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Study of high-frequency magnetic properties of Fe-Ti-B films obtained by magnetron sputtering

E N Sheftel¹, E V Harin¹, V A Tedzhetov^{1,*}, G Sh Usmanova¹, S Y Bobrovskii², K N Rozanov², P A Zezyulina² and Ph V Kiryukhantsev-Korneev³

¹ A A Baikov Institute of Metallurgy and Materials Science, Russian Academy of Sciences, Moscow, Russian Federation

² Institute for Theoretical and Applied Electromagnetics, Russian Academy of Sciences, Moscow, Russian Federation

³ National University of Science & Technology (MISIS), Moscow, Russian Federation * corresponding author, e-mail: vtedzhetov@imet.ac.ru

Abstract. The Fe-Ti-B films were obtained by dc magnetron sputtering of the Fe + 15% TiB₂ and Fe + 30% TiB₂ targets. The chemical composition and structure were studied in the assputtered state and after vacuum annealing at 500°C. The static (saturation magnetization M_s and coercive field H_c) and rf magnetic properties (magnetic permeability μ ', frequency of natural ferromagnetic resonance f_r and the frequency range in which μ' is kept) were determined.

1. Introduction

The main trends in modern magnetic microelectronics are miniaturization and performance, while ensuring efficient operation in the MHz and GHz frequency ranges of magnetic fields. Therefore, the development of new magnetic materials characterized by the properties that ensure the implementation of these trends is the most relevant task of materials science. The revolutionary changes in magnetic microelectronics that began in the mid-90s of the last century were largely initiated by experimental demonstration that nanocrystalline ferromagnets have unique magnetic properties [1]. This led to the development and the application of a number of Fe-based nanocrystalline alloys obtained by the quenching from the melt in the form of ribbons 10-50 µm thick [2].

At the same time, the scientific community showed interest in a new class of nanocrystalline soft magnetic alloys of Fe-Me-X systems (Me is one of the transition metals of IV group of the Periodic Table, X is one of the light elements N, C, O, B), obtained in the form of a film [3-5]. Particular interest in such films was shown in connection with the prospect of their use in miniature and highperformance audio- and video-equipment devices as magnetic cores of recording/playback heads [6]. For a new class of films, the physicochemical approach to the selection of chemical composition and structure formation conditions was first formulated in [7], which, as was shown later on the example of Fe-Zr-N films [8], provides a combination of high saturation magnetization M_s , low coercive field H_c and high hardness.

The approach consists in choosing a eutectic alloy in the Fe-MeX quasibinary system, producing films of this alloy in an amorphous or cluster state by magnetron sputtering, and a subsequent annealing, which forms a nanocrystalline structure in the films. The nanocrystalline structure should be represented by the main ferromagnetic Fe-based phase and the inclusions of the non-ferromagnetic thermodynamically-stable hard intercalation MeX phase (Fe/MeX nanocomposite structure).

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The described approach was applied to soft-magnetic Fe-based bulk alloys obtained by casting. As a result, alloys were developed, including alloy (Fe - 9.5% wt. Si - 5.5% wt. Al) - TiB₂, characterized by a complex of magnetic (high saturation magnetization and magnetic permeability in the MHz frequency range) and mechanical properties (hardness, wear resistance), not achieved on other bulk soft-magnetic alloys [9-10].

Planar technologies, in particular magnetron deposition, make it possible to obtain films of controlled thickness and integrate them into multilayer structures of an electronic device in one technological process, which greatly simplifies the manufacturing technology of a device and reduces its cost. The structure of the films is controlled by the magnetron deposition parameters and/or the subsequent annealing. As applied to Fe-Ti-B films, studies were started by us relatively recently [11].

The present work is devoted to the study of static (saturation magnetization M_s and coercive field H_c) and rf magnetic properties (magnetic permeability μ ', natural ferromagnetic resonance frequency f_r and frequency range in which μ ' is kept) of films with a two-phase structure of the Fe-TiB₂ nanocomposite type.

2. Objects and methods

The Fe-Ti-B films were prepared by the dc magnetron sputtering of composite targets in Ar atmosphere at a pressure of 0.2-0.5 Pa (other parameters of the deposition process are given in Table 1). Two different composite targets with TiB₂ ceramic chips, uniformly distributed within the target erosion zone, covering 15 or 30% of the target surface (Figure 1), were used. The TiB₂ chips were produced by self-propagating high-temperature synthesis. The time of deposition of the films was from 8 to 25 min. The glass (glass slide 1.5 mm thick and cover glass 0.13 mm thick) and Ti substrates were used.



Figure 1. Scheme of the $Fe + TiB_2$ targets.

| The area of TiB ₂ | U, V | I, A | Ar pressure, | Deposition time, | Fe-Ti-B film thickness, µm |
|------------------------------|------|------|--------------|------------------|----------------------------|
| on target, % | | | Ра | min | |
| 15 | 600 | 1.5 | | 20 | 2.4 |
| | | | 0.2 | 15 | 1.9 |
| | | | | 8 | 0.26 |
| 30 | | | 0.5 | 25 | 2.0 |
| | | | 0.4 | 20 | 1.57 |

Table 1. The deposition parameters of the Fe-Ti-B films under study.

The annealing of the films was performed at a 500°C temperature for 1 hour in a vacuum better than $2 \cdot 10^{-6}$ Torr. The annealing temperature was selected based on the previous studies [11].

The chemical composition, morphology, and film thickness were studied by EDX spectrometry using a Hitachi S3400N scanning electron microscope (SEM) with the Noran 7 Thermo Scientific attachment. The content of light elements (B, O, and N) was specified by the method of glow discharge optical emission spectroscopy (GDOES) using a Horiba Jobin Yvon Profiler-2 apparatus [12].

The phase-structural state of the films was studied by X-ray diffraction (XRD) on a Rigaku Ultima IV diffractometer equipped with a graphite monochromator in the Bragg-Bretano geometry using

CuK α -radiation. For quantitative phase analysis, a specialized software package [13] was used, that utilizes full-profile analysis according to the Rietveld method. The volume fractions of crystalline phases and their average grain sizes were determined. To estimate correctly the grain size, the physical broadening of the XRD peaks was analyzed taking into account not only the grain size *D*, but also the microstrains of the particles $\langle \varepsilon \rangle$ (we will not further consider microstrains). To reduce the error in determining the interplane distances Δd , the lattice parameters *a* and *c* were calculated from the position of the centers of gravity of the high-angle peaks using the Voigt approximation.

The static magnetic properties of the films were studied on a vibrating magnetometer. The rf magnetic permeability of the studied films was measured using an HP 8720C vector network analyzer in the frequency range 0.1-2 GHz [14].

3. Results and discussion

3.1. Phase composition and structure of the Fe-Ti-B films under study

According to the chemical analysis, the films in the as-sputtered state contain Fe, Ti, B, and O (the latter in an amount of not more than 1.5 at.%). None of other uncontrolled impurities (N, C) were found in these films. The distribution profiles of the elements obtained by the GDOES method (Figure 2a) indicate a uniform distribution of the elements over the thickness of the films. The films are characterized by a dense pore-free structure (Figure 2b). The film thicknesses, measured over cross sections using SEM (Figure 2b), are given in Table 1.



Figure 2. Distribution of chemical elements by thickness (a, GDOES) and microphotograph of the cross section (b, SEM) of the $Fe_{81.5}Ti_{13.1}B_{3.9}O_{1.5}$ films on the Ti alloy and glass substrates, respectively.

According to the XRD results (Table 2), in the Fe_{81.5}Ti_{13.1}B_{3.9}O_{1.5} films obtained by sputtering of the Fe + 15% TiB₂ target for 20 min, two-phase (bcc α Fe(Ti) + hcp TiB₂, Figure 3) nanocrystalline (grain sizes of 3.5 and 3.6 nm, respectively) structure is formed during the growth process with a volume phase ratio of ~13:1. The bcc phase formed in these films upon sputtering is a solid solution, most likely Ti in α Fe, as evidenced by the phase lattice parameter, which significantly exceeds that for pure α Fe (2.866 Å). The assumption that only Ti, and not B or O, is involved in the formation of a solid solution is based on the fact that this element has significant solubility in α Fe (up to 13.7 at.% at 882°C [15]). For B and O, the ultimate solubility does not exceed 0.001 and 0.1 at.%, respectively [15].

The Fe_{70.4}Ti_{8.8}B_{20.2}O_{0.6} films, obtained by sputtering of the Fe + 30% TiB₂ target for 25 min, are characterized by a single-phase α Fe-based amorphous (in terms of XRD) structure (Figure 3). Despite the presence of a significant amount of Ti and B, it is not possible to identify boride phases in these films.





The annealing at 500°C of the Fe_{81.5}Ti_{13.1}B_{3.9}O_{1.5} films leads to an increase of TiB₂ volume fraction up to 30 vol.% and to decrease of the bcc α Fe(Ti) phase lattice parameter which becomes equal to that of α Fe (2.866 Å). It means that depletion in Ti of the bcc solid solution phase carries out. Leaving the solid solution, Ti most likely interacts with B localized in the intergrain space (due to insolubility in α Fe), which leads to an increase in the volume fraction of the TiB₂ phase. At the same time, the average grain size of the bcc phase increases up to 10 nm, while for the TiB₂ phase it practically does not change.

The amorphous (in terms of XRD) state, formed in the $Fe_{70.4}Ti_{8.8}B_{20.2}O_{0.6}$ films under sputtering, is saved after annealing at 500°C.

| Deposition | Chem.composition | Annealing - | The results of the XRD analysis | | | |
|-------------------------|--|-------------|---------------------------------|----------------|-------|--------|
| parameters | of as-sput. films | | Phase [vol.%] | <i>a/c</i> , Å | D, nm | <ɛ>, % |
| Eo±150/ | | as- | bcc α Fe(Ti) [93] | 2.875 | 3.5 | 0.193 |
| T:D 20 | | sputtered | hcp TiB ₂ [7] | 3.014/3.290 | 3.6 | 0.601 |
| $11B_2, 20$ | Fe _{81.5} I I _{13.1} D _{3.9} O _{1.5} | 500°C | bcc αFe [69] | 2.866 | 10.1 | 0.506 |
| min | | | hcp $TiB_2[31]$ | 3.030/3.292 | 3.7 | 0.149 |
| Fe+30% TiB ₂ | | as-sputt. | XRD-amorphous | | | |
| 25 min | re _{70.4} 11 _{8.8} B _{20.2} O _{0.6} | 500°C | XRD-amorphous | | | |

Table 2. The results of the XRD analysis of the Fe-Ti-B films under study.

3.2. Static and rf magnetic properties of the Fe-Ti-B films under study

For the films of the Fe_{81.5}Ti_{13.1}B_{3.9}O_{1.5} composition obtained from the Fe + 15% TiB₂ target with a decrease in the deposition time from 20 to 8 minutes, the real magnetic permeability μ' increases from 4±2 to 130±50 (Table 3, Figure 4a). On Figure 4a, an uncharacteristic increase in the real magnetic permeability μ' is seen at frequencies above 1 GHz, when the imaginary part falls below zero. This may be due to the fact that the half-wave resonance in the measuring cell is superimposed on the ferromagnetic resonance of the sample, which is a purely measuring error. A decrease in the deposition time leads to a smaller film thickness; therefore, the substrate is less heated and the grain size is smaller. Table 3 also shows the frequency *f*, below which μ' is kept, and the frequency of the natural ferromagnetic permeability estimated from the hysteresis loop (Figure 5), $\mu = M_s/H_c$ [16], is consistent with μ' (Table 3).

The films of the Fe_{70.4}Ti_{8.8}B_{20.2}O_{0.6} composition obtained from the Fe + 30% TiB₂ target, with a decrease in the deposition time from 25 to 20 minutes, also show an increase in μ ' from 20±10 to 30±5 (Figure 4b). On Figure 4b, the imaginary permeability μ " curve shows two resonances at frequencies of 0.7 and 0.9 GHz, which is caused by two anisotropy fields [17], and they are visible on the hysteresis loop: coercive field and perpendicular anisotropy (Figure 5b). For the same deposition time (20 min), the magnetic permeability μ ' of the film from the Fe + 30% TiB₂ target is higher than from

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the Fe + 15% TiB₂ target (Table 3). Perhaps this is due to a single-phase XRD-amorphous structure in the first case. Measurements in two perpendicular directions in the film plane for all films showed very weak anisotropy (Figure 4).



Table 3. Static and rf magnetic properties of the Fe-Ti-B films under study

Figure 4. Frequency dependence of the magnetic permeability of the Fe-Ti-B films deposited from a target with 15 (a) and 30% (b) TiB₂ for 20 min. The real (μ ') and imaginary (μ '') magnetic permeabilities are measured in two perpendicular directions in the film plane.



Figure 5. The hysteresis loops of the films deposited from a target with 15 (a) and 30% (b) TiB_2 for 15 and 20 minutes, respectively. The second loop is measured in two perpendicular directions in the film plane.

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4. Summary

The Fe-Ti-B films with a thickness of 0.26 to 2.4 μ m were obtained for 8-25 min by dc magnetron sputtering of the Fe + 15% TiB₂ and Fe + 30% TiB₂ targets.

According to the results of chemical and phase analysis, in the $Fe_{81.5}Ti_{13.1}B_{3.9}O_{1.5}$ films obtained by sputtering of the Fe + 15% TiB₂ target for 20 min in the process of growth, two-phase (bcc $\alpha Fe(Ti)$ + hcp TiB₂) nanocrystalline (grain size about 4 nm) structure with a volume phase ratio of 93:7 is formed. The vacuum annealing at a temperature of 500°C led to an increase in the TiB₂ fraction up to 31%.

The Fe_{70.4}Ti_{8.8}B_{20.2}O_{0.6} films obtained by sputtering of the Fe + 30% TiB₂ target for 25 min are characterized by a single-phase α Fe-based XRD-amorphous structure. Despite the presence of a significant amount of Ti and B, it is not possible to identify boride phases in these films. The annealing at a temperature of 500°C did not lead to noticeable structural changes.

With a decrease in the deposition time of the Fe-Ti-B films (20, 15, 8 min and 25, 20 min for targets with 15 and 30% TiB₂, respectively), the real magnetic permeability μ' increases (from 4 to ~100 and from 20 to 30). For the same deposition time (20 min.), the magnetic permeability μ' of the film from the Fe + 30% TiB₂ target is higher than from the Fe + 15% TiB₂ target (30 and 4). The magnetic permeability estimated from the hysteresis loop, $\mu = M_s/H_c$, is consistent with μ' . The frequency f, below which the μ' value is kept, and the frequency of the natural ferromagnetic resonance f_r were determined. Measurements in two perpendicular directions in the film plane for all films showed very weak anisotropy.

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References

- [1] Yoshizawa Y, Oguma S and Yamauchi K 1988 J. Appl. Phys. 64 6044
- [2] McHenry M E and Laughlin D E 2000 *Acta Mater*. **48** 223
- [3] Viala B, Minor M K and Barnard J A 1996 J. Appl. Phys. 80 3941
- [4] Chakraborty A, Mountfield K R, Bellesis G H, Lambeth D N and Kryder M H 1996 J. Appl. Phys. 80 1012
- [5] Nago K, Sakakima H and Ihara K 1992 IEEE Transaction J. on Magn. in Japan. 7 119
- [6] Rask M T and Longworth L L 1991 US Patent 5001589A
- [7] Grigorovich V K and Sheftel' E N et al. 1995 Report on the contract IMET-Philips PLW-938018-D-WZ-86512
- [8] Sheftel E N 2010 Inorganic Materials: Applied Research 1 17
- [9] Bannykh O A, Sheftel' E N, Grigorovich V K, Strug R E, Mkrtumov A, Polyukhova I R and Evdokimov A V 1992 *Russian patent* 4775860/02 [In Russian]
- [10] Grigorovich V K, Sheftel' E N, Strug R E and Polyukhova I R 1993 *Izvestia Akademii nauk SSSR. Metally* No. 6, 173 [In Russian].
- [11] Sheftel E N, Tedzhetov V A, Harin E V, Kiryukhantsev-Korneev F V and Usmanova G Sh 2016 Physica Status Solidi C: Current topics in solid state physics 13 965
- [12] Kiryukhantsev-Korneev Ph V 2012 Protection of Metals and Physical Chemistry of Surfaces 48 585
- [13] Shelekhov E V and Sviridova T A 2000 Metal Science and Heat Treatment 42 309
- [14] Starostenko S N, Rozanov K N, Osipov A V 2008 J. Appl. Phys. 103 07E914
- [15] Lyakishev N P (Ed.) Phase diagrams of binary metallic systems (Moscow: Mashinostroenie) 1997 Vol 2 p 586-591; 1999 Vol 3 Book 1 p 724-726 [In Russian]
- [16] Dionne G F 2003 IEEE Transactions on Magnetics 39 3121
- [17] Zezyulina P A et al. 2016 IEEE Magnetics Letters 7 3705804