# **Direct evidence of laser-induced magnetic domain structures in metallic glasses**

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The metallic glasses  $Fe_{81}B_{13.5}Si_{3.5}C_2$  ( $\lambda_s=27$  ppm) and  $Fe_{40}Ni_{40}Mo_{4}B_{16}$  ( $\lambda_s=12$  ppm) were exposed to pulsed excimer laser irradiation ( $\lambda = 248$  nm,  $\tau = 8$  ns) with different numbers of applied laser pulses per spot  $(N=2, 5,$  and 10). Transmission and conversion electron Mössbauer spectroscopies were used to infer the dependence of bulk and surface magnetic textures on laser processing parameters. Higher magnetostriction samples were found to develop an out-of-plane magnetic anisotropy while having the surface magnetic texture unchanged. Lower magnetostriction samples were found to acquire a random orientation of magnetic moment directions as a function of the number of applied laser pulses. Magnetic force microscopy determinations provided direct evidence of the occurrence of spectacular submicron magnetic domain structures in the higher magnetostriction samples. Moreover, our investigations showed that no magnetic structures are induced in the laser-irradiated samples having lower magnetostriction values.

## **I. INTRODUCTION**

Metallic glasses such as  $Fe_{81}B_{13.5}Si_{3.5}C_2$  and  $Fe_{40}Ni_{40}Mo_{4}B_{16}$  are commonly available in the form of ribbons produced by rapid quenching from the melt. Cw laser annealing can be used to promote structural relaxation or crystallization in the amorphous ferromagnetic ribbons. $1-3$ Improved thermal stability and different crystallization kinetics have been shown to characterize the effect of ultrashort pulsed-laser annealing in several binary amorphous alloys.4 In addition, different stress distributions can be obtained in metallic glasses with negative magnetostriction, as a result of inhomogeneous heat flows produced by local laser annealing.<sup>5</sup>

To date, pulsed excimer lasers have been extensively used to irradiate semiconductor materials,<sup>6</sup> but they are expected to play an important role in laser processing of amorphous magnets as well. Recently, changes in the magnetic anisotropy of Fe-B amorphous alloys have been induced without onset of crystallization through the use of multipulseexcimer-laser irradiation.<sup>7-10</sup>

In the present work, samples of metallic glasses with high and low magnetostriction have been exposed to pulsedexcimer-laser processing with different numbers of applied laser pulses per spot. Mossbauer spectroscopy was used to yield the bulk and surface magnetic textures as functions of laser irradiation parameters. Here, magnetic force  $microscopy<sup>11</sup>$  (MFM) provided direct evidence of laserinduced magnetic domain structure formation in the higher magnetostrictive systems only.

#### **II. EXPERIMENT**

Amorphous alloys  $Fe_{81}B_{13.5}Si_{3.5}C_2$  (Metglas 2605SC) and  $Fe_{40}Ni_{40}Mo_{4}B_{16}$  (Metglas 2826MB) were supplied by Allied Signal Inc. in the form of  $25-\mu m$ -thick ribbons. Their magnetostriction constants had the values  $\lambda$ <sub>s</sub>=27 and 12 ppm, respectively. Square samples  $(2\times2$  cm) were cut from foils and exposed on the shiny side to the  $\lambda = 248$  nm radiation generated by a KrF excimer laser (Lambda Physik), having a pulse width at half maxima  $\tau=8$  ns. Pulse energies of 450 mJ/pulse were employed, and the samples were irradiated with  $N=2$ , 5, and 10 laser pulses/spot at a repetition rate of 10 Hz. An acceptable degree of radiation homogeneity was



FIG. 1. Room-temperature Mössbauer spectra of  $Fe_{81}B_{13.5}Si_{3.5}C_2$  samples (a) in the amorphous as-quenched state, (b) after irradiation with 2 (laser pulses)/spot, and  $(c)$  after irradiation with 5 (laser pulses)/spot.



FIG. 2. Room-temperature Mössbauer spectra of  $Fe_{40}Ni_{40}Mo_{4}B_{16}$  samples (a) in the amorphous as-quenched state, (b) after irradiation with 5 (laser pulses)/spot, and  $(c)$  after irradiation with  $10$  (laser pulses)/spot.

obtained by laser beam scanning of the whole samples surface.

Room-temperature transmission Mössbauer measurements were made with a constant acceleration spectrometer (Ranger Scientific). The 25-mCi gamma ray source was  ${}^{57}Co$ in a Rh matrix, maintained at room temperature. Transmission spectra of as-received and laser-irradiated samples were collected with incident gamma rays perpendicular to the ribbon plane. Amorphous specimens were analyzed with the NORMOS-DIST program.<sup>12</sup> Conversion electron Mössbauer spectroscopy (CEMS) spectra of as-cast and laser-treated surfaces were collected using a flowing  $He-CH<sub>4</sub>$  electron counter. The backscattered electrons into  $2\pi$  solid angles were recorded, so that the behavior of the surface layers of 100 nm could be detailed.

Magnetic force microscopy was performed on metallic glasses. We used a Digital instrument able to discriminate between the topographic and magnetic surface features. Both higher- and lower-magnetostriction systems were investigated.



FIG. 3. Conversion electron Mössbauer spectra recorded on the  $Fe_{81}B_{13.5}Si_{3.5}C_2$  system (a) in the amorphous as-quenched state and  $(b)$  after irradiation with 5 (laser pulses)/spot.



FIG. 4. Dependence of the magnetic texture parameter  $R_{21}$  on the number of applied laser pulses: circles for higher magnetostriction samples, squares for lower magnetostriction samples, and triangles for the surface magnetic texture.



FIG. 5. Magnetic force micrographs of the  $Fe_{81}B_{13.5}Si_{3.5}C_2$  system after irradiation with 5 (laser pulses)/spot. The left side corresponds to the topographical features.



FIG. 6. Magnetic force micrographs of the  $Fe_{81}B_{13.5}Si_3.5C_2$  system after irradiation with 10 (laser pulses)/spot. The left side corresponds to the topographical features.

## **III. RESULTS AND DISCUSSION**

Room-temperature transmission Mössbauer spectra of the Metglas 2605SC and 2826MB samples in the amorphous as-quenched state are shown in Figs.  $1(a)$  and  $2(a)$ , respectively. For the 14.4-keV  $\gamma$  rays of <sup>57</sup>Fe, the relative intensity of the second  $(fifth)$  to the first  $(sixth)$  lines is given, in the thin absorber approximation, by  $R_{21} = 4 \sin^2 \alpha / [3(1$  $(\cos^2 \alpha)$ , where  $\alpha$  is the angle between the  $\gamma$ -ray propagation direction and the direction of the magnetic hyperfine moment. The ratio  $R_{21}$  varies from 0 to 4/3 as  $\alpha$  changes from 0 to 90° and, for a completely random distribution of magnetic moment directions, takes the value 0.67.

Figures 1(b) and 1(c) show the transmission Mössbauer spectra of  $Fe_{81}B_{13.5}Si_{3.5}C_2$  samples after irradiation with 2 and 5 laser pulses/spot. It can be seen that the in-plane orientation of the easy magnetization direction was changed, due to the laser treatment performed, to an out-of-plane orientation. As inferred from the plot in Fig. 4, below, the preferred out-of-plane orientation induced in the highermagnetostriction samples is preserved through the entire laser treatment.

Figures  $2(b)$  and  $2(c)$  display the transmission Mössbauer spectra of  $Fe_{40}Ni_{40}Mo_{4}B_{16}$  specimens after irradiation with 5 and 10 laser pulses/spot. As shown by the  $R_{21}$  values in Fig. 4, middle, the laser irradiation treatment resulted in a random orientation of the magnetic moment directions, for all values of *N* investigated. This result can be correlated with the lower value of the saturation magnetostriction constant in this system.

Figure 3 shows selected CEMS spectra of amorphous  $(a)$ and laser-irradiated samples (b). As can be seen in Fig. 4, above, the original in-plane orientation is preserved by the surface magnetic texture during the irradiation treatment performed. This result demonstrates the existence of a distribution of magnetic moment orientations through the thickness of the foil. The three types of kinetics (out-of-plane for higher magnetostriction samples, random for lower magnetostriction samples, and in-plane for the surface magnetic texture) are summarized and presented in Fig. 4.

Figures 5 and 6 show the MFM micrographs of the  $Fe_{81}B_{13.5}Si_{3.5}C_2$  samples irradiated with 5 and 10 laser



FIG. 7. Magnetic force micrographs of the  $Fe_{40}Ni_{40}Mo_{4}B_{16}$ samples after irradiation with 10 (laser pulses)/spot. The left side corresponds to the topographical features.

pulses/spot, respectively. The left side represents the topographical features of the irradiated surfaces, while the right side shows the newly developed magnetic domain structures. Striking submicron domains can be noticed in these pictures, in contradistinction to the MFM photos of the lower magnetostriction samples. Indeed, the MFM micrographs of the  $Fe_{40}Ni_{40}Mo_{4}B_{16}$  system after laser processing, shown in Fig. 7, clearly show the absence of any kind of domain structures in the laser treated alloy. This result is in agreement with the lower-magnetostriction value of this system as well as its different behavior upon irradiation evidenced by Mössbauer spectroscopy findings. Consequently, the formation of submicron magnetic domain structures upon laser irradiation of highly magnetostriction metallic glasses correlates well with the development of a preferred out-of-plane orientation of the average direction of bulk magnetization, as demonstrated by Mössbauer data. Magnetic domain structures have been induced by laser processing of amorphous magnets and clearly evidenced by magnetic force microscopy.

#### **IV. CONCLUSIONS**

Higher-magnetostriction metallic glasses develop out-ofplane magnetic anisotropy as the effect of pulsed-excimerlaser processing, while lower-magnetostriction systems exhibit random orientation of the magnetic moment directions, under the same conditions of treatment. The surface magnetic texture remains in plane. Correspondingly, the highermagnetostriction samples develop submicron magnetic domain structure, which are totally absent after irradiation in lower-magnetostriction samples. Our present results open the way to obtaining amorphous magnets with tailored properties by means of laser processing.

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