Directional effects of heavy-ion irradiation in Tb/Fe multilayers

J. Juraszek, A. Fnidiki, and J. Teillet
Groupe de Métallurgie Physique, UMR 6634 CNRS–Université de Rouen, F-76821, Mont Saint Aignan Cedex, France

M. Toulemonde
Laboratoire CIRIL, Boîte Postale 5133, F-14070 Caen Cedex 05, France

A. Michel
Laboratoire de Métallurgie Physique, Boîte Postale 179, F-86960 Chasseneuil–Futuroscope Cedex, France

W. Keune
Laboratorium für Angewandte Physik, Gerhard-Mercator-Universität Duisburg, D-47048 Duisburg, Germany
(Received 18 June 1999)

Deposition of a ^{57}Fe probe layer at the top of iron layers in Tb/Fe multilayers has allowed us to study locally the atomic mixing induced by high-energy heavy-ion irradiation at the Tb-on-Fe interface. The structural and magnetic transformations induced by ion irradiation of the probe layer were investigated by ^{57}Fe conversion electron Mössbauer spectrometry. To study directional effects of the ion beam, we have irradiated a stack of two pieces of the same multilayer set face to face. No change in the average composition of the mixed Tb-on-Fe interface was observed when the incident ion direction is changed from a Tb layer to a Fe layer, or from a Fe layer to a Tb layer.

I. INTRODUCTION

The total energy loss of an energetic ion penetrating into a solid includes the electronic energy loss $(dE/dx)_e$, resulting from interactions of the particle with the electronic subsystem of the target, and the nuclear energy loss $(dE/dx)_n$, due to elastic collisions with the target nuclei. The latter one is preponderant at low ion velocities, while $(dE/dx)_e$ strongly dominates $(dE/dx)_n$ at high ion velocities ($\geqslant$1 MeV/nucleon).[1] For heavy ions, very high rates of electronic energy deposition up to few tens of keV/nm are reached. Such a huge amount of electronic excitations is known to induce on a nanometer scale damage creation, phase transformation, and amorphous track formation in a wide range of materials, including insulators[2] as well as pure metals[3] or metallic compounds.[4] However, the transformation of the electronical energy into atomic motion is quite complex and the fundamental mechanisms are not well understood up to now. Nanometric multilayered systems, due to the multiplicity of surfaces and interfaces are a useful tool to study the mechanisms of interdiffusion and phase transformation induced in metallic systems by electronic energy deposition. A very strong mixing as a result of electronic excitations was detected in Ni/Ti bilayers,[5] Fe/Si,[6] and Tb/Fe multilayers[7] (ML's) above a $(dE/dx)_e$ threshold. In the case of a Ti/Ni/Ti trilayer, a strong disymmetry concerning the composition of the mixed layers was observed. The composition of the mixed Ni-on-Ti interface is nearly equiatomic while the mixed Ti-on-Ni interface is very rich in Ti.[8]

The aim of this work is to check if the interfacial mixing produced by the electronic slowing down of swift heavy ions in multilayered A/B materials is influenced by the relative position of the A and B layers with respect to the incident beam direction. A direct comparison of ion-beam-mixed A-on-B and B-on-A interfaces is not appropriate in this case because different initial structure of the interfaces is expected due to atomic size mismatch. The best way is to study with a monolayer resolution technique a sharp interface irradiated under normal incidence in the two opposite directions. Such study can be made by conversion electron Mössbauer spectrometry (CEMS) in iron-based multilayers by using a ^{57}Fe probe layer introduced in the near-interface region of the investigated system. Since the Mössbauer signal of a ^{57}Fe layer is approximately 50 times greater than one of natural iron, the ^{57}Fe Mössbauer spectrum of the sample would be characteristic of the position of the probe layer. This method was applied succesfully to evidence the asymmetry of the interfaces in the Tb/Fe multilayers,[9] in agreement with previous studies on bilayers[10] and with *in situ* resistivity measurements.[11] The Tb-on-Fe interface is sharp, while the Fe-on-Tb interface is amorphous and diffuse.[9] In this paper we present the CEMS study of heavy-ion irradiation of the Tb-on-Fe interface in nanometric Tb/Fe multilayers. Directional effects of the ion beam were investigated through the irradiation of a face-to-face set of specimens.

II. EXPERIMENTAL

The (3.3 nm Fe/0.5 nm ^{57}Fe/1.9 nm Tb) multilayers were grown at 150 K by thermal evaporation of pure Tb (99.99 at. %), ^{57}Fe (99.9 at. %), and natural Fe (99.99 at. %) in an ultrahigh vacuum system. The deposition of the 20 Fe/Tb bilayers was made onto a 150 μm thick Si (111) substrate [Fig. 1(a)]. All the Fe/Tb stack was sandwiched by a 5 nm thick SiO_2 film to avoid contamination and oxidation of the multilayer. During evaporation, the pressure was kept below 2×10^{-9} mbar. The deposition rate and film thickness were monitored by calibrated quartz microbalances located

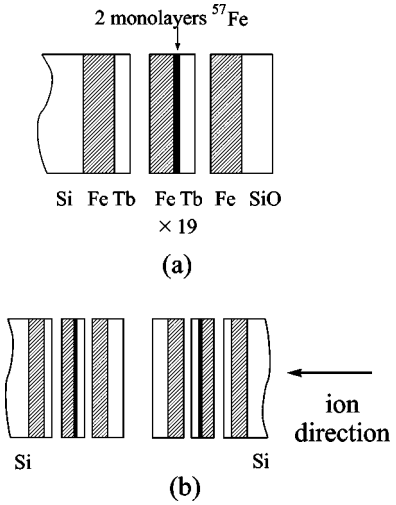

FIG. 1. (a) Description of the prepared samples with ^{57}Fe probe layer deposited at the top of natural iron layers. (b) Irradiation set-up: Two pieces of the same multilayer were put face-to-face and irradiated under normal incidence.

close to the substrate position. The deposition was held at a rate of 0.02 nm s^{-1} for Fe and 0.05 nm s^{-1} for Tb. The first and last Fe layers were only composed of natural iron, so that detection of the Fe/SiO_2 interfaces by Mössbauer spectrometry was negligible as compared to the ^{57}Fe probe layers signal.

The layered structure and the bilayer period were checked by grazing x-ray reflectometry. The crystallographic structure of the multilayer was investigated globally by high-angle x-ray diffraction (XRD) and locally by high-resolution transmission electron microscopy (HRTEM) using a JEOL 3010 transmission electron microscope. Cross-sectional views show many small Fe crystals in the metal layers, as expected because the layer thickness of 3.8 nm is beyond the critical crystallization thickness, which is estimated to be about 3.5 nm.[12] In contrast, only amorphous structures are visible in Tb layers and selected-area diffraction patterns show diffuse and broad rings for Tb. Because of the crystallization within the Fe layers, there is significant interface roughness.

Irradiation with heavy ions was performed at room temperature at the high-energy line IRABAT facility of the GANIL accelerator (Caen, France).[13] The beam flux was monitored by the secondary emission produced by the ion beam passing through a polarized 2.5 μm thick Ta foil, and calibrated with a Faraday cup. The relative uncertainty of the ion flux is estimated to be 20%. The multilayers were irradiated under normal incidence with 6 GeV ^{208}Pb ions at the fluence $\Phi t = 5\times10^{12}$ ions cm^{-2}. The beam flux Φ was kept below 3×10^{8} ions cm^2 s^{-1}, so that beam heating does not exceed a few tens degrees. The projected range, electronic and nuclear stopping powers were calculated using the TRIM code.[14] The values are reported in Table I. Due to the large projected range of 6 GeV ^{208}Pb ions in Si ($R_p = 319\,\mu$m), it was possible to irradiate a stack of two pieces of the same multilayers set face to face in order to compare the transformations of the probe layer irradiated with ions coming from Tb to Fe layers or from Fe to Tb layers. Before penetrating the first multilayer, an incoming ion has to cross the 150 μm thick Si top substrate, degrading its energy of approximately 3 GeV [Fig. 1(b)]. Then, the beam provides average values of $(dE/dx)_e$ inside the multilayers of $\sim$55.6 keV/nm in Fe and $\sim$42.3 keV/nm in Tb. It is noteworthy that both multilayers are entirely traversed by the beam and that implantation is made deeply into the second Si substrate. Thus, the ratio $(dE/dx)_e/(dE/dx)_n$ remains very large and the ion energy is mainly deposited in the multilayers by electronic excitations.

TABLE I. Main characteristics of the slowing down of GeV Pb ions in Si, Fe, and Tb targets calculated by the TRIM code (Ref. 14).

Ion	E (GeV)	Target	$(dE/dx)_e$ (keV/nm)	$(dE/dx)_n$ (keV/nm)	R_p (μm)
^{208}Pb	6.032	Si	15.6	8.9×10^{-2}	319
	3	Fe	55.6	4.8×10^{-2}	55.9
		Tb	42.3	3.7×10^{-2}	79.6

^{57}Fe conversion electron Mössbauer spectra were recorded at room temperature using a homemade helium/methane proportional counter with ^{57}Co in a Rhodium matrix as the source. During the Mössbauer analysis, the samples were set perpendicular to the γ beam. The fitting procedure used a least-square technique using the histogram method.[15] Isomer shifts are given with respect to standard α iron at 300 K. Direct information about the average Fe-spins orientation may be provided by comparing the relative line intensities $3:x:1:1:x:3$ of the six Mössbauer lines of a magnetically split sextet.[10] Indeed, the average ''cone-angle'' $\langle\beta\rangle$ between the γ-ray direction and the average direction of the magnetic hyperfine field B_{hf} is related to x by $x = 4\sin^2\langle\beta\rangle/(1+\cos^2\langle\beta\rangle)$.

III. RESULTS AND DISCUSSION

Each spectrum has been fitted with a distribution of hyperfine field $P(B_{hf})$ to take into account all the environments experienced by the ^{57}Fe nuclei, providing average values of the hyperfine parameters (Table II). The Mössbauer spectra of the as-deposited and ion-irradiated multilayers are shown in Fig. 2 and their corresponding hyperfine field distributions $P(B_{hf})$ are shown in Fig. 3. The Mössbauer spectrum of the as-deposited multilayer is composed of the superimposition of a sharp sextet and a broad magnetic component. The hy-

TABLE II. Parameters deduced from the fit of the Mössbauer spectra at 300 K for the Fe/^{57}Fe/Tb multilayers irradiated by GeV Pb ions from Tb to Fe layers (Tb→Fe) and from Fe to Tb layers (Fe→Tb). $\langle\delta\rangle$, $\langle B_{hf}\rangle$, $\langle\beta\rangle$ are, respectively, the mean isomer shift, the mean hyperfine field, and the average cone angle between the normal of the sample and the Fe spins direction. $A_{\alpha\text{-Fe}}$ is the relative fraction of Fe atoms which are in the bcc α-Fe phase compared to all Fe atoms.

Irradiation procedure	Fluence (ions cm^{-2})	$\langle\delta\rangle$ (mm s^{-1})	$\langle B_{hf}\rangle$ (T)	$\langle\beta\rangle$ (deg)	$A_{\alpha\text{-Fe}}$
	0	−0.018	27.2	64	0.59
Tb→Fe	5×10^{12}	−0.032	22.9	89	0.34
Fe→Tb	5×10^{12}	−0.033	22.6	89	0.33

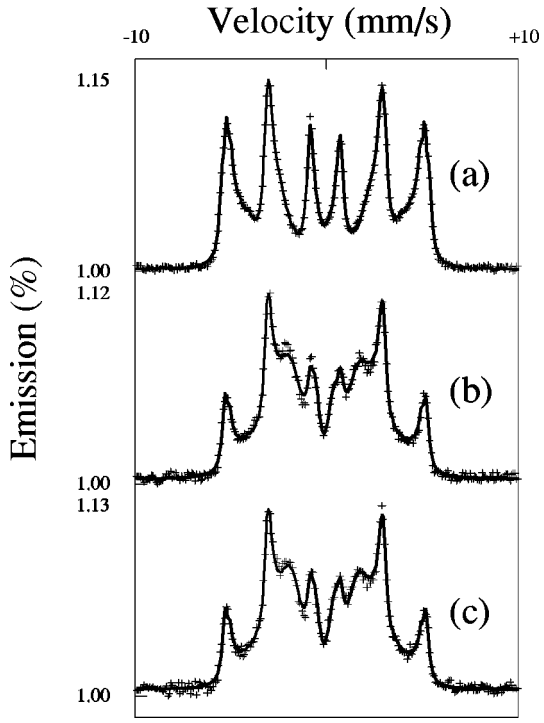

FIG. 2. Room-temperature Mössbauer spectra of the as-deposited sample (a) and Pb-ion irradiated at the fluence 5×10^{12} ion cm^{-2} with ion coming from Tb-to-Fe layers (b) or from Fe-to-Tb layers (c).

perfine parameters of the sharp sextet are close to those of bulk bcc-Fe values ($B_{hf}=33$ T and $\delta=0$ mm s^{-1}), evidencing for the crystallization of iron layers, in agreement with XRD and HRTEM results. The broad magnetic component comes from a distribution of Tb neighbors around Fe nuclei in a disordered structure due to a varying composition at the interface. The relative spectral area of the noncrystalline iron is 41% and the most probable hyperfine field, which defines the maximum of the distribution, is close to the bcc-Fe value, indicating a sharp interface.

Irradiation of the probe layer with 3 GeV ^{208}Pb ions coming from Tb to Fe layers results on the Mössbauer spectrum [Fig. 2(b)] in the decrease of the α-Fe sharp sextet spectral area (Table II) at the increase of the broad component intensity, indicating substantial ion-beam-mixing of the multilayer. The shape of the $P(B_{hf})$ distribution reflects the changes in the microstructure of the interfacial component

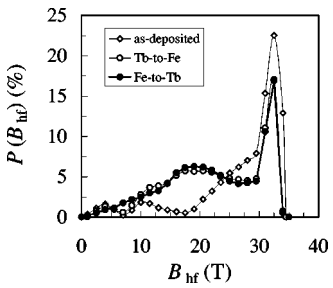

FIG. 3. Distributions of hyperfine fields $P(B_{hf})$ for the Tb-on-^{57}Fe interface: as-deposited ($\diamond$) and irradiated with Pb ions coming from Tb-to-Fe layers ($\circ$) or from Fe-to-Tb layers ($\bullet$).

(Fig. 3). It shows, regardless of the α-Fe peak centered at 33 T, a bell-like shape structure characteristic of an amorphous alloy, with a maximum centered at 18 T. Furthemore, the mean isomer shift $\langle\delta\rangle$ and the mean hyperfine field $\langle B_{hf}\rangle$ (Table II) evolve towards the values of the amorphous FeTb alloy [$\langle B_{hf}\rangle=18$ T and $\langle\delta\rangle=-0.08$ mm s^{-1}] with the same nominal Tb composition of the multilayer ($X_{Tb}=0.19$).[16,17] Thus, the loss of crystallinity of Fe layers is associated with the formation of an amorphous FeTb alloy, indicating a strong atomic mixing and amorphization induced at the interface in the vicinity of the ion path.[18]

Figure 2(c) presents the Mössbauer spectrum of the ^{57}Fe probe layer irradiated with ions coming from Fe to Tb layers. As previously, the spectrum exhibits a decrease of the α-Fe relative spectral area and a growth of a broad amorphous component, evidencing for a very efficient atomic mixing. Comparison of the two irradiated spectra [Figs. 2(b) and 2(c)] and corresponding hyperfine parameters (Table II) shows that (i) There is no visible change in the shape of the spectra and hyperfine fields distributions (Fig. 3), indicating that the structure of the mixed layer and the distribution of ^{57}Fe sites in the mixed zones are the same when the direction of the incident ions is changed.

(ii) The slight difference in the mean isomer shift $\langle\delta\rangle$ and mean hyperfine field $\langle B_{hf}\rangle$ values for the two irradiation configurations (Table II) remains in the limit of accuracy. Since these hyperfine parameters are known to be very sensitive to the local environment at the ^{57}Fe nuclei, the mean composition of the mixed layers is thought to be the same in the two cases.

The analysis of the Mössbauer spectra can also give informations about the ion-induced magnetic anisotropy modifications probed by the variation of the mean Mössbauer angle $\langle\beta\rangle$. The two samples show an increase of $\langle\beta\rangle$ towards 90°, evidencing for a reorientation of the magnetic anisotropy towards the film plane. It has been shown that the fluence evolution of $\langle\beta\rangle$ can be explained by assuming that mixed regions formed at the interfaces are surrounded by thermally relaxed zones in which the magnetic anisotropy turns into the plane.[18] In our case, the variation of the magnetic anisotropy is not affected by a change of the beam direction.

In a multilayered material, the succession of layers with different atomic species and with different sensitivities to dense electronic excitations leads to an inhomogeneous deposition of energy along the ion path. Our result shows that the Fe-Tb mixing induced by electronic excitations do not depend on a directional effect of the incident beam direction. According to Leguay *et al.*[8] irradiation of a Ti/Ni/Ti trilayer results in a strong dissymetry in the composition of the mixed Ti-on-Ni and Ni-on-Ti interfaces. In their case, the observed changes would result essentially from the initial different structural properties of both interfaces.

IV. CONCLUSION

The main conclusion of the paper is thus that the transformations of a ^{57}Fe probe layer deposited at the Tb-on-Fe interface do not depend on a directional effect of the incident ion. More precisely, when the ions travel the Tb-on-Fe interface from a terbium layer to an iron layer, or reversely from

an iron layer to a terbium layer, no measurable change in the structure and the composition of the mixed layer occurs. This experimental result suggests that the basic mechanism which allows to convert the energy transferred to the electronic subsystem into atomic motion leading to interfacial mixing is independent of the relative position of the different layers with respect to the incident beam direction.

ACKNOWLEDGMENTS

The authors are grateful to U. von Hörsten (Laboratorium für Angewandte Physik, Universität Duisburg, Germany) for providing the samples and to F. Levesque (CIRIL, Caen, France) for his efficient technical support during the irradiation experiment.

[1] E. Balanzat and S. Bouffard, Solid State Phenom. **30-31**, 7 (1993).
[2] M. Toulemonde, S. Bouffard, and F. Studer, Nucl. Instrum. Methods Phys. Res. B **91**, 108 (1994).
[3] Z.G. Wang, Ch. Dufour, E. Paumier, and M. Toulemonde, J. Phys.: Condens. Matter **10**, 9669 (1998).
[4] A. Audouard, E. Balanzat, S. Bouffard, J.C. Jousset, A. Chamberod, A. Dunlop, D. Lesueur, G. Fuchs, R. Spohr, J. Vetter, and L. Thomé, Phys. Rev. Lett. **65**, 875 (1990).
[5] R. Leguay, A. Dunlop, F. Dunstetter, N. Lorenzelli, A. Brasleau, F. Bridou, J. Corno, B. Pardo, J. Chevallier, C. Colliex, A. Menelle, and J.L. Rouvière, Nucl. Instrum. Methods Phys. Res. B **106**, 28 (1995).
[6] C. Dufour, Ph. Bauer, G. Marchal, J. Grilhée, C. Jaouen, J. Pacaud, and J.C. Jousset, Europhys. Lett. **21**, 671 (1993); Ph. Bauer, C. Dufour, C. Jaouen, G. Marchal, J. Pacaud, J. Grilhé, and J.C. Jousset, J. Appl. Phys. **81**, 116 (1997).
[7] J. Teillet, F. Richomme, A. Fnidiki, and M. Toulemonde, Phys. Rev. B **55**, 11 560 (1997).
[8] R. Leguay, A. Dunlop, F. Dunstetter, N. Lorenzelli, A. Brasleau, F. Bridou, J. Corno, B. Pardo, J. Chevallier, C. Colliex, A. Menelle, J.L. Rouvière, and L. Thomé, Nucl. Instrum. Methods Phys. Res. B **122**, 481 (1997).
[9] F. Richomme, B. Scholtz, R.A. Brandt, W. Keune, and J. Teillet, J. Magn. Magn. Mater. **156**, 195 (1996).
[10] B. Scholz, R.A. Brand, and W. Keune, Phys. Rev. B **50**, 2537 (1994).
[11] C. Dufour, K. Cherifi, A. Bruson, G. Marchal, and Ph. Mangin, Phys. Status Solidi A **125**, 561 (1991).
[12] F. Richomme (unpublished).
[13] S. Bouffard, J. Dural, F. Levesque, and J.M. Ramillon, Ann. Phys. (Paris) **4**, 395 (1989).
[14] J.P. Biersack and L.G. Haggmark, Nucl. Instrum. Methods Phys. Res. B **174**, 257 (1980).
[15] J. Teillet and F. Varret, MOSFIT program (unpublished).
[16] V.S. Rusakov, B.S. Vvedensky, S.N. Gadetsky, E.T. Voropaeva, V.V. Kochetov, A.V. Stupnov, and E.N. Nikolaev, J. Magn. Soc. Jpn. **17-S1**, 35 (1993).
[17] T. Ruckert, J. Tappert, R.A. Brand, and W. Keune, J. Magn. Magn. Mater. **165**, 411 (1997).
[18] J. Juraszek, A. Fnidiki, J. Teillet, F. Richomme, N.H. Duc, M. Toulemonde, and W. Keune, Appl. Phys. Lett. **74**, 2378 (1999).