Magnetostriction of ternary Fe–Ga–X (X = C,V,Cr,Mn,Co,Rh) alloys

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Magnetostriction of ternary Fe–Ga–X (X = C, V, Cr, Mn, Co, Rh) alloys

Abstract
Binary iron-gallium (Galfenol) alloys have large magnetostrictions over a wide temperature range. Single crystal measurements show that additions of 2 at. % or greater of 3d and 4d transition elements with fewer (V, Cr, Mo, Mn) and more (Co, Ni, Rh) valence electrons than Fe, all reduce the saturation magnetostriction. Kawamiya and Adachi [J. Magn. Magn. Mater. 31–34, 145 (1983)] reported that the DO₃ structure is stabilized by 3d transition elements with electron/atom ratios both less than iron and greater than iron. If DO₃ ordering decreases the magnetostriction, the maximum magnetostriction should be largest for the (more disordered) binary Fe–Ga alloys as observed. Notably, addition of small amounts of C (0.07, 0.08, and 0.14 at. %) increases the magnetostriction of the slow cooled binary alloy to values comparable to the rapidly quenched alloy. We assume that small atom (C, B, N) additions enter interstitially and inhibit ordering, thus maximizing the magnetostriction without quenching.

Keywords
iron alloys, gallium alloys, vanadium alloys, chromium alloys, manganese alloys, cobalt alloys, rhenium alloys, magnetostriction, magnetisation

Disciplines
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Comments
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Binary iron–gallium (Galfenol) alloys have large magnetostrictions over a wide temperature range. Single crystal measurements show that additions of 2 at. % or greater of 3d and 4d transition elements with fewer (V, Cr, Mo, Mn) and more (Co, Ni, Rh) valence electrons than Fe, all reduce the saturation magnetostriction. Kawamiya and Adachi [J. Magn. Magn. Mater. 31–34, 145 (1983)] reported that the DO3 structure is stabilized by 3d transition elements with electron/atom ratios both less than iron and greater than iron. If DO3 ordering decreases the magnetostriction, the maximum magnetostriction should be largest for the (more disordered) binary Fe–Ga alloys as observed. Notably, addition of small amounts of C (0.07, 0.08, and 0.14 at. %) increases the magnetostriction of the slow cooled binary alloy to values comparable to the rapidly quenched alloy. We assume that small atom (C, B, N) additions enter interstitially and inhibit ordering, thus maximizing the magnetostriction without quenching. © 2007 American Institute of Physics.

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INTRODUCTION

Iron–gallium alloys (Galfenol) are interesting materials because of both their high magnetostriction and their desirable mechanical properties. Magnetostriction can be as high as 400 ppm in single crystals and 275 ppm in highly textured polycrystals.1,2 Fe–Ga is mechanically strong and can support tensile stresses up to 500 MPa, unlike current active materials such as Terfenol-D, lead zirconic titanate (PZT), and lead magnesium niobate (PMN). Fe–Ga alloys can also be machined and welded with conventional metal working techniques. The magnetostriction shows a very small temperature dependence. For example, magnetostriction in a quenched Fe79.3Ga20.6 sample rises slowly from 385 ppm at 300 K to 423 ppm at 4.4 K.1 However, the most astonishing property is that after annealing under a compressive stress the material maintains its full magnetostriction when subjected to as much as 50 MPa of applied tensile stress.3

Because Hall4 showed that small additions of transition metals (V, Cr, Mo) increase the λ100 magnetostriction of pure iron, it was of interest to see if transition metal additions would have the same effect on the Galfenol alloys. The addition of some ternary elements to improve the magnetic and/or mechanical properties of the binary system has been studied previously. Reference 5 examined additions of Ni, Mo, Al, and Sn. With the exception of Sn, all the additions lowered the magnetostriction. Small amounts of Sn, which has a much larger atomic radius than Ga, surprisingly left the magnetostriction unchanged for Ga concentrations near 18 at. % and increased the magnetostriction for low Ga concentrations. This paper continues the exploration of ternary additions and examines V, Cr, Mn, Co, and Rh additions in the 2–15 at. % range and C additions in the 0.07–0.17 at. % range.

EXPERIMENT AND RESULTS

Alloys were prepared by arc melting and single crystals were grown by the Bridgman method. Oriented disks of 6.35 mm in diameter and 2.5 to 3 mm thick were cut from larger boules with the [100] and [011] directions normal to the disk. In-plane directions of [100] and [111] were identified and marked on the surface of the disks. A strain gage was glued parallel to either the [100] direction (⟨100⟩ samples) or the [111] direction (⟨011⟩ samples). Magnetostriction was measured by rotating the disk in a 15 kOe magnetic field applied parallel to the face of the disk. The resulting strain was a very good fit to the lowest order magnetostriction term, which follows a cos^2 θ angular dependence. The next higher order magnetostriction term, which follows a cos^4 θ angular dependence, was generally less than 10 ppm. Measurements at 20 kOe demonstrated that the samples were saturated during measurements. Figure 1 shows 3/2 λ100 and 3/2 λ111 strain measurements for Gal-

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fenol with Cr and Co ternary additions. From the relative phases between the \( \lambda_{100} \) and \( \lambda_{111} \) (smaller amplitude) curves, it can be seen that \( \lambda_{111} \) is negative in the 2 at. % Cr case and positive in all other cases.

Table I shows the compositions and measured magnetostrictions of the samples used in these experiments. Data from Table I are plotted in Figs. 2, 3, and 4 along with the results of previously reported ternary additions.5

### DISCUSSION

As seen in Fig. 2, the addition of V, Cr, Mn, Co, and Rh reduced the tetragonal magnetostriction, \( \lambda_{100} \), compared to the Fe–Ga binary with a similar Ga concentration. Kawamiya and Adachi6 and Nishino et al.7 have pointed out that additions of elements with either more or fewer electrons per atom than iron stabilize the \( D_03 \) phase, which is conjectured to lower the \( \lambda_{100} \) magnetostriction.8 Stabilization of \( D_03 \) also has been argued to account for a change in sign from negative to positive for \( \lambda_{111} \) as a function Ga content in the binary alloys.9 The increased \( D_03 \) stabilization may be attributed to the substitution of the ternary atoms on different sites of the \( D_03 \) structure—atoms with electron/atom ratios less than the average Fe 3Ga electron/atom ratio reside on the mixed simple cubic lattice \( Fe I \) and atoms with electron/atom ratios greater than the Fe 3Ga electron/atom ratio reside on the pure iron sublattice \( Fe II \).6 In both cases, this increases the difference of the average atomic sizes between the two simple cubic lattices, stabilizing the formation of an ordered phase.

#### FIG. 1. Room temperature strain vs angle curves at 15 kOe. (a) ~2 at. % Cr, (b) ~5.1 at. % Cr, (c) ~9.8 at. % Co, and (d) ~14.9 at. % Co. The large amplitude strains are \( 3/2 \lambda_{100} \) and the small amplitude strains are \( 3/2 \lambda_{111} \). Positive \( \lambda_{111} \) is in phase with \( \lambda_{100} \) as in (b)–(d); negative \( \lambda_{111} \) is out of phase with \( \lambda_{100} \) (a). Note the different scales on the figure. See Table I for complete details.

#### FIG. 2. Saturation magnetostriction \( (3/2 \lambda_{100}) \) of ternary alloys \( (Fe_{100-x}Ga_xX, with X=V, Cr, Mn, Co, or Rh including previously reported Ni, Sn, and Mo) vs Ga content x \) (see Ref. 5). The lines are guides to the eye for the binary alloy (see Fig. 4): solid, unquenched; dashed, quenched.

<table>
<thead>
<tr>
<th>Composition</th>
<th>3/2 ( \lambda_{100} )</th>
<th>3/2 ( \lambda_{111} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe(<em>{82.3})Ga(</em>{17.6})C(_{0.07})</td>
<td>322 (298)</td>
<td>...</td>
</tr>
<tr>
<td>Fe(<em>{83.3})Ga(</em>{16.0})C(_{0.08})</td>
<td>268</td>
<td>...</td>
</tr>
<tr>
<td>Fe(<em>{81.1})Ga(</em>{18.6})C(_{0.08})</td>
<td>369 (334)</td>
<td>...</td>
</tr>
<tr>
<td>Fe(<em>{81.3})C(</em>{0.17})</td>
<td>342</td>
<td>...</td>
</tr>
<tr>
<td>Fe(<em>{82.5})Ga(</em>{15.9})V(_{1.6})</td>
<td>257</td>
<td>...</td>
</tr>
<tr>
<td>Fe(<em>{80.7})Ga(</em>{17.4})V(_{1.9})</td>
<td>253</td>
<td>...</td>
</tr>
<tr>
<td>Fe(<em>{82.1})Ga(</em>{15.3})Cr(_{2.1})</td>
<td>255</td>
<td>...</td>
</tr>
<tr>
<td>Fe(<em>{78.1})Ga(</em>{16.7})Mn(_{1.0})</td>
<td>76</td>
<td>...</td>
</tr>
<tr>
<td>Fe(<em>{74.1})Ga(</em>{15.3})Cr(_{3.1})</td>
<td>3</td>
<td>...</td>
</tr>
<tr>
<td>Fe(<em>{82.0})Ga(</em>{15.3})Cr(_{3.2})</td>
<td>...</td>
<td>-13</td>
</tr>
<tr>
<td>Fe(<em>{73.5})Ga(</em>{15.3})Cr(_{5.0})</td>
<td>...</td>
<td>3</td>
</tr>
<tr>
<td>Fe(<em>{82.5})Ga(</em>{16.7})Mn(_{3.8})</td>
<td>...</td>
<td>-21</td>
</tr>
<tr>
<td>Fe(<em>{80.7})Ga(</em>{16.6})Mn(_{3.8})</td>
<td>303 (285)</td>
<td>...</td>
</tr>
<tr>
<td>Fe(<em>{83.3})Ga(</em>{15.3})Mn(_{0.8})</td>
<td>258 (261)</td>
<td>...</td>
</tr>
<tr>
<td>Fe(<em>{79.8})Ga(</em>{16.6})Mn(_{3.8})</td>
<td>...</td>
<td>-11 (-16)</td>
</tr>
<tr>
<td>Fe(<em>{78.0})Ga(</em>{16.7})Mn(_{4.1})</td>
<td>...</td>
<td>15</td>
</tr>
<tr>
<td>Fe(<em>{81.1})Ga(</em>{17.7})Co(_{1.0})</td>
<td>323</td>
<td>...</td>
</tr>
<tr>
<td>Fe(<em>{78.0})Ga(</em>{19.9})Co(_{1.9})</td>
<td>253</td>
<td>...</td>
</tr>
<tr>
<td>Fe(<em>{74.1})Ga(</em>{16.9})Co(_{7.7})</td>
<td>116</td>
<td>...</td>
</tr>
<tr>
<td>Fe(<em>{87.7})Ga(</em>{17.4})Co(_{14.8})</td>
<td>43</td>
<td>...</td>
</tr>
<tr>
<td>Fe(<em>{84.2})Ga(</em>{16.9})Co(_{8.8})</td>
<td>...</td>
<td>10</td>
</tr>
<tr>
<td>Fe(<em>{87.7})Ga(</em>{17.7})Co(_{9.5})</td>
<td>...</td>
<td>29</td>
</tr>
<tr>
<td>Fe(<em>{88.8})Ga(</em>{18.0})Rh(_{1.6})</td>
<td>99 (105)</td>
<td>...</td>
</tr>
<tr>
<td>Fe(<em>{79.3})Ga(</em>{18.3})Rh(_{2.2})</td>
<td>264 (250)</td>
<td>...</td>
</tr>
</tbody>
</table>
Increased $D_0$ stability is also reflected in the rhombohedral magnetostriction measurements $\lambda_{111}$ of Table I and Fig. 1. Table I shows that as the amount of the ternary alloy (Cr, Mn, and Co) is increased, the magnetostriction of $\lambda_{111}$ becomes more positive as is found in the $D_0$ binary alloys. The change in sign from minus to plus as the concentration of Cr is increased from 2 at. % to 5 at. % is also illustrated by the phase relations in Figs. 1(a) and 1(b).

Figure 3 compares the rhombohedral magnetostriction, $\lambda_{111}$, of the binary and ternary alloys. In agreement with the increased stability of the $D_0$ phase with transition metal additions, ternary alloys show magnetostriction consistent with the $D_0$ phase (positive $\lambda_{111}$ magnetostriction) at Ga concentrations less than the binary alloys.

In contrast to the V, Cr, Mn, Co, and Rh substitutions, the addition of small amounts of carbon to the Fe–Ga binary produces scientifically and technically interesting results. These results are shown in Fig. 4 along with the magnetostriction of Fe–Ga binary plotted as a function of Ga concentration for both quenched and unquenched samples. At Ga concentrations larger than ~18 at. %, the saturation magnetostriction is known to be improved by quenching the binary samples from a high temperature, typically 800 or 1000 °C. Unquenched carbon-containing samples, with Ga > 18 at. % lie above the unquenched binary curve. This suggests that the carbon additions (and possibly other small atoms such as B or N) have the same salutary effect on the magnetostriction as quenching, without the added step of quenching the alloys, implying that the phase stability of $D_0$ relative to $A_2$ is reduced. We suggest that carbon atoms enter interstitially, whereas the transition metals enter substitutionally. Carbon interstitially incorporated into the Fe–Ga alloys could also generate tetragonal distortions within the Fe lattice similar to those observed in the Fe–Al alloys. These tetragonal distortions may play an important role in the enhanced magnetoelasticity of the Fe–Ga alloys. Magnetoelastic measurements of additional compositions and detailed structural studies needed to confirm this picture are underway.

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