

## Soft magnetism, magnetostriction, and microwave properties of FeGaB thin films

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A series of  $(\text{Fe}_{100-y}\text{Ga}_y)_{1-x}\text{B}_x$  ( $x=0-21$  and  $y=9-17$ ) films were deposited; their microstructure, soft magnetism, magnetostrictive behavior, and microwave properties were investigated. The addition of B changes the FeGaB films from polycrystalline to amorphous phase and leads to excellent magnetic softness with coercivity  $<1$  Oe, high  $4\pi M_s$ , self-biased ferromagnetic resonance (FMR) frequency of 1.85 GHz, narrow FMR linewidth ( $X$  band) of 16–20 Oe, and a high saturation magnetostriction constant of 70 ppm. The combination of these properties makes the FeGaB films potential candidates for tunable magnetoelectric microwave devices and other rf/microwave magnetic device applications. © 2007 American Institute of Physics. [DOI: 10.1063/1.2804123]

Magnetoelectric (ME) composite materials consisting of both magnetic and ferroelectric materials with strong ME coupling have led to many different applications,<sup>1–4</sup> which require magnetic materials with a large saturation magnetostriction constant and a low saturation field.<sup>5</sup> Strong ME coupling has been achieved in bulk ME composite materials, but it has been challenging in achieving strong ME coupling in thin film magnetoelectric composite materials. High quality metallic soft magnetic films can be deposited by physical vapor deposition with a low cost and at room temperature, which lead to easy integration into different integrated circuits. Metallic magnetic films with large saturation magnetostriction constant and low saturation field, however, are not readily available. In particular, such metallic magnetic films with low loss tangents at rf/microwave frequencies, if made available, can lead to integrated electrostatically tunable rf/microwave magnetoelectric devices.

Terfenol-D, with a saturation magnetostriction constant up to 1600 ppm, is well known as a giant magnetostrictive material. It has been utilized in many low frequency devices,<sup>3,4</sup> but is hard to be saturated.<sup>6</sup> On the other hand, binary FeGa alloys (also called Galfenol) show great promise with a high saturation magnetostriction constant of 400 ppm for single crystals<sup>7</sup> and 275 ppm for directional solidified polycrystalline alloys,<sup>8</sup> a low saturation field in the order of 100 Oe, and a large saturation magnetization of 18 kG.<sup>9</sup> However, FeGa single crystal films have been very lossy at microwave frequencies with large ferromagnetic resonance (FMR) linewidth of 450–600 Oe at  $X$  band,<sup>10</sup> which is too high to be incorporated into microwave magnetoelectric devices.

Incorporation of metalloid element carbon into FeGa alloys was most recently shown to destabilize the  $DO_3$  phase that is detrimental to high saturation magnetostriction, lead-

ing to magnetostriction constants higher than those of the FeGa binary alloys.<sup>11</sup> Boron is a well known metalloid element that is widely used in soft magnetic alloy films such as the FeCoB films.<sup>12–14</sup> The addition of B into FeCo films causes refined grain size and diminished magnetocrystalline anisotropy, which lead to excellent magnetic softness and microwave performance.<sup>12–14</sup> In addition, the most recent investigations on the effect of B addition into FeGa bulk polycrystalline alloys<sup>15</sup> and melt-spun ribbons<sup>16</sup> show that B can indeed have a beneficial effect on magnetostriction.

In this letter, the effects of B addition to the microstructure, soft magnetism, magnetostriction, as well as microwave properties of FeGaB films were investigated. It is shown that a combination of very low coercivity, narrow FMR linewidth, high self-biased FMR frequency, and high saturation magnetostriction constant have been achieved, making these FeGaB films potential candidates for electrostatically tunable microwave magnetoelectric devices and other rf/microwave magnetic devices.

FeGaB films with varied B contents were sputtered onto silicon substrates from two separate targets of  $\text{Fe}_{80}\text{Ga}_{20}$  (at. %) and boron under an *in situ* magnetic field which induces a uniaxial in-plane magnetic anisotropy. B content of FeGaB films was varied by changing boron gun power while keeping the  $\text{Fe}_{80}\text{Ga}_{20}$  gun power at 50 W on a 2 in. diameter target. FeGaB films were deposited with a thickness of 100 nm in 3 mTorr Ar atmosphere at a rate of 60 Å/min in a vacuum chamber with a base pressure better than  $1 \times 10^{-7}$  Torr. Compositions of FeGaB films were examined by X-ray photoelectron spectroscopy (XPS), and their crystal structures were characterized by X-ray diffraction (XRD) with the Cu  $K\alpha$  source. Magnetic properties of these FeGaB films were measured by vibrating sample magnetometer and magnetostriction constants of them were measured on a customer-made magnetostriction tester with a rotating field. The magnetostriction tester has a large drive of field up to

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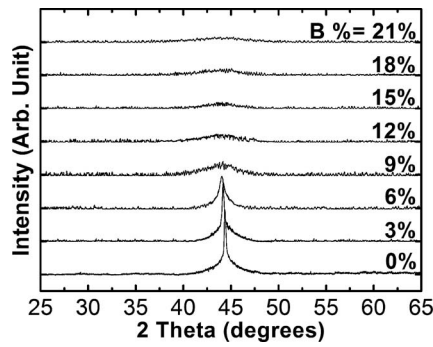


FIG. 1. XRD patterns for FeGaB films with different B contents.

300 Oe and a good resolution of 0.1 ppm for the magnetostriction constant for 10 nm thick films on 200  $\mu\text{m}$  thick silicon substrates. Microwave properties were characterized by both broadband and narrowband techniques. The broadband customer-made permeameter has a bandwidth of 0.05–5 GHz, and the narrowband technique is an X-band ( $\sim 9.6$  GHz) FMR spectrometer.

From the results of XPS, compositions of these alloys can be expressed by  $(\text{Fe}_{100-y}\text{Ga}_y)_{1-x}\text{B}_x$  (at. %) with B contents in the range of 0–21, and Ga in the range of 9–17. The Ga contents are slightly off from the  $\text{Fe}_{81}\text{Ga}_{19}$  composition, which has the highest saturation magnetostriction constant.<sup>17</sup> The higher the B power, the less the Ga to Fe ratio, even though the FeGa target composition is fixed at  $\text{Fe}_{80}\text{Ga}_{20}$  and the power for the FeGa target is also fixed.

XRD analyses of these FeGaB films indicate that these films show only one bcc (110) diffraction peak, which broadens as the B content increases from 0 to 6 at. % (Fig. 1). This indicates that the addition of B atoms into the FeGaB alloys leads to refined grain size and/or a more disordered lattice. The (110) diffraction peak position also shifts to lower angles with the addition of B, indicating a gradually expanded bcc lattice with the lattice parameters changing from 2.89 to 2.91 nm. The expanded lattice indicates that B atoms reside in the interstitial sites of the bcc FeGa. This result matches with what has been observed in FeCoB films.<sup>18</sup> The (110) peak changes into a broad hump when the B content is 9 at. % or higher, suggesting that an amorphous phase is formed.

The soft magnetic properties of these FeGaB films are shown in Fig. 2. As the B content increases, the saturation magnetization drops gradually and the entire trend is nearly linear, similar to what was reported for FeCoB.<sup>14,18</sup> Binary FeGa film has a saturation magnetization of 17.5 kG, which

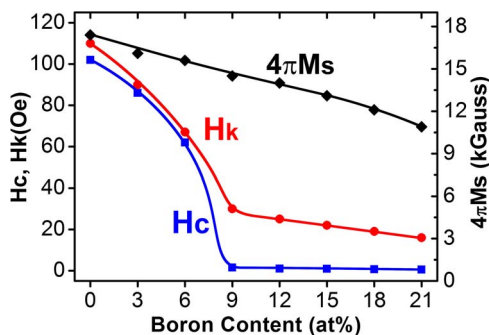


FIG. 2. (Color online) Coercivity ( $H_c$ ), in-plane anisotropy field ( $H_k$ ), and  $4\pi M_s$  of FeGaB films with different B contents.

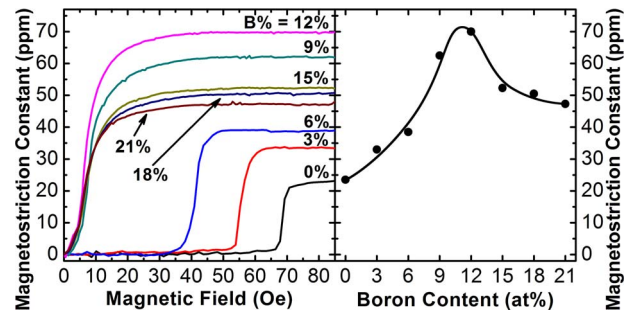


FIG. 3. (Color online) (a) Magnetostriction vs magnetic field. (b) Saturation magnetostriction constant vs B content.

is close to what was reported by Clark *et al.*,<sup>9</sup> while the FeGaB film with 21 at. % B has a saturation magnetization of only 11 kG. With the addition of B, the soft magnetic properties of these films are dramatically improved, similar to what was found in FeCoB films.<sup>12–14</sup> The coercivity of the FeGaB films was significantly reduced from  $\sim 110$  Oe for binary FeGa film to less than 1 Oe with B content  $\geq 9$  at. % and reaches 0.4 Oe at 21 at. % B. The effective in-plane anisotropy field  $H_k$ , which is induced by an *in situ* magnetic field during deposition, shows a similar trend, which drops dramatically from 120 to 30 Oe at a B content of 9 at. % and reaches 15 Oe at 21 at. % B. For FeGaB films without a well defined uniaxial anisotropy, i.e., films with B content  $\leq 6$  at. %, the  $H_k$  is defined as the field to reach 95% saturation magnetization. The changes in soft magnetic properties of these FeGaB films can be understood on the basis of XRD results. When the B content in FeGaB films is 6 at. % or less, B addition leads to a refined grain size, which results in gradually reduced coercivities and anisotropy fields. At a B content of 9 at. % and higher, the FeGaB films turned amorphous. The elimination of magnetocrystalline anisotropy results in a sudden drop of the coercivity and anisotropy fields and significantly improved magnetic softness.

Magnetostriction behavior of these FeGaB films is shown in Fig. 3(a) as a function of the rotating magnetic field for FeGaB films with different B contents. At B contents of 0, 3, and 6 at. %, there is almost no magnetostrictive response until the applied magnetic field reaches 67, 53, and 38 Oe, respectively. While the amorphous FeGaB films with  $\geq 9$  at. % B content show magnetostrictive response at very low fields. These magnetostrictive behaviors are clearly consistent with the coercivity and anisotropy field data shown in Fig. 2. The saturation magnetostriction constants for these FeGaB alloy films are also shown in Fig. 3(b). Interestingly, the saturation magnetostriction constant of binary  $\text{Fe}_{83}\text{Ga}_{17}$  film is only 23 ppm, which is one order of magnitude lower than reported values for bulk  $\text{Fe}_{81}\text{Ga}_{19}$  alloys.<sup>7–9</sup> The magnetostriction constant of a binary FeGa alloy film with a {110} fiber texture can be expressed by  $\lambda_{\{110\}} = (3\lambda_{100} + 5\lambda_{111})/8$ ,<sup>19</sup> which should lead to an expected value of 73 ppm with  $3/2\lambda_{100} = 300$  ppm and  $3/2\lambda_{111} = -20$  ppm.<sup>11</sup> The large discrepancy between measured and expected magnetostriction constants for FeGa films could be due to the difference in the microstructure between deposited films and bulk FeGa alloys, and needs further investigation.

When B atoms incorporate into the FeGa bcc lattice, the magnetostriction constant firstly increases and reaches a maximum of 70 ppm at a B content of 12 at. %, and then drops to 46 ppm at a B content of 21 at. %. The maximum

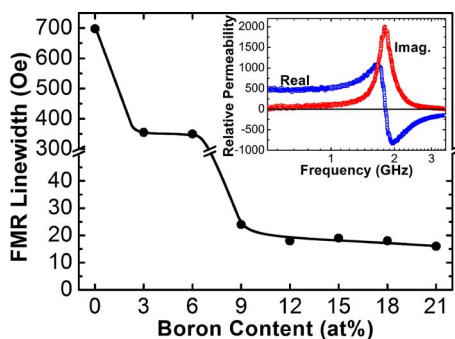


FIG. 4. (Color online) X-band FMR linewidth vs B content. (Inset) Permeability spectrum of FeGaB<sub>12</sub> film under zero bias field.

magnetostriction constant is three times of that of the binary FeGa film. This nonlinear magnetostriction behavior as a function of B content was also discovered in other amorphous alloys such as FeB (Ref. 20) and FeCoB.<sup>13</sup> The reason for this, according to Clark *et al.*,<sup>17</sup> is that a low concentration of metalloid atoms could lead to atomic pairs such as Ga–Ga or B–B, which enhance the magnetostrictive behavior, while a relatively higher amount of metalloid atoms would form clusters that decrease the magnetostriction constant. It is also believed<sup>11</sup> that a small amount of B atoms, just like C atoms, would stay in the interstitial sites of the bcc lattice and destabilize the DO<sub>3</sub> phase, leading to enhanced magnetostriction constants. Further confirmation on these statements is under investigation with extended X-ray absorption fine structure at national synchrotron X-ray source.

FMR measurements on these FeGaB films were carried out at X band with external field applied parallel to the in-plane easy axis direction. Figure 4 shows the relation between FMR linewidth and B content. For the binary FeGa film, its FMR linewidth is about 700 Oe, which is comparable to that of single crystal FeGa films.<sup>10</sup> The FMR linewidth of FeGaB films drops dramatically with the incorporation of B atoms to 24 Oe at a B content of 9 at.%, and stays below 20 Oe at  $\geq 12$  at.% of B, and reaches 16 Oe at 21 at.% B. These FMR linewidth values of 16–20 Oe at X band are record low values for  $\sim 100$  nm thick metallic magnetic amorphous films. Again, the B content dependence of FMR linewidth is closely related to their soft magnetic properties, as shown in Fig. 2, and can be explained by their microstructures. The permeability spectrum of FeGaB films with a B content of 12 at.% under zero bias field is also shown in the inset of Fig. 4, which indicates an initial relative permeability of 500 and a self-biased FMR frequency of 1.85 GHz. However, according to Fig. 2, with a saturation magnetization of 14.5 kG and a uniaxial anisotropy of 25 Oe, the FeGaB films with a 12 at.% B content should possess a FMR frequency of 1.69 GHz. The slight difference between these two FMR frequencies could be due to the

difference between static and dynamic magnetic anisotropies of these FeGaB films, as observed for Permalloy.<sup>21</sup>

It is notable that all discussions above are made as a function of B content, while the Fe to Ga ratio differs slightly for these FeGaB films, which may contribute to observed changes of film properties. Additional work is being done to investigate the effects of the Fe to Ga ratio.

It was found that with B addition to the FeGa films, the microstructure of these films changed from polycrystalline to amorphous phase, which led to dramatically enhanced soft magnetic and microwave properties. In addition, the incorporation of B atoms in these FeGaB films led to a nearly tripled saturation magnetostriction constant at a B content of 12 at.%. The combination of soft magnetism, large magnetostriction constant, and excellent microwave magnetic properties makes these FeGaB films potential candidates for magnetoelectric materials and for other rf/microwave device applications.

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<sup>1</sup>M. I. Bichurin, I. A. Kornev, V. M. Petrov, A. S. Tatarenko, Yu. V. Kiliba, and G. Srinivasan, *Phys. Rev. B* **64**, 094409 (2001).

<sup>2</sup>A. Ustinov, G. Srinivasan, and B. A. Kalinikos, *Appl. Phys. Lett.* **90**, 031913 (2007).

<sup>3</sup>S. X. Dong, J. F. Li, and D. Viehland, *Appl. Phys. Lett.* **83**, 2265 (2003).

<sup>4</sup>S. X. Dong, J. F. Li, and D. Viehland, *Appl. Phys. Lett.* **85**, 2307 (2004).

<sup>5</sup>S. X. Dong, J. F. Li, and D. Viehland, *IEEE Trans. Ultrason. Ferroelectr. Freq. Control* **50**, 1253 (2002).

<sup>6</sup>G. Engdahl and I. D. Mayergoyz, *Handbook of Giant Magnetostrictive Materials* (Academic, New York, 2000), pp. 209–217.

<sup>7</sup>A. E. Clark, M. Wun-Fogle, J. B. Restorff, and T. A. Lograsso, *Mater. Trans., JIM* **43**, 881 (2002).

<sup>8</sup>N. Srisukhumbornchai and S. Guruswamy, *J. Appl. Phys.* **90**, 5680 (2001).

<sup>9</sup>A. E. Clark, J. B. Restorff, M. Wun-Fogle, T. A. Lograsso, and D. L. Schlagel, *IEEE Trans. Magn.* **36**, 3238 (2000).

<sup>10</sup>A. Butera, J. Gómez, J. L. Weston, and J. A. Barnard, *J. Appl. Phys.* **98**, 033901 (2005).

<sup>11</sup>A. E. Clark, J. B. Restorff, M. Wun-Fogle, K. B. Hathaway, T. A. Lograsso, M. Huang, and E. Summers, *J. Appl. Phys.* **101**, 09C507 (2007).

<sup>12</sup>H. Imrane, J. Lou, Z. Cai, K. S. Ziemer, and N. X. Sun (unpublished).

<sup>13</sup>C. L. Platt, M. K. Minor, and T. J. Klemmer, *IEEE Trans. Magn.* **37**, 2302 (2001).

<sup>14</sup>I. Kim, J. Kim, K. H. Kim, and M. Yamaguchi, *IEEE Trans. Magn.* **40**, 2706 (2004).

<sup>15</sup>Y. Gong, C. Jiang, and H. Xu, *Acta Metall. Sin.* **42**, 830 (2006).

<sup>16</sup>T. Kubota and A. Inoue, *Mater. Trans.* **45**, 199 (2004).

<sup>17</sup>A. E. Clark, M. Wun-Fogle, J. B. Restorff, T. A. Lograsso, and J. R. Cullen, *IEEE Trans. Magn.* **37**, 2678 (2001).

<sup>18</sup>M. Munakata, S. Aouki, and M. Yagi, *IEEE Trans. Magn.* **41**, 3262 (2005).

<sup>19</sup>A. Hosono and Y. Shimada, *J. Appl. Phys.* **67**, 6981 (1990).

<sup>20</sup>N. Tsuya and K. I. Arai, *J. Appl. Phys.* **50**, 1658 (1979).

<sup>21</sup>R. Lopusnik, J. P. Nibarger, T. J. Silva, and Z. Celinski, *Appl. Phys. Lett.* **83**, 96 (2003).