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Magnetic texture in amorphous $Fe_{40}Ni_{35}Si_{10}B_{15}$ alloy irradiated with swift heavy ions*

M Kopcewicz[1] **and A Dunlop**[2]

[1] Institute of Electronic Materials Technology, Wólczyńska 133, 01-919 Warsaw, Poland
[2] Laboratoire des Solides Irradiés, Commissariat à l'Energie Atomique/Ecole Polytechnique, 91128 Palaiseau, France

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Abstract
The influence of swift-heavy-ion irradiation on the orientation of the hyperfine field in amorphous $Fe_{40}Ni_{35}Si_{10}B_{15}$ alloy is studied by Mössbauer spectroscopy. The change of the magnetic texture induced by 6 GeV Pb-ion irradiation is studied as a function of ion energy and the linear rate of electronic energy deposition for ion fluences ranging from 1×10^{11} to 2.4×10^{13} ions cm^{-2}. The Mössbauer measurements revealed that in a substantial volume fraction of the irradiated samples the spins changed their orientation from the in-plane orientation to the perpendicular one. This effect was attributed to the formation of cylinders of 'modified' amorphous structure along the ion path which may induce a stress as a result of which a substantial fraction of spins are aligned along the beam direction, i.e. perpendicular to the plane of the sample.

1. Introduction

The slowing down of high-energy heavy ions penetrating a target occurs via two nearly independent processes: (i) elastic collisions with nuclei, the most important mechanism at low ion energy, and (ii) electronic excitations and ionization, which strongly dominates at high ion energies (typically above 1 MeV/nucleon). The inelastic collisions with target electrons lead to high electronic excitations and ionizations and result in some modifications of the structure of the metallic system localized in the vicinity of the ion trajectory; such ion tracks could be identified by e.g. transmission electron microscopy. Along the path of GeV monatomic heavy ions or MeV fullerene clusters: (i) amorphous tracks were observed in crystalline metallic alloys such as $NiZr_2$ [1, 2], Ni_3B [3], NiTi [4]; (ii) defective crystalline tracks were found in Ti [5, 6], Zr [6], Fe [7]; and (iii) there was even a new crystalline phase detected in Ti [8]. It was suggested that in amorphous metallic alloys, such as $Pd_{80}B_{20}$, $Fe_{85}B_{15}$ and Ni_3B [9–12], high-electronic-energy deposition localized along the path of the energetic ions could lead to the formation of cylindrical zones of modified amorphous matter surrounding ion trajectories.

* The irradiations were performed using the GANIL accelerator, Caen (France).

The existence of such 'modified' amorphous cylinders was later confirmed by transmission electron microscopy observations [13, 14]. Since the formation of such tracks may induce local stresses in the amorphous structure, one could expect this to possibly affect some magnetic properties of soft ferromagnetic amorphous alloys, e.g., the orientation of the magnetization. Irradiation-induced anisotropy, which affects the direction of the magnetization, was observed in crystalline magnetic oxides ($Y_3Fe_5O_{12}$, $BaFe_{12}O_{19}$ [15] and $ZnFe_2O_4$ spinel [16]), but was not observed for ferromagnetic amorphous alloys.

The problem of magnetic texture in amorphous alloys and its dependence on stress was studied by a number of researchers and is well known in Mössbauer spectroscopy. For example, a stress-induced magnetic easy axis was observed in amorphous $Fe_{40}Ni_{40}P_{14}B_6$ alloy [17]. Internal stress [18] and sample curvature [19] influenced the magnetic texture in $Fe_{40}Ni_{38}Mo_4B_{18}$ alloy. Magnetoelastic effects related to surface crystallization of amorphous alloys affected the preferential spin alignment and indicated reorientation in the domain structure [20–23]. Magnetic texture was detected by Mössbauer spectroscopy in amorphous $Fe_{78}B_{13}Si_9$ alloy [24] and the influence of stress on the hyperfine-field distributions in amorphous $Fe_{81.5}B_{14.5}Si_4$ alloy was discussed [25]. The perpendicular anisotropy induced by thermal treatment was observed in amorphous $Fe_{82}B_{12}Si_6$ alloy [26] and a strange anisotropy was detected when a static magnetic field was applied perpendicular to the amorphous $Fe_{83}P_5C_{12}$ ribbon [27]. In most of these studies the stress-induced changes of preferential spin orientation were related to the magnetoelastic coupling which is magnetostriction. In some studies (e.g., [17]) a sophisticated Mössbauer polarimetry technique was used to analyse the spin texture. However, information regarding magnetic texture was usually obtained from the relative intensities of Mössbauer lines in the transmission or conversion-electron spectra of magnetically ordered materials which consisted of Zeeman sextets. The same, commonly used method is applied in the present study.

In this paper the change of the orientation of the magnetization (spin texture) in amorphous alloy induced by swift-heavy-ion irradiation was observed for the first time. This effect was detected for the amorphous $Fe_{40}Ni_{35}Si_{10}B_{15}$ alloy by means of Mössbauer spectroscopy.

2. Experiment

Amorphous $Fe_{40}Ni_{35}Si_{10}B_{15}$ ribbons, 10 mm wide and about 25 μm thick, were used as targets mounted in a liquid nitrogen cryostat and irradiated with 6 GeV Pb ions in the GANIL accelerator (Caen, France). A stack of four samples mounted one behind another, with the shiny side of the ribbon facing the beam, was irradiated with the ion beam at normal incidence under a controlled ion flux ($<5 \times 10^8$ ions cm^{-2} s^{-1}) in order to avoid excessive heating by the ion beam and to limit the sample temperature to 90 K. In front of the sample stack three foils (0.8 μm Al, 1 μm Ti and 0.8 μm Al) were placed for the determination of the ion fluence.

The ion energy after penetrating each sample in the stack and the electronic energy loss were calculated using the TRIM code [28]. The energies of the ions in the centre of the foils were respectively 5.38, 4.07, 2.62 and 1.07 GeV going from the first to the fourth sample in the stack. The corresponding linear rates of electronic energy deposition of Pb ions in the four samples increased gradually as the ions slowed down, and were respectively 41.4, 45.6, 51.2 and 54.4 keV nm^{-1}.

The samples were irradiated to fluences up to 1×10^{11}, 1×10^{12} and 2.4×10^{13} ions cm^{-2}.

All irradiated samples were characterized by Mössbauer spectroscopy in the transmission geometry. Additionally, conversion-electron Mössbauer spectroscopy (CEMS) was used for the study of near-surface (about 120 nm thick) regions of the samples. The Mössbauer measurements were performed at room temperature using a constant-acceleration spectrometer.

^{57}Co-in-Rh sources with activities of about 15 mCi for the transmission measurements and of about 25 mCi for the CEMS measurements were used. The Mössbauer spectra were fitted with the hyperfine-field distribution, $P(H)$, method. The constrained Hesse–Rübartsch method was used [29, 30]. The isomer shifts were relative to the α-Fe standard. Fitting of the spectra was performed using the NORMOS program [31].

3. Results and discussion

Mössbauer spectroscopy provides information on the orientation of the magnetic field in the sample with respect to the direction of the Mössbauer gamma radiation from the ratio of line intensities in the Zeeman sextet (see, e.g., [32–35]). In all Mössbauer measurements discussed here the γ-rays were perpendicular to the plane of the sample. In the general case the line intensity ratio in the six-line spectrum of the magnetically ordered sample, calculated taking into account the angular dependence of the allowed transitions in a pure nuclear Zeeman pattern, is 3:α:1:1:α:3, where

$$\alpha = \frac{4\sin^2\Theta}{1+\cos^2\Theta}. \tag{1}$$

(Θ is the angle between the direction of the hyperfine field, H_{hf}, and the direction of propagation of the gamma rays.) The parameter α varies from 0 to 4 depending on Θ. For instance, $\alpha = 0$ for $\Theta = 0^\circ$ (H_{hf} parallel to the γ-ray direction, i.e. spins are perpendicular to the plane of the sample), $\alpha = 4$ for $\Theta = 90^\circ$ (spins are aligned in the plane of the sample) and $\alpha = 2$ for random spin orientation.

The procedure used for fitting the Mössbauer spectra [29–31] allows the calculation of the α-parameter for the $P(H)$ distribution and hence the determination of the orientation of the magnetic field with respect to the plane of the sample. Changes of the spin texture due to swift-ion irradiation can thus be determined directly from the parameter α.

Since the ion energy and the electronic energy deposition were different for each sample in the stack it was possible to study the influence of the ion irradiation on the magnetic texture versus ion energy and the energy loss for various ion fluences. Typical Mössbauer spectra recorded for the nonirradiated amorphous $Fe_{40}Ni_{35}Si_{10}B_{15}$ sample and those irradiated with various ion fluences are shown in figures 1 and 2 for the first and the last sample in the stack, respectively. As can be seen from figure 1(a) the spectrum of the nonirradiated sample consists of the broadened sextet typical for ferromagnetic amorphous alloy. Slight asymmetry of the spectrum (the second line has somewhat larger intensity than the fifth line) was taken into account by assuming, as a first approximation, a linear correlation between the isomer shift and the hyperfine field. The parameter α resulting from the fit of the $P(H)$ distribution is about 3.60 ± 0.1, which strongly suggests a preferential (but not complete) spin alignment in the plane of the sample. At the highest effective ion energy (the first sample in the stack), irradiation with low ion fluence (1×10^{11} ions cm^{-2}) does not markedly change the spin texture (figure 1(b)). The parameter α is about 3.80 ± 0.1. The increase of ion fluence affects the spin orientation significantly. $\alpha \approx 2.3$ and 2.6 for ion fluences of 1×10^{12} ions cm^{-2} (figure 1(c)) and 2.4×10^{13} ions cm^{-2} (figure 1(d)), respectively, which strongly suggests that the average spin direction is tilted out of the sample plane. The $P(H)$ distributions (figures 1(a′)–1(d′)) remain almost unchanged. Similar behaviour was observed for the last sample in the stack which was irradiated with the lowest ion energy (of about 1.07 GeV) and in which the electronic energy deposition was the largest (about 54 keV nm^{-1}); see figure 2. Irradiation even with a low ion fluence (figure 2(b)) induces a noticeable change of the parameter α from 3.6 for the nonirradiated sample to 3.2. The increase of the ion fluence to 1×10^{12} ions cm^{-2} (figure 2(c))

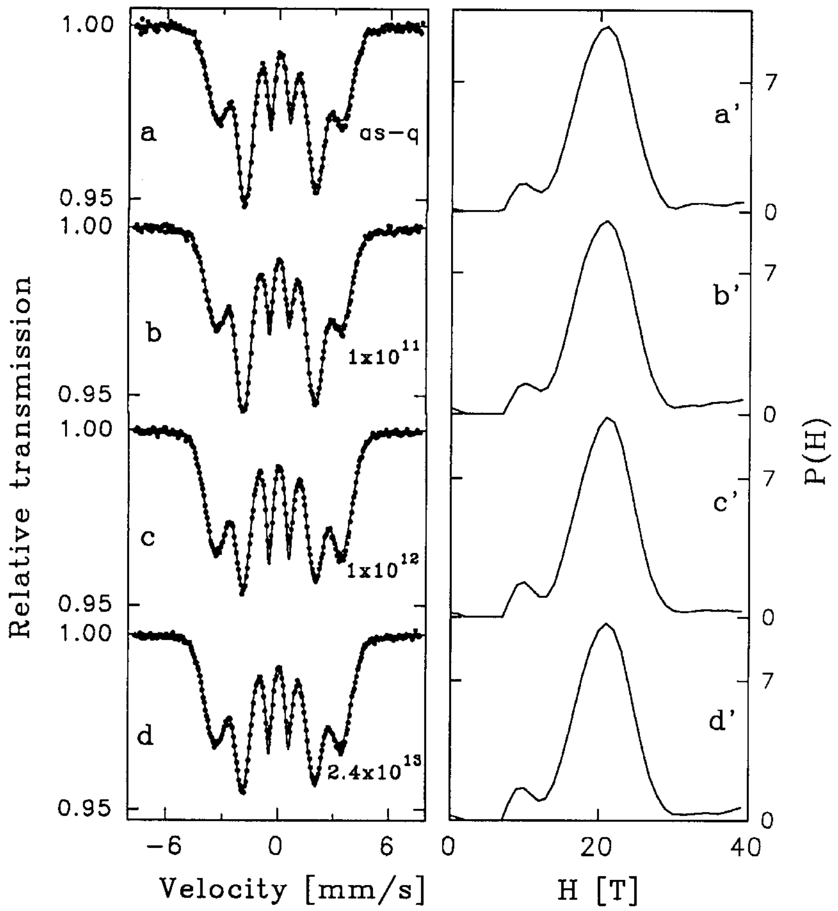

Figure 1. The Mössbauer spectra measured in transmission geometry for the nonirradiated amorphous $Fe_{40}Ni_{35}Si_{10}B_{15}$ sample (a) and an irradiated sample (first sample in the stack) with the ion fluences indicated ((b), (c), (d)) and the corresponding $P(H)$ distributions ((a′)–(d′)).

and 2.4×10^{13} ions cm^{-2} (figure 2(d)) caused a significant decrease of the parameter α to 1.8 and 2.1, respectively. Again, the $P(H)$ distributions remained almost unchanged.

The dependence of the parameter α on the position of the sample in the stack, i.e., on the ion energy and electronic energy deposition, is shown in figure 3 for the largest ion fluence of 2.4×10^{13} ions cm^{-2}. As can be seen, the parameter α decreases with decreasing ion energy and increase of the linear rate of the electronic energy deposition, from 3.6 for the nonirradiated sample to 2.6, 2.3, 2.0 and 2.1 for the consecutive samples in the stack (figures 3(b)–3(e)).

The observed change of the parameter α from 3.6, which evidences a strong spin alignment in the plane of the nonirradiated sample, to about 2 for samples irradiated with large ion fluence and with relatively low energy and high-electronic-energy deposition suggests either an irradiation-induced randomization of the spin orientations or that in a substantial volume fraction of the sample the spins changed their orientation from the in-plane orientation to the perpendicular one resulting in an average spin angle of about 55° and $\alpha \approx 2$. It seems that the second possibility is more realistic. Heavy-ion irradiation with normal-incidence geometry may result in the formation of cylinders of 'modified' amorphous matter directed perpendicular

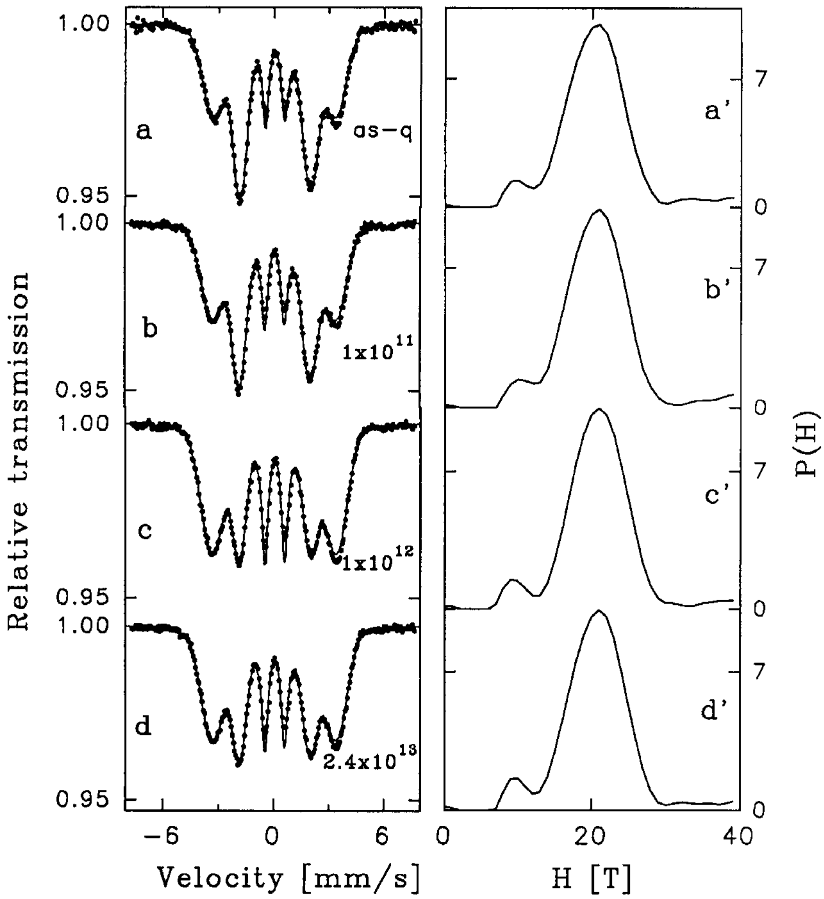

Figure 2. The Mössbauer spectra measured in transmission geometry for the nonirradiated amorphous $Fe_{40}Ni_{35}Si_{10}B_{15}$ sample (a) and an irradiated sample (fourth sample in the stack) with the ion fluences indicated ((b), (c), (d)) and the corresponding $P(H)$ distributions ((a′)–(d′)).

to the plane of the sample. Formation of such cylinders may induce stress in the amorphous structure, which may affect the spin texture, turning a substantial fraction of spins out of the sample plane to the direction parallel to the cylinders. As a result, the angle of spin orientation averaged over the entire volume of the sample may decrease from about 90° to about 55° leading to $\alpha \approx 2$, coincidentally a value corresponding to the random spin orientation.

A similar change of the direction of the hyperfine field due to swift-heavy-ion irradiation was observed earlier for crystalline $Y_3Fe_5O_{12}$, $BaFe_{12}O_{19}$ [15] and for the crystalline $ZnFe_2O_4$ spinel [16]. However, such an effect had not been observed previously for the amorphous alloy.

The change of the spin texture occurs mainly in the bulk of the sample as evidenced by the CEMS measurements performed for the same amorphous $Fe_{40}Ni_{35}Si_{10}B_{15}$ samples. Figure 4 shows, as an example, the CEMS spectra recorded for the nonirradiated amorphous $Fe_{40}Ni_{35}Si_{10}B_{15}$ sample and for four samples forming the stack irradiated with 2.4×10^{13} ions cm^{-2}. The parameter $\alpha \approx 3.9$ calculated from the fit of the CEMS spectrum in figure 4(a) for the surface region of the nonirradiated sample is larger than that corresponding to the bulk (figure 1(a)) which suggests that the spin alignment in the plane of the sample is stronger in the surface region than in the bulk.

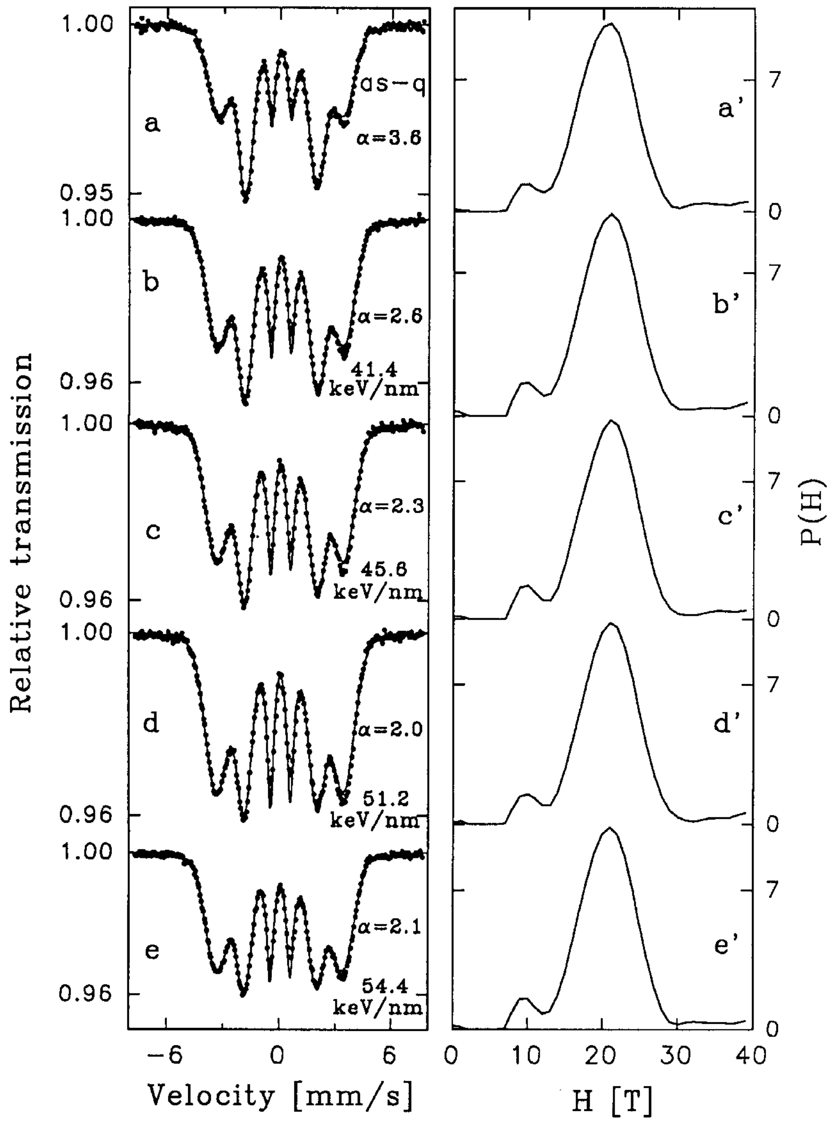

Figure 3. The Mössbauer spectra measured in transmission geometry for the nonirradiated amorphous $Fe_{40}Ni_{35}Si_{10}B_{15}$ sample (a) and for successive samples in the stack (with corresponding rates of energy deposition) irradiated with an ion fluence of 2.4×10^{13} ions cm^{-2} ((b), (c), (d), (e)) and the corresponding $P(H)$ distributions ((a′)–(e′)).

As can be seen from figure 4 the spin texture remains almost unchanged. The α-parameter decreases only slightly from 3.9 for the nonirradiated sample (figure 4(a)) to about 3.6 and 3.3 for the third and fourth samples in the stack (figures 4(d), 4(e)). The $P(H)$ distribution does not change significantly (figures 4(a′)–4(e′)). Similar behaviour was observed for all ion fluences. A much weaker change of the spin texture at the sample surface suggests that the surface anisotropy, responsible for the in-plane spin alignment, exceeds the stress-induced anisotropy, which tilts the spins out from the plane, related to the formation by the ion beam of the cylinders of ‘modified’ amorphous structure.

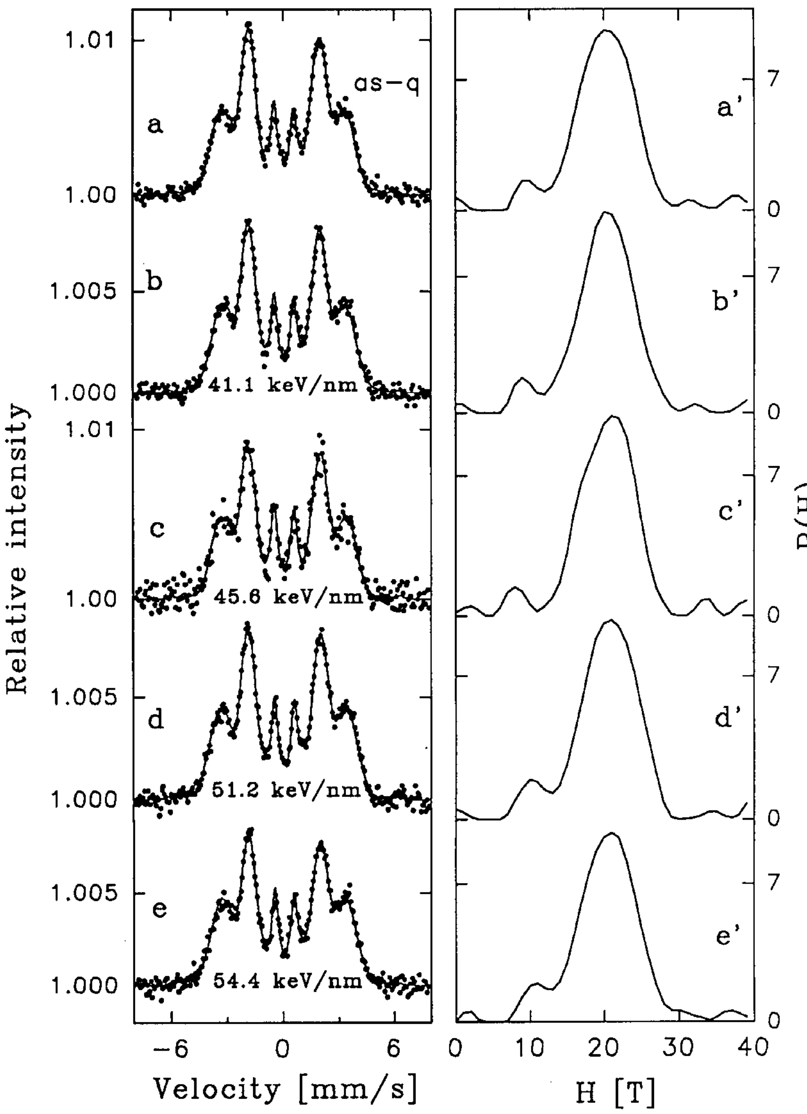

Figure 4. The conversion-electron Mössbauer spectra measured for the shiny side of the nonirradiated amorphous $Fe_{40}Ni_{35}Si_{10}B_{15}$ sample (a) and for successive samples in the stack (with corresponding rates of energy deposition) irradiated with an ion fluence of 2.4×10^{13} ions cm^{-2} ((b), (c), (d), (e)) and the corresponding $P(H)$ distributions ((a′)–(e′)).

4. Conclusions

The first evidence for the change of the orientation of the hyperfine field (and hence the magnetization) with respect to the plane of the sample induced in a ferromagnetic amorphous alloy by swift-heavy-ion irradiation is reported. The change of the spin texture, studied as a function of ion energy and the linear rate of electronic energy deposition for various ion fluences, was detected by Mössbauer spectroscopy. The change of the spin texture was attributed to the formation of cylinders of 'modified' amorphous structure due to high-electronic-energy deposition along the ion trajectory. Formation of such cylinders may induce a stress which may tilt a substantial fraction of spins out from the in-plane alignment to the direction parallel to the beam, i.e., perpendicular to the plane of the sample. It was found that the change of the spin texture is much larger in the bulk of the sample than at the sample surface.

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