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Effects of eddy current and dispersion of magnetic anisotropy on the high-frequency permeability of Fe-based nanocomposites

M. Han [a,*], K.N. Rozanov [b], P.A. Zezyulina [b], Yan-Hui Wu [a]

[a] State Key Laboratory of Electronic Thin Films & Integrated Devices, University of Electronic Science and Technology of China, Chengdu, China
[b] Institute for Theoretical and Applied Electromagnetics, Russian Academy of Sciences, Moscow, Russia

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ABSTRACT

Fe–Cu–Nb–Si–B microflakes have been prepared by ball milling. The structural, magnetostatic and microwave permeability of the flakes and flake-filled composites have been studied. Two ferromagnetic phases, nanograins and amorphous matrix, are found in the flakes. The Mössbauer study shows that the nanograins are α-$Fe_3(Si)$ with $D0_3$ superlattice structure. High resolution transmission electron microscopy shows that the nanograins are well dispersed in the matrix. The microwave permeability of composites containing the flakes has been measured. The comparison of the intrinsic permeability of the flakes obtained from the permeability measurements and from the anisotropy field distribution reveals a disagreement in the magnetic loss peak location. It is concluded that the low-frequency loss in the composites is not due to the effect of eddy currents. The low-frequency loss may be attributed to other sources, such as domain wall motion or peculiarities of the magnetic structure of the flakes in the composite.

1. Introduction

To design magnetic components operable at high frequencies, the magnetic permeability ($\mu = \mu' - i\mu''$) is an important parameter that has to be taken into account. Some magnetic components require large real part of complex permeability and small imaginary part (i.e., low magnetic loss), e.g., magnetic cores in common-mode choke coils, power transformers, and active filters [1]. Other components, such as electromagnetic noise suppressors, demand large magnetic loss at high frequency.

Ferromagnetic alloys with soft magnetic properties are one of the critical materials in electrical engineering. For applications at frequencies above 1 GHz (GHz), ferromagnetic alloys are employed as tiny particles of various shapes to reduce the effect of eddy currents and to retain therefore large values of high-frequency permeability. The most studied shapes are microwires, nanowires, nanobelts and microflakes [2–4].

From the perspective of engineering, microflakes can be fabricated on a large scale. The strong shape anisotropy is beneficial to increase the natural resonance frequency (f_{res}) above 1 GHz, which enables the flakes to be employed as fillers in electromagnetic wave absorbing composites with high magnetic loss resulting from the natural resonance. Properly annealed Fe–Cu–Nb–Si–B alloys (FINEMET) have good soft magnetic properties with high initial permeability (about 100,000) [5]. Although the high-frequency permeability of ferromagnetic flakes has been intensively studied [6,7], the frequency dependences of permeability of such materials are frequently hard to be understood.

Several approaches have been suggested recently to analyze the dispersion of high-frequency permeability. For example, the skin criterion [8] exploits the equation

$$\mu''/\left(\mu'^2 f\right) = K_\delta a^2 \sigma \quad (1)$$

where the left part is found from measured permeability. If the left part is a constant in the low frequency range, then it is believed that the product of characteristic particle size (a, the thickness for platelet inclusions) and conductivity (σ) can be found from this constant. The value of K_δ depends on the inclusion shape and is close to $(\pi/c)^2$, where c is the light velocity for sphere, cylinder and film [9]. Eq. (1) holds for the permeability of the inclusions in a composite rather than to the effective permeability of the composite [9].

It is readily shown that a low-frequency region, where the left part of (1) is independent of frequency, exists for any frequency dependence of permeability. Indeed, the analytical consideration of frequency dependence of material parameters [10] shows that the real permeability is an even function of frequency and the

* Corresponding author
E-mail address: mangui@gmail.com (M. Han).

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imaginary permeability is an odd function of frequency. Therefore, any reasonable frequency dependence must involve a low-frequency region, where the real permeability is close to a constant and the imaginary permeability is proportional to the frequency. Therefore, the left part of (1) is a constant. Actually, the application of the skin criterion is based on the assumption that the low-frequency loss is attributed to the effect of eddy currents. This assumption has to be validated.

Magnetostatic data may also be helpful for understanding the dispersion of high-frequency permeability. For example, the saturation magnetization determines integral magnetic loss in the material (Acher's law) [11]. This law is useful for estimating high-frequency performance of magnets [12,13].

Other information inferred from the magnetization curve may also be useful. In Ref. [13], the distribution function $P(H_k)$ for magnetic anisotropy fields derived from the measured initial magnetization curves has been suggested:

$$P(H_k) = -H\frac{d^2m}{dH^2} \quad (2)$$

where $m=M/M_0$ is the normalized magnetization,M_0 is the saturation magnetization,$H$is the external magnetic field. Eq. (2) is valid for thin magnetic films and with magnetization measurement made along hard magnetic axis. It is derived from the Stoner–Wohlfarth (SW) theory under assumption that the anisotropy fields in different regions of the material may have different magnitude though always have the same direction. Eq. (2) is useful for understanding high-frequency permeability ofthin ferromagnetic films [15].

In this paper, we will study the effect of eddy currents and of magnetic anisotropy dispersion on the permeability of composites containing Fe–Cu–Nb–Si–B (also named FINEMET alloy) microflakes. In flake particles, the magnetic structure is determined by the shape anisotropy and is the same as in thin films. Hence, the use of anisotropy field distribution is suggested for the analysis of permeability frequency dispersion in composites. It is known that the distribution in parameters of inclusions affects the properties of composites [16]. In composites, the anisotropy axes of inclusions are directed randomly, so the original approach seems to be not valid [14]. However, it is known that the anisotropy distribution may be also obtained from the magnetization curve when it is measured not along the hard axis [17]. In this case, the location of the distribution peak is about the same, though its amplitude and width may differ. Hence, the approach may be employed for randomly directed anisotropy fields. The anisotropy field distribution found from the magnetization curve is related to the distribution of the natural resonance frequencies and, further, to the frequency dependence of magnetic loss. The amplitude of the loss peak is obtained with the use of the relation between the saturation magnetization and integral magnetic loss [11].

2. Experimental details

The Fe-based alloy ribbons with a composition of $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ were produced with iron, copper, FeNb alloy, and FeB alloy as starting materials by melting in an induction furnace. The as-prepared ribbons were made by a melt-spinning technique. The as-prepared ribbons were then annealed at 540 °C for 1 h under argon atmosphere. In order to transform the ribbons into flaky shape, the annealed ribbons were respectively milled for 10 and 30 h with a ball milling procedure. The weight ratio of milling balls over ribbons was about 25.

X-ray diffraction (XRD) measurements were taken to examine the phases of ribbons. High resolution-transmission electron microscopy (HR-TEM, Model TECNAI-G^2 F20) was employed to study the nanostructure of the annealed samples. HR-TEM images and selected area electron diffraction (SAED) were used to study the size of nanograins and the formation of nano phases.

The Mössbauer spectroscopy experiments with transmission mode were conducted for the annealed ribbons. The radiation source was ^{57}Co embedded in an Rh matrix. The velocity of radiation source is calibrated with a standard α-Fe foil. The Mössbauer spectra were fitted using WinNormos-for-Igors® software. The high-frequency permeability spectra were measured within the frequency range of 0.5–10 GHz for composites containing the flakes and with paraffin as the matrix on a Vector Network Analyzer (Agilent 8720 ET) based on the transmission/reflection mode. The initial magnetization curves of the composites were taken on the Physical Properties Measurement System (PPMS, Quantum Design). The measurement samples were a thin layer of flakes containing between two sheets of foil.

3. Characterization of the nanocomposites

To find the nanocrystallization temperature for the as-prepared ribbons, the differential scanning calorimeter (DSC) study was made. Two exothermal peaks are observed, one at 530 °C (called T_1) and another at 672 °C (called T_2). Based on the

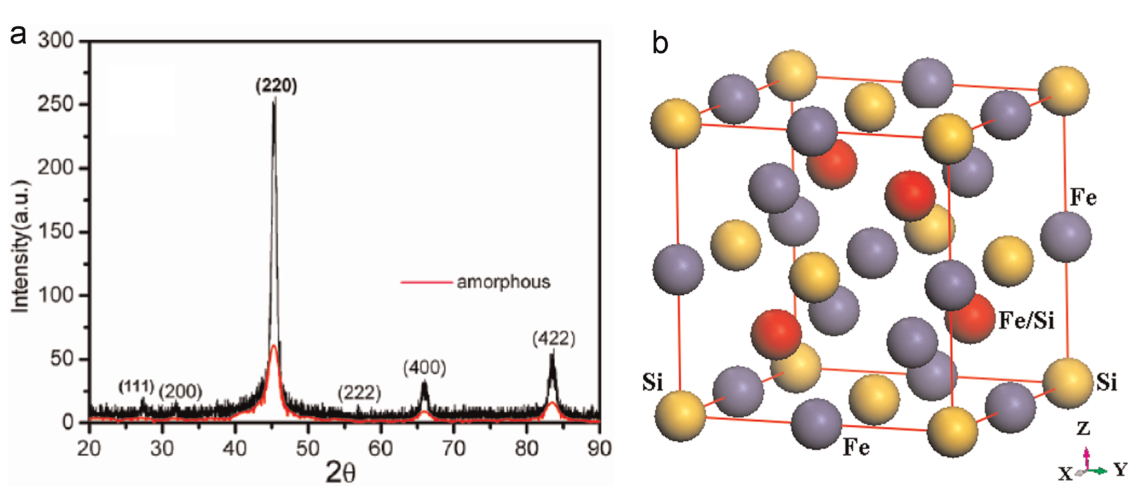

Fig. 1. (a) The XRD patterns of FINEMET ribbons; and (b) D03 superlattice.

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