Modification of Fe/Cu multilayers under 400 keV Xe^{20+} irradiation*

WEI Kong-Fang(魏孔芳)[1,2] WANG Zhi-Guang(王志光)[1;1)] LIU Chun-Bao(刘纯宝)[1,2]
ZANG Hang(臧航)[1,2] YAO Cun-Feng(姚存峰)[1,2] SHENG Yan-Bin(盛彦斌)[1]
MA Yi-Zhun(马艺准)[1,2] SONG Yin(宋银)[1] LU Zi-Wei(卢子伟)[1]

1 (Institute of Modern Physics, Chinese Academy of Sciences, Lanzhou 730000, China)
2 (Graduate University of Chinese Academy of Sciences, Beijing 100049, China)

Abstract Two kinds of Fe/Cu multilayers with different modulation wavelength were deposited on cleaved Si(100) substrates and then irradiated at room temperature using 400 keV Xe^{20+} in a wide range of irradiation fluences. As a comparison, thermal annealing at 300—900 ℃ was also carried out in vacuum. Then the samples were analyzed by XRD and the evolution of crystallite structures induced by irradiation was investigated. The obtained XRD patterns showed that, with increase of the irradiation fluence, the peaks of Fe became weaker, the peaks related to Cu-based fcc solid solution and Fe-based bcc solid solution phase became visible and the former became strong gradually. This implied that the intermixing at the Fe/Cu interface induced by ion irradiation resulted in the formation of the new phases which could not be achieved by thermal annealing. The possible intermixing mechanism of Fe/Cu multilayers induced by energetic ion irradiation was briefly discussed.

Key words ion irradiation, Fe/Cu multilayers, Cu-based fcc solid solution

PACS 32.30.Rj, 61, 68.65.Ac, 34.35.+a

1 Introduction

The fcc-iron alloys show unique physical properties and attract considerable interests in physics and technology[1]. Some experiments verified that it can be got when iron alloyed with other fcc metals such as Ni, Pt, Cu, or Pd[2]. The Fe-Cu system, with positive heat of mixing of +19 kJ/mol, has extremely low miscibility at room temperature, and only ∼4% Fe could dissolve into fcc Cu and ∼10% Cu into bcc Fe near their respective melting points[3]. Mechanical alloying technique as a common method has been well used and got some progress in the fabrication of fcc Fe-Cu alloys in a wide alloying composition region[4—6]. Recently, energetic ion irradiation as a new nonequilibrium processing technique was proposed and used in the studies of intermixing, formation of amorphous or metastable phases of equilibrium immiscible bilayer or multilayer systems[7—11].

In the present work, intermixing and formation of new phases in Fe/Cu multilayers induced by energetic ion irradiation have been analyzed by X-ray diffraction (XRD). Both crystallite structure changes and intermixing of the layers corresponding to irradiation fluences during irradiation were observed. The structural evolution of the Fe/Cu multilayers during annealing was also investigated.

2 Experimental procedure

Multilayers with structure of Si/Fe(10 nm)/[Cu (4 nm)/Fe(4 nm)$]_5$/Cu(4 nm) and Si/Fe(10 nm)/[Cu (2.5 nm)/Fe(2.5 nm)$]_8$/Cu(5 nm) (the subscripts refer to layer numbers) were prepared by alternating depo-

Received 8 July 2008
* Supported by NSFC (10475102, 10125522) and Science Foundation of the Chinese Academy of Sciences
1) E-mail: zhgwang@impcas.ac.cn

sitions of pure iron (99.99% Fe) and copper (99.99% Cu) on cleaved Si(100) substrates by magnetron sputtering at room temperature (RT). In order to simplify the notation, these multilayers are hereafter referred to as $Cu_{2.5}/Fe_{2.5}$ and Cu_4/Fe_4. Every multilayer film was deposited on a 10 nm iron buffer layer and covered by a Cu protective layer. The base vacuum was less than 2.4×10^{-4} Pa and the vacuum during deposition was kept at about 0.68 Pa. The deposited thickness of each layer was monitored in-situ by a film thickness monitor.

The multilayers were irradiated at RT by 400 keV Xe^{20+} ions and the irradiation fluences ranged from 10^{14} to 10^{16} ions/cm^2. Some of the as-deposited multilayers were also annealed at 300 ℃, 600 ℃ and 900 ℃ in vacuum of 1.5×10^{-3} Pa for 2 hours. The XRD investigation of all the multilayer samples was performed on a Philips Expert Pro spectrometer under a glance angle of about 1 degree, and Cu K_α line was selected as the incident light.

3 Results and discussion

3.1 Structure of as-deposited multilayers

Figure 1 shows typical XRD patterns of as-deposited multilayers with equi-thick of Fe and Cu elemental layers, named (a) $Cu_{2.5}/Fe_{2.5}$, (b) Cu_4/Fe_4. The Fe (200), Fe (211) peaks were visible, but the principal peaks of Cu (111) and Fe (110) disappeared. This may be from that the grain size reduced when diminishing the modulation wavelength, which broadened the XRD peaks of Fe and Cu and caused the overlap of Cu (111) and Fe (110) peaks.

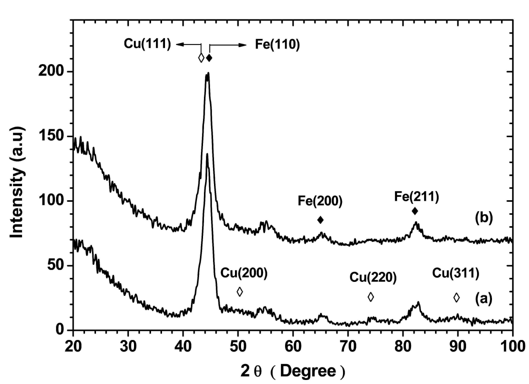

Fig. 1. XRD patterns of as-deposited Fe/Cu multilayers. (a) Si/Fe(10 nm)/[Cu(2.5 nm)/Fe (2.5 nm)]$_8$/Cu(5 nm); (b) Si/Fe(10 nm)/[Cu (4 nm) /Fe(4 nm)]$_5$/Cu(4 nm).

3.2 Modification of multilayers under irradiation

The structural evolution of multilayers was studied as function of irradiation fluences and the modulation wavelength. Fig. 2 shows the XRD patterns of Cu_4/Fe_4 sample. For the as-deposited sample, Fe (200) and Fe (211) peaks were observed. The strongest and broadening peak at $2\theta\sim44.375°$ may be caused by the overlapping of the principal peaks of Cu (111) and Fe (110). After 1×10^{14} ions/cm^{2}irradiation, two new peaks at $2\theta\sim43.915°$ and $2\theta\sim44.395°$ became visible, the Fe (200) and Fe (211) peaks became weaker and broadening. After 1×10^{16} ions/cm^{2}irradiation, the Cu (111), Cu (200), Cu (220), Cu (311) peaks became visible but shifted to lower angles and became narrow considerably, the Fe (200) and Fe (211) peaks became invisible and the Fe (110) peak at $2\theta\sim44.425°$ shifted to lower angles comparing with that of bulk Fe and became weak gradually, which showed that Cu-based fcc solid solution(Cu-fcc ss) and Fe-based bcc solid solution (Fe-bcc ss) phases appeared during irradiation. All these results indicated that the intermixing at Fe/Cu interface induced the formation of Cu-fcc ss and Fe-bcc ss phases during irradiation. XRD patterns of $Cu_{2.5}/Fe_{2.5}$ before and after irradiation are given in Fig.3. For the sample, two new peaks at $2\theta\sim43.165°$ and $2\theta\sim44.425°$ did not appear until the sample was irradiated at 1×10^{15} ions/cm^2.

It is interesting to compare with the mechanical alloyed Fe-Cu system. By ball milling for 160 hours, high Cu concentration fcc-like and low Cu concentration bcc-like Fe-Cu alloys could be obtained[12]. In our study, it was found that the structure of multilayers could transfer from bcc Fe and fcc Cu to Cu-fcc ss and Fe-bcc ss. With increasing the irradiation fluence, the XRD peak of Cu-fcc ss became strong and the peak of Fe-bcc ss became weak gradually, which indicated that the Cu concentration at the mixed region enhanced due to the intermixing of Fe and Cu layers. The intermixing induced by ion irradiation can be divided into two steps: atomic collision cascade step and consequent relaxation step. Atomic cascade, a process of far-from equilibrium state, alters the atomic mobility and enhances Fe and Cu interdiffusion to form a mixture phase of Fe and Cu with high formation energy. Controlled by thermodynamic and kinetic factors, the mixture phase produced by ion irradiation will relax and transfer towards to a stable phase. However, the relaxation period is extremely short lasting only for 10^{-10}—10^{-9}s, the phase transfer is not complete and thus a possible intermediate or metastable state could be obtained.

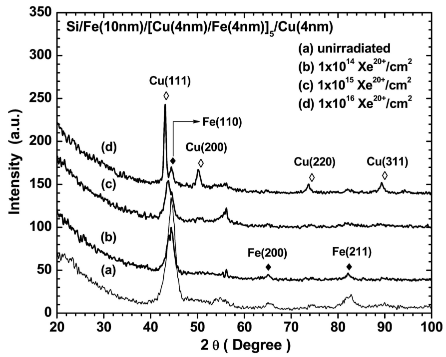

Fig. 2. XRD patterns of Si/Fe(10 nm)/[Cu(4 nm)/Fe(4 nm)]$_5$/Cu(4 nm).

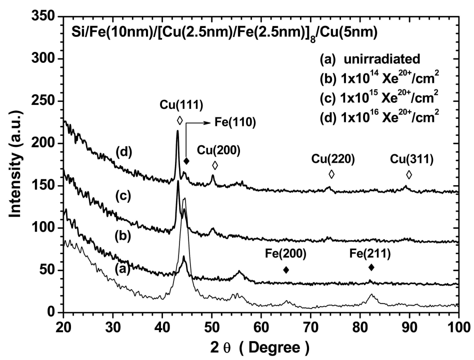

Fig. 3. XRD patterns of Si/Fe(10 nm)/[Cu(2.5 nm)/Fe(2.5 nm)]$_8$/Cu(5 nm).

3.3 Modification of multilayers after annealing

The obtained XRD results showed that, except for FeSi and $FeSi_2$, only a small fraction of Fe-bcc ss phase could be formed in the annealed samples. Fig. 4 gives the typical XRD patterns of Fe_4/Cu_4 multilayers after thermal annealing. It is clear that a new peak located at $2\theta \sim 44.56°$ (Fe-bcc ss) formed at annealing temperature up to 900 ℃. The $Cu_{2.5}/Fe_{2.5}$ sample shows a similar result to the Cu_4/Fe_4 sample.

From the comparison between the XRD results of ion irradiation and thermal annealing, it is evident that the ion irradiation resulted in the formation of new metastable phases that could not be achieved by the thermal annealing.

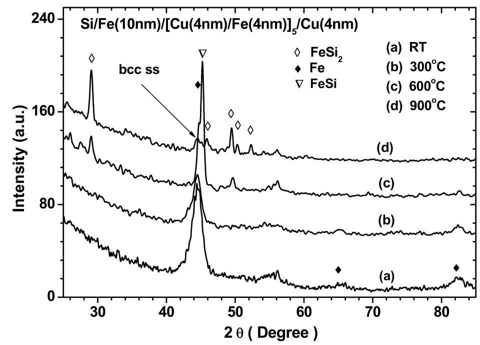

Fig. 4. XRD patterns of Si/Fe(10 nm)/[Cu(4 nm)/Fe(4 nm)]$_5$/Cu(4 nm) before and after annealing up to 300 ℃, 600 ℃ and 900 ℃.

4 Conclusions

The modification of Fe/Cu multilayers induced by 400 keV Xe^{20+} irradiation has been studied. After irradiation, the structure of multilayers gradually changed from bcc Fe and fcc Cu to Cu-based fcc solid solution and Fe-based bcc solid solution, which proved that ion irradiation induced intermixing of Fe/Cu multilayers. For the thermal annealed multilayers, Cu-based fcc solid solution phase was not observed. The structural evolution induced by ion irradiation was different from that induced by thermal annealing, which implied that collision cascades induced by ion irradiation altered the atomic mobility and then resulted in the formation of metastable new phase.

References

1 Pepperhoff W, Acet M. Constitution and Magnetism of Iron and its Alloys. Berlin: Springer-Verlag, 2001
2 Khmelevskyi S, Mohn P. Phys. Rev. B, 2005, **71**: 144423
3 Hansen M. Constitution of Binary Alloys. New York: McGraw-Hill, 1958
4 Yavari A R, Desre P J, Benameur T. Phys. Rev. Lett., 1992, **68**(14): 2235—2238
5 Harris V G, Kemner K M, Das B N et al. Phys. Rev. B, 1996, **54**(10): 6929—6940
6 LI T, LI Y Z, ZHANG Y H et al. Phys. Rev. B, 1995, **52** (2): 1120—1122
7 LIU B X, ZHANG Z J. J. Phys.: Condens. Matter, 1996, **8**: 165—171
8 CAI M, Veres T, Roorda S et al. J.Appl. Phys., 2004, **95**(4): 1996—2005
9 Amirthapandian S, Panigrahi B K, Srivastava A K et al. Nucl. Instrum. Methods B, 2003, **212**: 140—145
10 WEI L C, Averback R S. J. Appl. Phys., 1997, **81**(2): 613—623
11 LIU B X, LAI W S, ZHANG Q. Mater. Sci. Eng.R-Reports, 2000, **29**(1-2): 1—48
12 WEI S Q, YAN W S, LI Y Z et al. Physica B, 2001, **305**: 135—142