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PREPARATION AND STRUCTURE OF THIN Fe$_{1-x}$B$_x$ SPUTTER FILMS

F. Stobiecki, S. Schwarzl, T. Stobiecki and H. Hoffmann

Institut für Angewandte Physik, Universität Regensburg, 8400 Regensburg, R.F.A.
+ Institute of Molecular Physics of the Polish Academy of Sciences, Poznań, Poland
++ Solid State Physics Department Academy of Mining and Metallurgy, Kraków, Poland

Introduction

Amorphous metal-metalloid /M-TM/ alloys, including Fe$_{1-x}$B$_x$ alloys, obtained by chemical or rapid quenching methods have been subjected to intensive studies for more than ten years [1]. Recently, thin films of these alloys obtained usually by the sputtering technique, have won great interest [2-4].

As these alloys in an amorphous state exhibit some interesting magnetic properties numerous works are concerned with their structural studies - short range ordering. For Fe-B alloys the studies are performed by various methods: X-Ray diffraction [5], neutron diffraction [6] and Mössbauer technique [7].

This paper is concerned with some technological problems arising in obtaining the thin Fe$_{1-x}$B$_x$ films and delivers a description of their structure and crystallization process obtained by the studies performed with the use of an electron microscope.

Sample preparation

All the thin films studied were obtained by rf-sputtering under the following conditions:

- Background pressure = $10^{-6}$ Torr
- Argon pressure = $2\times10^{-2}$ Torr
- Cathode current = 100 mA
- Cathode dc bias = -700 V
- Anode dc bias = -60 V

As substrates served the freshly cleaved NaCl single crystals, for studies by a transmission electron microscope (TEM), and glasses when the coercive field was measured by Kerr effect.

The powder pressed target composed of Fe 80 B 20 at % was used for sputtering. Composition of the films was changed by a regular covering the target surface with pieces of Fe or B. The concentration of individual elements in a film was determined by microanalysis and Auger analysis.

Fig. 1 presents the dependence of the film composition on the degree of covering the target surface with the pieces of Fe or B.
The relation presented proves that under the applied conditions of sputtering a great difference appears between the composition of the powder target: \( x = 0.20 \) and the composition of the film: \( x = 0.60 \).

In case of films intended to be examined on TEM their thickness, controlled by Alpha - step, was 80 nm, and in case of those to be studied as to their magnetic properties the thickness was 80 and 400 nm.

The sputtering rate decreased with increasing B concentration in a film, according to the relation shown in Fig. 2.

\[
\text{Sputtering Rate} = \frac{1}{1 + (x - 0.20)^2}
\]

Fig.2. Dependence of the sputtering rate on the \( \text{Fe}_{1-x} \text{B}_x \) film composition.

The dependence of the sputtering rate on the film composition or on the degree of covering the target surface with the pieces of Fe or B together with the difference between the composition of the film and of the target result from a considerable difference in sputtering yield for Fe and B.

**Structural analysis**

Structural studies were performed applying a 200 kV transmission electron microscope made by JEOL. The bright field micrographs obtained show inhomogeneities with a period of 200 - 400 Å superposed by a faint structure with a period of 40 - 80 Å, for example for the \( \text{Fe}_{0.68} \text{B}_{0.32} \) film see Fig. 3a. The origin of these inhomogeneities is not definitely clarified. The possible origins are:

a/ compositional fluctuations /Fe and B rich clusters/,  
b/ columnar structure with voids,  
c/ surface roughness,

For all films as deposited and in the course of annealing the total intensity of electron scattering \( I(s) \) was registered on a photographic film and next, plotted with a microdensitometer. The films were annealed in an electron microscope under pressure of \( p < 10^{-5} \) Torr with an annealing rate of about 2 degree/min.

The \( I(s) \) dependences obtained allowed to conclude that by the sputtering technique the thin films of \( \text{Fe}_{1-x} \text{B}_x \) in an amorphous state can be obtained for \( 0.12 \leq x \leq 0.75 /\text{Fig.4a/}. \) In case the content of B is smaller e.g. \( x = 0.06 \) the as-deposited films are crystalline. The crystallization of the films caused by
Fig. 4. Microdensitograms of a Fe$_{1-x}$B$_x$ diffraction patterns, /a/ 0.12≤x≤0.75 as-deposited /amorphous state/, /b/ 0.12≤x≤0.75 after annealing and x=0.06. x=0 as-deposited /crystalline state/, /c/ x=0.32 for various annealing temperatures.

Annealing runs in steps, as it shown Fig. 4c. For example in the case of Fe$_{0.38}$B$_{0.32}$ thin film the process of annealing does not produce any apparent change in the I(s) dependence.

At the temperature of 730 K the crystallization starts what is evidenced by the formation of crystallities with a diameter between 100 and 200 Å /see Fig. 3b/ and by the changes in the I(s) course /see Fig.4c/.

The temperature at which the crystallization starts depends on the composition of the thin films in the following way:

for x=0.37 it reaches the greatest value and decreases for both smaller and greater x values /see Table/.

<table>
<thead>
<tr>
<th>Temperature of crystallization [K]</th>
<th>650</th>
<th>700</th>
<th>730</th>
<th>750</th>
<th>650</th>
<th>550</th>
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</tr>
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Studying the dependence of the coercive field on the annealing temperature it was found that for the amorphous films the field is low /Hc < 0.5 Oe/ and increases strongly at a temperature close to that at which the film crystallization starts.

The I(s) dependences obtained for all the amorphous thin films allow to establish definitely the occurrence of the α-Fe phase. For the systems of B concentration about 30 at % the Fe$_2$B phase can also be distinguished.

However, it is not possible to prove the existence of other phases as for example a metastable phase Fe$_3$B observed in ribbons [7,8].

Radial distribution function - RDF

Following the method as described by H. Hoffmann and R. Winkler [9] we corrected the measured intensities from the background due to inelastic scattering, whereas small angle scattering due to inhomogeneities was neglected. With this correction it was possible to obtain an interference function i(s) which gives /by Fourier transformation, a rather good estimate of the interatomic distances, we have:

\[ G(r) = 4\pi \rho_i r^2 = \int S_i(s) \sin(sr) dr \]

The reduced RDF obtained from the above formula for the Fe$_1$B$_x$ films in both amorphous and crystalline states, are presented in Fig.5 and compared with the RDF of the film obtained by sputtering a target of pure iron. As it follows from
Fig. 5. Reduced RDF's for Fe$_{1-x}$B$_x$ films, 
a/ 0.12 ≤ x ≤ 0.75 as-deposited, 
b/ 0.12 ≤ x ≤ 0.75 after annealing and 
  x=0.06, x=0 as-deposited.

the dependencies shown in Fig.5 the nearest neighbour distance for iron thin films
is 2.7 Å and decreases only about 5% with the B content increasing up to 60at%.
For greater B contents the nearest neighbour distance considerably decreases and
reaches 2.2 Å in case of 75at% B. This effect may be due to the fact that as the
scattering amplitude of B atoms is small, for the lower B concentrations the first
peak may correspond to Fe-Fe pairs. For high B concentration x=0.75 the nearest
neighbour distance may correspond to Fe-B pairs, in spite of a considerable diffe-
rence in the scattering amplitudes of B and Fe.

Conclusions
1. Amorphous thin films of Fe$_{1-x}$B$_x$ are 
obtainable within a wide range of 
  concentrations /0.12 ≤ x ≤ 0.75/.
2. The temperature at which crystallization 
of thin films starts depends on their 
  compositions. It reaches the greatest 
  value for films containing about 30at% 
  B and decreases both for smaller and 
  greater B concentrations.
3. The nearest neighbour distance is about 
  2.6 Å for films of B concentration de-
  fined by 0.12 ≤ x ≤ 0.60, corresponding 
  to Fe-Fe pairs, and for x = 0.75 this 
  distance is 2.2 Å corresponding to Fe-B 
  pairs.
4. A rapid increase in coercive field oc-
curs during annealing of films at a 
temperature close to that at which the 
crystallization starts.

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