Mössbauer effect studies of thin iron films synthesized by ion beam assisted deposition technique

A L Zinnatullin1, N M Lyadov1,2, I A Faizrakhmanov1,2 and F G Vagizov1

1 Kazan (Volga Region) Federal University, Kremlievskaya 18, 420008 Kazan, Russia
2 Zavoysky Physical-Technical Institute, Sibirsky Trakt 10/7, 420029 Kazan, Russia

Abstract. The paper presents the results of Mössbauer effect studies of thin iron films obtained by ion-assisted deposition technique. It was found that the films obtained consist of several phases and have a perpendicular magnetic anisotropy. Magnetization direction can be changed by external magnetic field in sample plane.

1. Introduction

To increase the storage capacity of magnetic memory, it is necessary to use new magnetic materials that have certain properties. Such materials should have high coercivity at room temperature. The size of the magnetic domain should be as small as possible. Finally, it is important that the materials have perpendicular magnetic anisotropy (PMA). This feature of magnetization gives us possibilities to extremely increase the density of domains and, consequently, to increase the storage capacity of magnetic memory [1].

Thin films which show perpendicular magnetic anisotropy have been studied in the 70s of the last century [2]. The first magnetic memory devices with perpendicular recording appeared in 2005 [3]. Such devices have a relatively big capacity, more performance then hard disks with longitudinal recording. Furthermore, materials with perpendicular magnetic anisotropy can be used in magneto-optical and spintronic devices [4].

The development of a method for the production of such films is a significant practical task. The key factors here are the reproducibility, control of the elemental and phase composition of the films, as well as the low cost of the materials produced. Films with perpendicular magnetic anisotropy can be produced by magnetron sputtering [5], chemical deposition from the gas phase [6], and electrolytic deposition [7]. Ion beam assisted deposition (IBAD) technique is of a great interest. Main feature of IBAD method is that films in unusual, metastable crystalline and amorphous states can be synthesized under the continuous action of low-energy ions (<1 keV) [8].

In this paper, we present the experimental results on investigations of thin iron films synthesized by ion beam assisted deposition. To analyze the phase composition and magnetic properties of the obtained samples, a gamma-resonance spectroscopy (Mössbauer spectroscopy of conversion electrons) was used.

2. Experimental part

Thin iron films on silicon and glass substrates, called as "Sample 1" and "Sample 2", respectively, were synthesized by ion beam assisted deposition technique. Xenon ions with energy of 0.7 keV...
sputtered the metallic iron target enriched with $^{57}$Fe isotope up to 60%. The xenon ions incident on the surface of the substrate at small angles caused amorphization of the synthesized layer and stimulated the growth of the iron clusters under conditions which are different from the thermodynamic equilibrium. The density of the ion current was about 200 $\mu$A/cm$^2$. Films were deposited for 20 minutes. The film thickness, estimated from the image of scanning electron microscopy ("Zeiss EVO 50 XVP"), was about 160 nm for the “Sample 1” (Figure 1).

Figure 1. Scanning electron microscopy image of iron thin film deposited on the glass substrate.

Mössbauer spectra of conversion electrons were measured at room temperature on a conventional constant-acceleration spectrometer (WissEl, Germany), equipped with a commercial source of gamma-radiation (Co-57 in the rhodium matrix, Retverc). To detect the conversion electrons, a gas flow detector, operating on a mixture of helium (He) and methane (CH$_4$, 5% vol.), was used. Spectrometer was calibrated using the metallic iron absorber at room temperature. The isomer shifts of the spectrum components were determined relatively to the center of gravity of the metallic iron spectrum.

3. Results and discussion
Conversion electron Mossbauer spectra of iron thin films at room temperature and the results of fitting are shown in Figure 2. A model of four components was used for the fitting. Mathematical processing revealed the presence of three sextets corresponding to magnetically ordered phases of iron atoms and one doublet that correspond to paramagnetic state of the iron.

Figure 2. Conversion electron Mossbauer spectra of thin iron films on silicon substrate without external magnetic field (a) and with magnetic field of 700 Oe (b); conversion electron Mossbauer spectra of thin iron film on glass substrate (c) without external magnetic field. Model spectra and components are shown. The solid lines represent the resulting fit curve and the components of the spectrum.
The hyperfine parameters of the spectrum components are given in Table 1. It should be noted that an asymmetric Gaussian distribution of hyperfine magnetic fields on resonant iron nuclei was assumed for more accurately approximation of spectra. Asymmetric distribution of hyperfine magnetic fields is typical for nanostructured materials [9].

Table 1. Hyperfine parameters* and relative content of the components.

|                  | IS, mm/sec | QS, mm/sec | HF, kOe | S, % | A₂  |
|------------------|-----------|------------|---------|------|-----|
| Sample 1         |           |            |         |      |     |
| Sextet-1         | 0.03      | -0.04      | 348     | 43.4 | 0.5 |
| Sextet-2         | 0.01      | 0.02       | 333     | 13.1 | 0.9 |
| Sextet-3         | 0.60      | 0.36       | 325     | 22.2 | 1.5 |
| Doublet          | 0.99      | 1.21       | -       | 20.2 | -   |
| Sample 1 in magnetic field of 700 Oe | | | | | |
| Sextet-1         | 0.03      | -0.07      | 346     | 47.3 | 1.15|
| Sextet-2         | 0.01      | 0.02       | 333     | 12.4 | 1.29|
| Sextet-3         | 0.56      | 0.28       | 315     | 22.5 | 1.26|
| Doublet          | 0.99      | 1.21       | -       | 17.7 | -   |
| Sample 2         |           |            |         |      |     |
| Sextet-1         | 0.02      | -0.05      | 347     | 42.4 | 0.5 |
| Sextet-2         | 0.02      | 0.02       | 332     | 13.7 | 0.8 |
| Sextet-3         | 0.52      | 0.31       | 313     | 25.3 | 1.0 |
| Doublet          | 0.99      | 1.25       | -       | 18.5 | -   |

*Estimate errors for isomer shift and quadrupole splitting are ~0.01 mm/sec, for hyperfine magnetic field ~1 kOe.

The hyperfine parameters of the sextet, indicated in Figure 2 as "Sextet-2", are close to the well known values of bulk metallic iron, α-Fe. However the parameters for other two sextets are slightly different relate to the parameters for bulk metallic iron and iron oxides. They can be attributed to small particles of metallic iron, as well as to amorphous iron [10]. The values of the doublet parameters are close to the values of the hyperfine parameters of iron (II) oxide.

Values of hyperfine parameters of components, called as Sextet – 2 on model spectra, close enough to the well known parameters for bulk metallic iron. However parameters for other two sextets are slightly different relate to the parameters for metallic iron and iron oxides. We assumed that these components correspond to the small metallic iron particles as well as to particles of amorphous iron [10]. Values of hyperfine parameters for the doublet are close to parameters for iron (II) oxide.

From the analysis of the Mössbauer spectra of iron films deposited on the silicon and glass substrate (Figure 2a, c) and the hyperfine parameters of the spectrum components (Table), it can be assumed that the substrate material has no significant effect on the phase composition of the resulting films under the same deposition conditions. The ratio of the intensity of the second (fifth) sextet lines relative to the intensity of the third (fourth) lines depends on the angle between the direction of the hyperfine magnetic field on the nucleus and the direction of the wave vector of the resonance photon:
\[ A_2 = \frac{4 \cdot \sin^2 \theta}{1 + \cos^2 \theta}, \]

where \( \theta \) is the angle between the direction of the magnetic field on the nucleus and the wave vector of the gamma-photon. Thus, this ratio gives us information about primary orientation of the magnetic moments [11]. In the case of random orientation of magnetic moments (isotropic case) this ratio should be equal to 2. As can be seen in Figure 2a, this ratio is different for our samples. Figure 2b shows the Mössbauer spectrum of the conversion electrons of the first sample placed in a longitudinal magnetic field with strength of 700 Oe. It can be seen that the intensities of the 2nd and 5th lines have increased noticeably.

In case of our measurements the wave-vector of gamma photons is perpendicular to the sample plane. So, as follow from above equation, such values of the ratio \( A_2 \) correspond to the case when the magnetization of the sample does not lie in the plane of the sample. An increasing of intensities of second and fifth lines might be connected with decrease of angle between the magnetization of sample and the film plane. For well-defined metallic iron sextet (Sextet-2) the angle \( \theta \) between the primary orientation of the magnetic moments and the wave vector of the gamma-photon changes from 23° to 34° for the sample in the magnetic field.

4. Conclusion
The results of Mössbauer effect studies show that the thin iron films synthesized by ion-assisted deposition technique have a pronounced perpendicular component of the magnetic anisotropy. The resultant magnetization of the sample makes an angle of 23° with respect to the plane normal and can vary with the application of an external magnetic field. It was found that the iron film consists mainly of alpha-iron and disordered iron. The nature of the observed anisotropy requires further study.

Acknowledgments
This work was partially funded by the Russian Foundation for Basic Research (Grant No. 18-02-00845-a), the Program of Competitive Growth of Kazan Federal University funded by the Russian Government.

References
[1] Tudu B. and Tiwari A. 2017 Vacuum 146, 329.
[2] Iwasaki S. and Nakamura Y. 1977 IEEE Trans. Magn. 13, 1272.
[3] Iwasaki S. 2009 Proc. Jpn. Acad. Ser. B Phys. Biol. Sci. 85, 37.
[4] Ikeda S, Miura K, Yamamoto H, Mizunuma K, Gan H D, Endo M, Kanai S, Hayakawa J, Matsukura F and Ohno H 2010 Nature Materials 9, 721.
[5] Wu D, Zhang Z, Li L, Zhang Z, Zhao H B, Wang J, Ma B and Jin Q Y 2015 Scientific Reports 5, 12352.
[6] Ootera Y, Shimada T, Kado M, Quinsat M, Morise H, Nakamura S and Kondo T 2015 Applied Physics Express 8, 113005.
[7] Zana I, Zangari G and Shamsuzzoha M 2005 Journal of Magnetism and Magnetic Materials 292, 266.
[8] Lyadov N M, Bazarov V V, Vagizov F G, Vakhitov I R, Dulov E N, Kashapov R N, Noskov A I, Khaibullin R I, Shustov V A and Faizrakhmanov I A 2016 Applied Surface Science 378, 114.
[9] Eibschütz M, Lines M E and Chen H S 1983 Physical Review B 28, 425.
[10] Long G J, Hautot D, Pankhurst Q A, Vandormael D, Grandjean F, Gaspard J P, Briois V, Hyeon T and Suslick K S 1998 Physical Review B 57, 10716.
[11] Wertheim G K 1964 Mossbauer Effect: Principles and Applications (New York: Academic Press).