

Physical properties of ion-beam mixed Fe/Si multilayered films

J. S. Park, C. O. Kim, Y. P. Lee*, Y. V. Kudryavtsev*, J. Dubowik**, B. Szymanski**, and J. Y. Rhee***

Department of Physics, Hanyang University, Seoul, 133-791, Korea

**Institute of Metal Phys., Nat'l Academy of Sci. of Ukraine, 36 Vernadsky str, 03142,
Kiev-142, Ukraine*

***Institute of Molecular Phys., Polish Academy of Sciences, 17 Smolukhowskiego 17, 60-179, Poznan, Poland*

****Department of Physics, Hoseo University, Asan, Choongnam, 336-795 Korea*

(Received October 10, 2001)

Abstract

We investigated physical properties of ion-beam mixed Fe/Si multilayered films(MLF) prepared by rf sputtering onto glass substrates at room temperature. Such an ion-beam treatment has led to noticeable changes in the structural and physical properties of the MLF; the formation of a new phase which is characterized by a crystalline silicide with a low coercivity and $T_C \approx 550$ K. In contrast to the as-prepared state, the ion-beam mixed MLF contains two magnetic phases. One of them is a very soft ($H_C < 2$ Oe), but microscopically homogeneous one with $M_{eff} = 6.7$ kG.

1. Introduction

The ion beam mixing(IBM) allows us to overcome either thermodynamic or kinetic barriers by employing the energetic particles, to surpass the limit of solid solubility and to achieve the compositional and structural metastability. For Fe/Si couple several publications were devoted to the study of iron-silicide formation caused by the IBM at a single Fe(Si)/Si(Fe) interface (*i.e.*, a thin Fe(or Si) film on a Si(or Fe) substrate) [1-5]. Irradiation by Ar^+ ions of a Si film deposited on Fe induces two phases; one of them is magnetically ordered and the other is paramagnetic [3]. The formation of a crystalline $B2$ -phase FeSi is also discussed [6-7]. This issue can also be realized in Fe/Si multilayered films(MLF). The purpose of this work is to comprehensively study the influence of ion-beam treatment on the structure and physical properties of Fe/Si MLF by using moderately surface-sensitive experimental tools. We used the low-angle and high-angle x-ray diffraction (LAXRD and HAXRD, respectively), vibrating-sample

magnetometry(VSM), ferromagnetic resonance(FMR), and magneto-optic(MO) and optical spectroscopies to clarify the silicide formation.

2. Experimental

Fe/Si MLF have been prepared by rf-sputtering onto glass substrates at room temperature(RT). The MLF has 50 bilayers composed of Fe and Si sublayers of 3.0 and 2.2 nm, respectively, in the nominal thickness. The topmost layer of the MLF was Fe. After the structural and physical properties of the as-deposited MLF had been investigated, the MLF were ion-beam mixed in a high vacuum ($\sim 1 \times 10^{-6}$ Torr) by Ar^+ ions directed normally to the free film surface at the following conditions; an ion energy of 80 keV, an ion flux of 1.5×10^{-6} A/cm², and ion dose of 1.5×10^{16} Ar⁺/cm². The structural characterization was performed by LAXRD and HAXRD. The magnetic property for MLF were measured at RT and elevated temperatures(up to 520 K) by using VSM and FMR. The details of the MO,

※ E-mail : yplee@hanyang.ac.kr

more specifically equatorial Kerr effect(EKE), and optical measurements can be found elsewhere [8]. In order to study the thermal stability of a structure formed by IBM the ion-beam mixed MLF were annealed at 720 K in a high vacuum and the physical properties were also studied.

3. Results and Discussion

Peak 3 in the HAXRD spectra of the as-deposited MLF(Fig. 1) can be attributed to the Fe(110) reflection. The ion-beam treatment has mixed the layers by about 50 - 60% of the total thickness(see below). Thus, the LAXRD spectrum is related to the unmixed part at the bottom. The enhancement and narrowing of peak 3 can be considered as an evidence for an improvement in the surface structure of MLF due to formation of a crystalline iron silicide at the top. The interplanar spacing related to peak 3 of the ion-beam mixed MLF is 0.1998 nm. Unfortunately, the HAXRD data do not allow us to conclude confidently what kind of silicide is formed at the top of the MLF after IBM, because several silicides have the most intense XRD lines

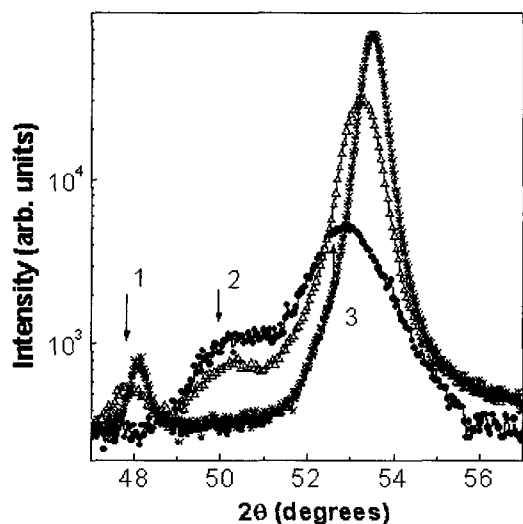


Fig. 1. High-angle XRD spectra for the as-deposited (circles), ion-beam mixed(triangles) and annealed [at 720 K] (asterisks)(3.0 nm Fe/2.2 nm Si)₅₀ MLF.

corresponding to nearly the same interplanar spacing [9]. On the other hand, peak 3 can also be related to the (110) reflection of the Fe-rich Fe-Si alloy, which can be formed as a result of IBM. It was shown by Fullerton *et al.* [10] that the HAXRD spectra for metallic MLF with the individual sublayer thicknesses of a few monolayers usually reveal only the diffraction peak corresponding to the weight-averaged lattice spacing of the constituent metals. Since the bottom layers were not mixed, the resultant HAXRD spectrum can be the averaged one from the “old” unmixed(at the bottom) and the “new” mixed(at the top) layers. However, the dominant contribution comes from the mixed layers.

An annealing of the ion-beam mixed MLF at 720 K leads to disappearance of the layered structure. The LAXRD as well as HARXD spectra do not exhibit satellites except a very tiny reflection around 3°. Peak 1 and 3 in the HAXRD spectra become more intense and narrower. Assuming that peak 1 and 3 after annealing belong to the same phase, we can suggest that these peaks are related to Fe₃Si₃.

The shape of $M(H)$ curve shown in Fig. 2 has changed drastically, indicating clearly an existence of at least two different magnetic phases in the ion-beam mixed state, *i.e.*, the old “hard” and new “soft” magnetic phases. The width of the second peak in the FMR spectra(not

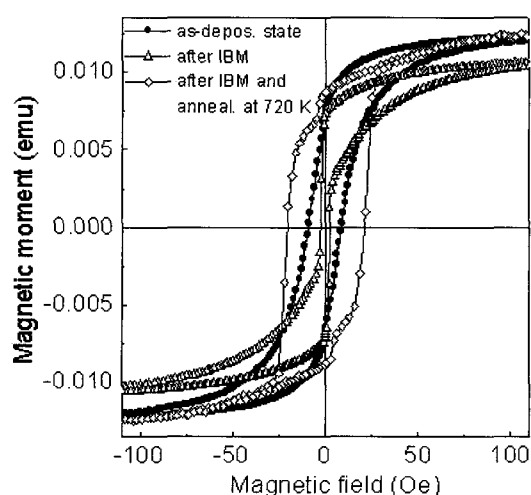


Fig. 2. Magnetic hysteresis loops of Fe/Si MLF.

shown here) is relatively narrow (about 40 Oe). This fact implies a small magnetic anisotropy as well as a high magnetic homogeneity of the ion-beam mixed part of sample. Furthermore, a low coercivity of the “soft” magnetic phase in the ion-beam mixed MLF also suggests the existence of a rather perfect crystalline structure.

The EKE spectrum of the as-deposited MLF is characterized by a feature at 1.7 eV [see Fig. 3(a)], which originates from the Fe sublayers [8]. The MO response of the as-deposited MLF is saturated at an external magnetic field of about 300 Oe. In the optical conductivity (OC, σ) spectrum of the as-deposited MLF a weak peak near 2 eV (marked by down arrow) is observed [see Fig. 3(b)]. The nature of this peak is closely related to the main absorption peak of Fe.

IBM alters noticeably the shape of EKE spectra and reduces the MO response in the 1.5 - 2.0 eV region. These changes point out a reduction of pure Fe content in the surface layers and can also be considered as a consequence of the modified electronic structure of the surface layers. Furthermore, from the AC magnetic-field dependence of EKE only the soft magnetic phase is detected; the MO response is fully saturated at 25 Oe. This fact is in an agreement with the magnetic

results.

The experimental hysteresis loops for the ion-beam mixed samples also indicate a combination of two uncoupled magnetic parts (see Fig. 2). By using a simple simulation method we can estimate that only about 60% of the MLF was mixed, i.e., the mixing affects only about 150 nm down the surface. The thickness of the mixed part is nearly equal to the Ar penetration depth [11]. Thus, the reaction in MLF does not reach the bottom layers. In our previous works the experimental MO properties of the as-deposited MLF were reasonably simulated with a model assuming that the actual bilayer structure in the MLF is Fe/FeSi [8]. IBM invokes the diffusion of the Fe atoms into the FeSi sublayers, forming $Fe_{1-x}Si_x$ ($0 < x < 0.5$) alloy regions and reducing the Fe content in the top layers (or the thickness of Fe sublayers).

For the case of full and homogeneous mixing the top layers of MLF should have a composition close to $Fe_{0.698}Si_{0.302}$. It is well known that in a thin-film alloy system some metastable phases can be formed. As can be seen in the phase diagram for bulk Fe-Si system [12], one can observe that $Fe_{0.70}Si_{0.30}$ alloy can be found in several structural states. The prominent enhancement of coercivity is due to appearance of a

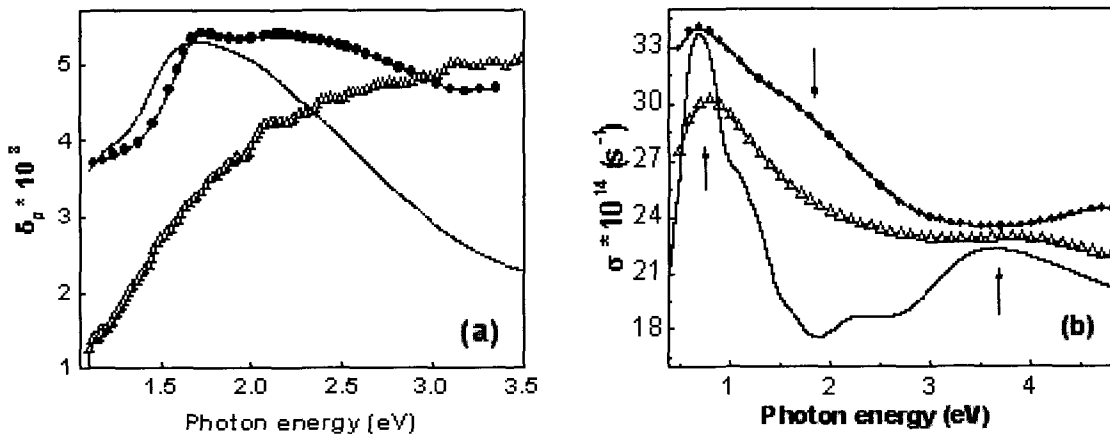


Fig. 3. (a) Equatorial-Kerr-effect and (b) optical-conductivity spectra for the as-deposited (circles) and ion-beam mixed (triangles) Fe/Si MLF. Pure Fe film (solid line) is included with a scaling factor of 0.5 in (a), and broadened and self-energy corrected optical-conductivity spectrum (solid line) for B2-phase Fe_2Si with a scaling factor of 0.83 is also shown in (b).

hard-magnetic Fe₅Si₃ phase [13].

Since, for the ion-beam mixed MLF, the changes in magnetic moment(m) are reversible for the thermal cycling up to 470 K, we may argue that the experimental data shown in Fig. 4 actually reflects the $m(T)$ dependence. The saturated magnetization can be expressed by the Weiss equation;

$$\frac{M_s}{M_0} = \tanh \left(\frac{\frac{M_s}{M_0} + \frac{H}{NM_0}}{\frac{T}{T_c}} \right) \quad (1)$$

where M_s and M_0 are the saturated magnetization at T and 0 K, respectively, H is an external magnetic field, and N is the molecular field constant [14]. In the calculations of the $M_s(T)/M_0$ dependence we used a saturated magnetic field of 200 Oe. The experimental temperature dependence of m can be reasonably described by Eq. (1) with a T_c of 550 K(see Fig. 4).

$m(T)$ for the ion-beam mixed MLF looks very similar to the $M_s(T)$ observed by Varga *et al.* [15] for a quenched Fe_{0.67}Si_{0.33} alloy(a metastable Fe₂Si B2-phase with a T_c of 508 K) and also by Yelsukov *et al.* [16] for Fe_{0.67}Si_{0.33} alloy prepared by mechanical grinding with a $T_c \approx 500$ K. T_c for Fe₅Si₃ and Fe₃Si phases(385 K [15] and 799 K [13], respectively), which are formed by the decomposition of Fe₂Si, are significantly different from that of Fe₂Si.

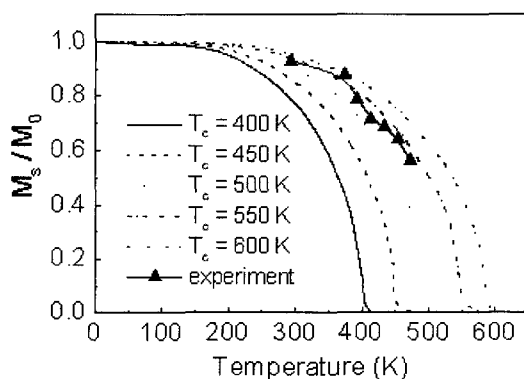


Fig. 4. Temperature dependence of the normalized magnetization for ion-beam mixed MLF.

In contrast to the aforementioned silicides, the calculated OC spectrum for the B2-phase Fe₂Si exhibits a reasonable resemblance to the experimental OC spectrum of MLF after IBM in shape and location of the absorption peaks, manifesting a formation of Fe₂Si after IBM. It is seen that both OC spectra exhibit an intense interband absorption peak located below 1 eV and also a weak peak near 4 eV [marked by up arrows in Fig. 3(b)].

Therefore, considering the overall results one can conclude that the ion-beam mixed Fe/Si MLF results in a formation of magnetically-soft metastable B2-phase Fe₂Si. A subsequent annealing at 720 K leads to decomposition of the Fe₂Si phase into magnetically-hard Fe₅Si₃ phase and, probably, Fe₃Si.

4. Summary

In summary, IBM destroys partially the layered structure of MLF and actually mixes only the surface layers down to a depth of about 110 - 150 nm. The bottom layers remain undisturbed and preserve the properties of the as-deposited Fe/Si MLF. By the optical and MO studies, it was concluded that IBM results in a formation of metastable B2-phase Fe₂Si. IBM induces a perfect crystalline silicide with a low coercivity and a T_c of about 550 K. An annealing of the ion-beam mixed Fe/Si MLF further destroys the layered structure at the bottom and also decomposes Fe₂Si to form a metastable magnetically-hard Fe₅Si₃ phase and, presumably, Fe₃Si.

Acknowledgments

This work was supported by Korea Research Foundation Grants(99-D00048 and 2001-015-DS0015) and by Hanyang University, Korea, made in the program year of 2001.

References

- [1] S. B. Ogale, R. Joshee, V. P. Godbole, S. M. Kanetkar, and V. G. Bride, *J. Appl. Phys.* **57**, 2915 (1985).

- [2] D. L. Santos, J. P. de Souza, L. Amaral, and H. Boudinov, *Nucl. Instrum. Methods*, B **103**, 56 (1995).
- [3] J. Jagielski, M. Kopciwicz, A. Turos, and F. Eichhorn, *Nucl. Instrum. Methods*, B **148**, 886 (1999).
- [4] N. P. Barradas, C. Jeynes, K. P. Homewood, B. J. Sealy, and M. Milosavljevic, *Instrum. Methods*, B **139**, 235 (1998).
- [5] M. Miloslavjevic, S. Dhar, P. Schaaf, N. Bibic, M. Nan, and K. P. Lieb, *Appl. Phys. A* **71**, 43 (2000).
- [6] E. G. Michel, *Appl. Surf. Sci.* **117-118**, 294 (1997).
- [7] M. Fanciulli, A. Zenkevich, and G. Weyer, *Appl. Surf. Sci.* **123-124**, 207 (1998).
- [8] Y. V. Kudryavtsev, V. V. Nemoshkalenko, Y. P. Lee, K. W. Kim, J. Y. Rhee, and J. Dubowik, *J. Appl. Phys.* **90**, (2001).
- [9] P. Veillars and L. D. Calvert, *Pearson's Handbook of Crystallographic Data for Intermetallic Phases* (American Society for Metals, 1985).
- [10] E. E. Fullerton, I. K. Schuller, H. Vanderstraeten, and Y. Bruynseraede, *Phys. Rev. B* **45**, 9292 (1992).
- [11] G. S. Chang, S. M. Jung, J. H. Song, H. B. Kim, J. J. Woo, D. H. Byun, and C. N. Whang, *Nucl. Instrum. Methods*, B **121**, 244 (1997).
- [12] *Binary Alloy Phase Diagrams*, edited by T. B. Massalski (American Society for Metals, 1986), Vol. I, p.1100.
- [13] L. K. Varga, F. Mazaleyerat, J. Kovac, and A. Kakay, *J. Magn. Magn. Mater.* **215-216**, 121 (2000).
- [14] R. M. Bozorth, *Ferromagnetism*(IEEE Press, New York, 1993), p.717.
- [15] G. Langer and P. Brown, *Philos. Mag.* **16**, 521 (1967).
- [16] E. P. Yelsukov, G. N. Konygin, E. V. Voronina, A. V. Korolyov, A. I. Ulyanov, S. K. Godovikov, and A. V. Zagainov, *J. Magn. Magn. Mater.* **214**, 258 (2000).