{j}\mu''$ and relative complex permittivity $\varepsilon=\varepsilon'-\mathrm{j}\varepsilon''$ of the composite samples with a HP8722ES vector network analyzer in the frequency range of 2–18 GHz. The maximal mass fraction of microwires in composite was 25% for the glabrous surface of coaxial composite samples with larger mass fraction was difficult to keep.

3 Results and discussion

3.1 Phase structure of microwires

Fig.2 shows the X-ray diffraction patterns of glass-coated FeCuNbSiB microwires. No phase peak is observed in the XRD pattern, which implies that the glass-coated microwires are amorphous. As can be seen from the SEM photograph(Fig.3), the diameter of the glass-coated microwires is 4.5 μm in metal core and 8 μm in glass-coating. The geometry of glass-coated microwires is successive and uniform with smooth surface and solid structure.

3.2 Magnetic properties of microwires
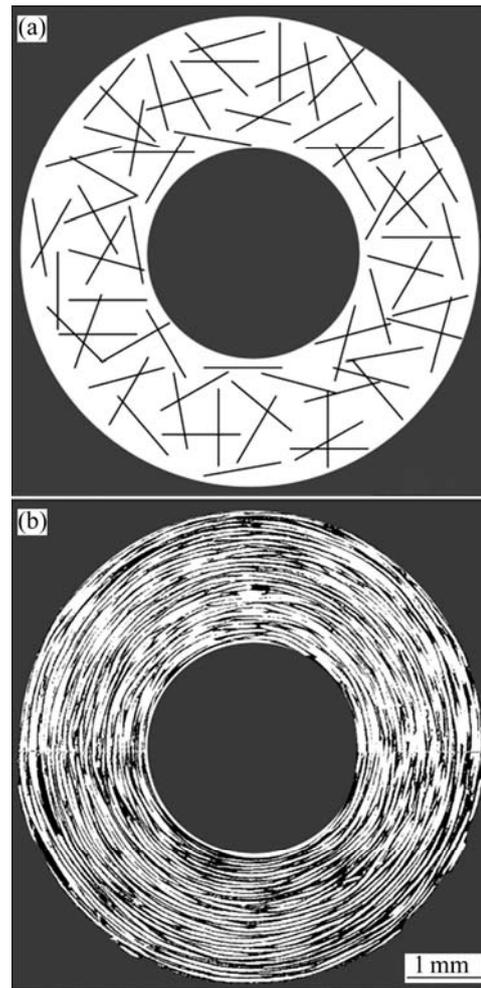
As illustrated in Fig.4, the hysteresis curve of a

Fig.1 Schematic structure of samples: (a) Isotropic A-type samples; (b) Anisotropic B-type sample
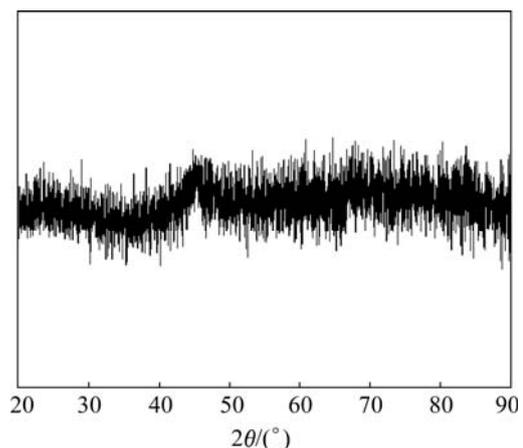

Fig.2 XRD pattern of glass-coated FeCuNbSiB microwires

single Fe-rich microwire exhibits typical squared loop with coercive force of 120 A/m, which is associated with the magnetic domain reversal of the metal inner core. As well known, the Fe-rich microwires have a longitudinally magnetized inner core and a radially magnetized thin outer shell[14–15]. The radius of the inner core, R_i, can be estimated according to the following equation:
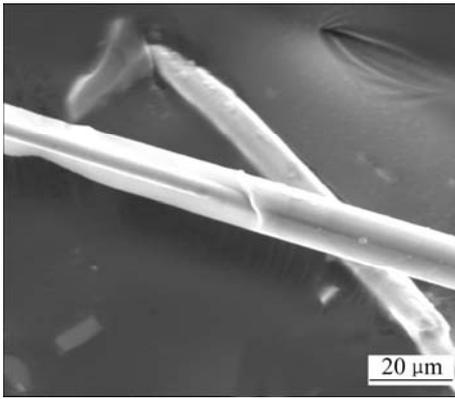
Fig.3 SEM photograph of glass-coated FeCuNbSiB microwires

Fig.5 High field axial and radial hysteresis loops of an array of 150 microwires with samples length of 1 and 10 mm

As the microwires are of limited length, a decrease in this length for an array of given microwires results in an increase in demagnetizing factor and hence a reduction in the ratio of the volume of the axially magnetized inner core to the total volume of the metallic wire. Thus the axial coercive field H_c and anisotropy field H_a along axis decrease.

3.3 Microwave permeability of microwires

The complex permeability spectra of the two types of composite with different microwires concentration and arrange orientation in the frequency range of 2−18 GHz are shown in Figs.6 and 7. Both the real and imagine part of permeability rise with the increase of the microwires concentration for both two types of composites. There exist some magnetic resonance for the composite containing high microwire concentration. The natural ferromagnetic resonance(NFMR) comes from the precession of the magnetic moment driven by the microwave magnetic field in an effective magnetic anisotropy field.
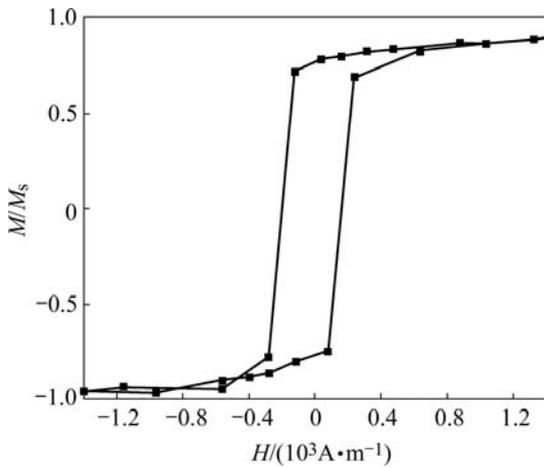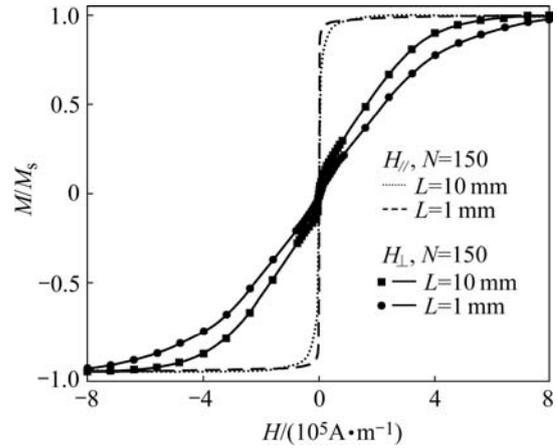

Fig.4 Axial hysteresis loop of one microwire with its axis parallel to external magnetic field

$$R_i \approx R_m \sqrt{M_r/M_s} \tag{1}$$

where R_m is the radius of the metal core, M_r/M_s is the remnant to saturation ratio of one microwire, which takes 0.8 according to Fig.4. So it can be estimated that the thickness of thin outer shell is about 0.2 μm.

The magnetic properties of an array of microwires depend on their length, as can be seen from Fig.5. The central part of hysteresis loops of all the samples corresponds to domain reversal of the metal inner cores. While the external part of the loops is dominated by domain rotational mechanism of the outer shell and the tangled domain of the metal core ends[16].

The magnetizing difference of one microwire and an array of microwires can be attributed to the influence of the long-range dipolar-dipolar interaction. When the microwire number increases to about 10, the dipolar interaction gives rise to an additional axial anisotropy, which results in a huge increase of H_a along the axis of the microwires[8]. This consequently would influence the macroscopic electromagnetic response of the system.
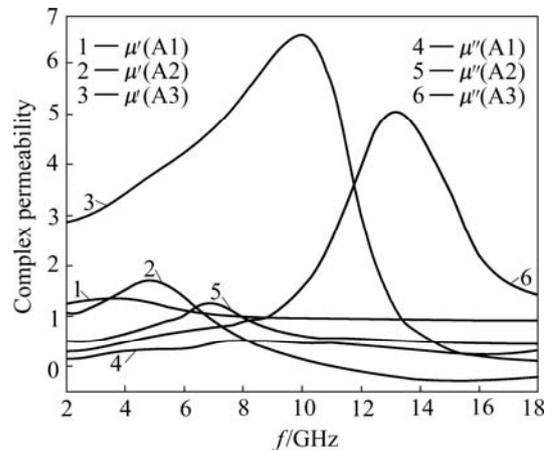

Fig.6 Permeability of composites with 1 mm long randomly oriented microwires versus frequency
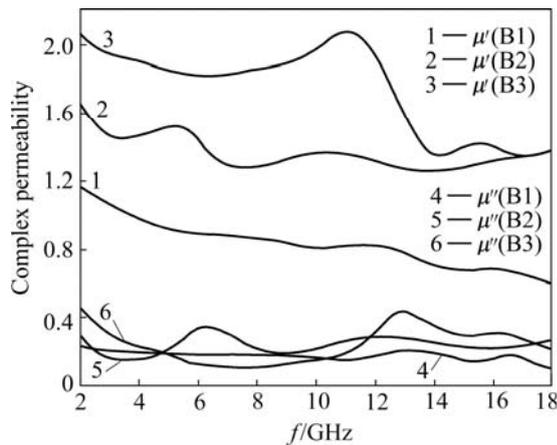
Fig.7 Permeability of composites with microwire wound round coaxial coil versus frequency

The magnetic resonance frequency shifts to higher frequency with higher microwire concentration. The real permeability of samples A1 is very low, while the imagine permeability is a little higher than zero with one weak resonance peak at 4 GHz. For sample A2 the average permeability shows a dispersive form in its real component and an obvious magnetic absorbing peak in its imaginary component. The complex permeability shows strong dispersion when the mass fraction of A-type composite increases to 25%.

The complex permeability of sample B1 is similar to that of A1 but having faster falling real component and basically invariable imaginary component. For sample B2 the magnetic resonance is irregular, as displayed in Fig.7. As the mass fraction increases to 25%, the resonance increases with more complicated frequency dispersion. The values of μ' for all of the two types of composite decrease with the increase of the frequency due to the eddy current loss and ferromagnetic resonance[10].

The skin depth δ of the electromagnetic wave in the microwires can be calculated with the following equation:

$$\delta = \sqrt{2/(\omega\mu\sigma)} \tag{2}$$

where ω is the angular frequency, σ is the conductivity of 0.8×10^6 S/m, μ is the intrinsic permeability taken as 5.5−6.5i at 5 GHz for the amorphous $Fe_{73.5}Cu_{1.0}Nb_{3.0}$-$Si_{13.5}B_{9.0}$ microwires. Thus δ for the microwires at 5 GHz is about 2.73 μm, larger than 0.2 μm of outer shell of the alloy core. At the NFMR frequency, μ suddenly increases by some times, causing δ value to decrease but still larger than the outer shell. So the microwave magnetic field can penetrate most of the inner metal core and consume large part of microwave magnetic energy in the measured frequency range.

In the excitation of microwave, there is magnetic component in the radial direction of most microwires of A-type composite. The moment of the microwires being most axially magnetized is excited by the microwave, resulting in the precession of the magnetic moment. Thus the NFMR takes place in especial frequency of f_r depending on the effective anisotropy field H_a.

The ferromagnetic resonance frequency f_r of glass-coated microwires can be calculated without external magnetic field (H=0) and $H_a \ll 4\pi M$[3]:

$$f_r = g\sqrt{MH_a/\pi} \tag{3}$$

where g is the gyromagnetic ratio of 2.21×10^5 m/(A·s), M is magnetization taken as 1.1 T, H_a is the anisotropy field. The effective anisotropy field for one microwire is about 2 400 A/m, and that for an array of 150 microwires is about 6 000 A/m. The high anisotropy field causes the high NFMR frequency that is in the range of 4−12 GHz.

The length of short microwires of 1−2 mm is close to the critical length of the microwires for the demagnetizing energy destroys the uniformly magnetized domain of the inner core. The H_a of the short microwires is non-uniform and less than that of the long microwire, resulting in the broadening of the resonance half-width.

The permeability spectra of the B-type composite may be partly from the non-strictly circumferential arrangement and non-uniform lengths of the microwires. There is also a thin outer shell and tangle magnetized domain in the two ends of the microwires that have low effective anisotropy field. There is little part of magnetic moments being perpendicular to the microwave magnetic field. Thus there is magnetic resonance instead of continuous reduction of the permeability, as illustrated in Fig.7.

With the increase of the microwire concentration in the composite, the distance between the microwires is shortened. So the stronger dipolar interaction causes the increase of the effective anisotropic field H_a. Therefore the resonance frequency rises with the increase of the microwires concentration.

For A-type of composite, the length l of microwires is in a certain range (1−2 mm). The microwires length l is comparable to $l_{eff}/2$ (l_{eff} is the effective wavelength of the electromagnetic field in the composite). The geometric resonance may be take place in some frequency.

3.4 Microwave permittivity of microwires

Resonance or relaxation type of permittivity spectra can be seen from Figs.8 and 9 for different microwires concentration and structure. The values of both ε' and ε'' for composite A2 are higher than those of A1 and far lower than those of A3. The dielectric absorption changes
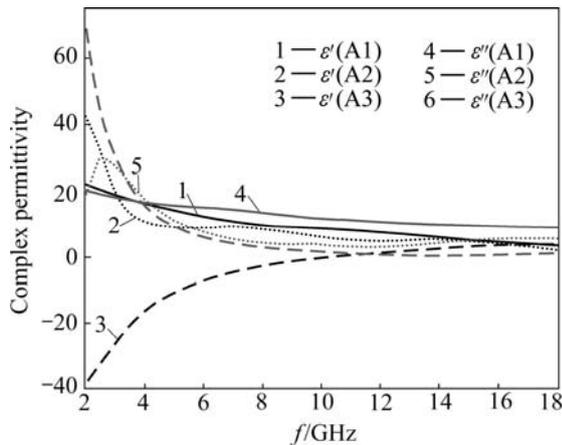
Fig.8 Permittivity of composites with 1 mm long randomly oriented microwires versus frequency
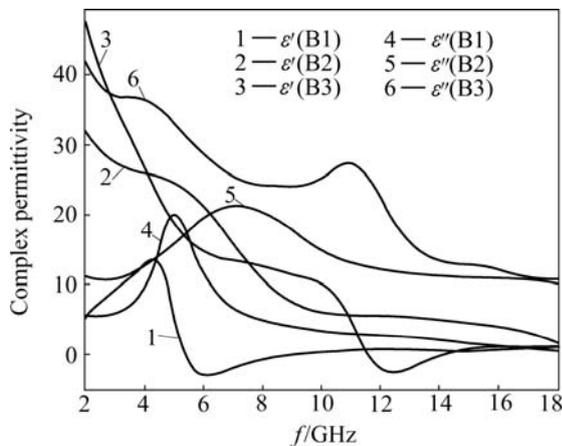

Fig.9 Permittivity of composites with microwire wound round coaxial coil versus frequency

from resonance-type to relaxation-type with the increase of the microwires concentration.

The alloy microwires behave as electric dipole in the excitation of the microwave electric field. The collective oscillation of free electrons is excited by the microwave electric field, generating the line current in the microwires. Thus the microwires consume the incident electromagnetic energy and cause the occurrence of electric dipole resonance(EDR) at given frequency, which is like micro-antennas. The EDR frequency of the composite with low microwires concentration is expressed as[17]:

$$f_{\mathrm{res},n} \approx \frac{c(2n-1)}{2l\sqrt{\varepsilon}} \tag{4}$$

where $f_{\mathrm{res},n}$ is the EDR frequency, c is the light velocity, n is natural number, l is the microwire length, ε is the permittivity of matrix taken as 2. The first electric dipole resonance frequency is at the lowest frequency with a maximal response. As can be seen from Eqn.(4), the

EDR frequency is inverse with the length of microwires. There is EDR for the long microwires in 2−18 GHz. The electric response is intensified for the interaction between the microwires in the composite with the high microwire concentration, resulting in the increase of the permittivity. Thus the permittivity shifts to higher frequency and the dielectric absorption peak broadens with larger mass of microwires.

The permittivity of short microwire composite is complicated. The permittivity of A-type composite with high microwires concentration is some like the randomly distributed conductive fiber-dielectric composite. For the composite with dilute short microwires, the permittivity decreases with increasing the measured frequency. As the short microwire concentration is not very low, such as sample A1, the moderate electric interaction between the radiant electromagnetic wave may result in the resonance of the permittivity. When the microwire concentration is high enough, the shortened distance between the microwires causes the increase of the contact probability between the microwires. Thus the intense metallicity of the microwire composites may result in the negative real permittivity.

4 Conclusions

1) The anisotropy of an array of long glass-coated amorphous $Fe_{73.5}Cu_{1.0}Nb_{3.0}Si_{13.5}B_{9.0}$ microwires is higher than that of an array of short amorphous microwires.

2) The electromagnetic properties depend on the microwires concentration and arrange orientation. The microwave permeability and permittivity of composite increase with the increase of the microwire concentration.

3) The NFMR is more remarkable for the randomly distributed microwires-dielectric composite. The EDR takes place as the long microwires are parallel to the microwave electric field. The NFMR and EDR frequency of composites shift to higher frequency with the higher microwire concentration.

4) The high microwave electromagnetic loss of glass-coated Fe-rich microwires makes them be used as EMI filters and applied to develop microwave absorbing materials.

References

[1]   PANINA L V, MAKHNOVSKIY D P, MOHRI K. Magnetoimpedance in amorphous wires and multifunctional applications: From sensors to tunable artificial microwave materials [J]. J Magn Magn Mater, 2004, 272/276(2): 1452−1459.

[2]   BARANOV S A. Use of a microconductor with natural ferromagnetic resonance for radio-absorbing materials [J]. Tech Phys Lett, 1998, 24(7): 549−550.

[3]   MARÍN P, CORTINA D, HERNANDO A. High-frequency behavior of amorphous microwires and its applications [J]. J Magn Magn

Mater, 2005, 290/291(2): 1597−1600.

[4]　CAO Ling-fei, WANG Ming-pu, XIE Dan, GUO Ming-xing, LI Zhou, TAN Wang, XU Gen-ying. Thermal behavior and structure of $Fe_{84}Nb_7B_9$ nanocrystalline powders [J]. Trans Nonferrous Met Soc China, 2006, 16(2): 299−303.

[5]　WEN Qi-ye, ZHANG Huai-wu, SONG Yuan-qiang, JIANG Xiang-dong. Theoretical and experimental investigation on giant magnetoresistive materials with amorphous ferromagnetic layer [J]. Trans Nonferrous Met Soc China, 2006, 16(s1): 59−62.

[6]　LU Bin, YI Dan-qing, YAN Biao, YIN Jun-lin, LIU Yan, LIU Hui-qun, WU Wei, MA Rui. High-pressure sintering and magnetic properties of $Fe_86Zr_{11-x}Nb_xB_3$(x=5.5, 6) amorphous alloys [J]. Trans Nonferrous Met Soc China, 2005, 15(4): 828−833.

[7]　ZHUKOV V, USOV N A, ZHUKOV A, GONZALEZ J. Length effect in a Co-rich amorphous wire [J]. Phys Rev B, 2002, 65(13): 134407-1−7.

[8]　SINNECKER E H C P, MENEZES F S, SAMPAIO L C, KNOBEL M, VÁZQUEZ M. Tailoring coercivity in an array of glass-coated microwires [J]. J Magn Magn Mater, 2001, 226/230(2): 1467−1469.

[9]　STAROSTENKO S N, ROZANOV K N, OSIPOV A V. Microwave properties of composites with glass coated amorphous magnetic microwires [J]. J Magn Magn Mater, 2006, 298(1): 56−64.

[10]　WU M Z, HE H H, ZHAO Z S, YAO X. Electromagnetic and microwave absorbing properties of iron fibre-epoxy resin composites [J]. J Phys D: Appl Phys, 2000, 33(8): 2398−2401.

[11]　LARIN V S, TORCUNOV A V, ZHUKOV A, GONZALEZ J, VAZQUEZ M, PANINA L. Preparation and properties of glass-coated microwires [J]. J Magn Magn Mater, 2002, 249(1/2): 39−45.

[12]　ACHER O, ADENOT A L, DEPROT S. Parallel permeability of ferromagnetic wires up to GHz frequencies [J]. J Magn Magn Mater, 2002, 249(1/2): 264−268.

[13]　WU M Z, HE H H, ZHAO Z S, YAO X. Electromagnetic anisotropy of magnetic iron fibres at microwave frequencies [J]. J Phys D: Appl Phys, 2001, 34(4): 1069−1074.

[14]　VELÁZQUEZ J, GARCÍA C, VÁZQUEZ M, HERNANDO A. Interacting amorphous ferromagnetic wires: A complex system [J]. J Appl Phys, 1999, 85(5): 2768−2773.

[15]　CHIZHIK A, ZHUKOV A, BLANCO J M. Interaction between Fe-rich ferromagnetic glass-coated microwires [J]. J Magn Magn Mater, 2002, 249(1/2): 99−103.

[16]　PEROV N S, RADKOVSKAYA A A, ANTONOV A S, SZYMCZAKD R, GONZALEZ J. Magnetic properties of short amorphous microwires [J]. J Magn Magn Mater, 1999, 196/197(5): 385−387.

[17]　MAKHNOVSKIY D P, PANINA L V. Field dependent permittivity of composite materials containing ferromagnetic wires [J]. J Appl Phys, 2003, 93(7): 4120−4129.

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