Materials with high magnetostriction

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Abstract. The magnetostrictive properties of cubic Laves phases such as TbFe₂, Terfenol D and SmFe₂ are summarised. It is shown that SmFe₂ can be an interesting candidate as a high magnetostrictive material even in the as cast “dirty” state. Fe-X alloys based on Fe-Ga but also Fe-Al are presented as materials with a medium magnetostriction, however with the big advantage of a magnetically soft material. Generally all nonmagnetic substutional elements which cause an increase of the interatomic distance in the Fe-lattice (such as Ga, Al, Si) cause an increase of the magnetostriction too. In Ni-Ga a reduction of the magnetostriction was found. Another interesting material with high magnetostriction is CoFe₂O₄ which is cheap, easy to produce and an oxidic material. Here magnetostriction values up to 400 ppm can be achieved by hydrostatic compaction followed by a field annealing.

1. Introduction
Magnetostriction is an intrinsic property of all kind of magnetically ordered materials. Due to the magnetic exchange the materials change its length (parallel and perpendicular to the external field) as well as its volume. In a single crystal magnetostriction is a tensor property which depends on the symmetry of the lattice. This change of length can be between 10⁻⁸ and 10⁻³ (Δl/l₀) depending on the involved elements as well as on the local symmetry. Magnetostriction is based on the coupling of the magnetic moment to the lattice, which means that here the orbital moment L of the atom has to be considered. Materials with high magnetostriction are technologically important for sensor or actuator applications.

In this paper different materials with high magnetostriction will be discussed and surveyed. Also a theoretical background of the origin of the magnetostriction will be given. Generally a single crystal is best suited for characterizing the magnetostriction, however often only data of polycrystalline material are available.

2. Rare earth based magnetostrictive systems
Pure rare earth elements (such as Tb, Dy,...) exhibit due to its orbital magnetism generally a huge magnetostriction, however only at low temperatures. Therefore alloys between rare earths and 3d-elements, which have ordering temperatures above room temperature have to be considered. The highest magnetostriction at room temperature was found in TbFe₂ as well as in SmFe₂ [1,2]. These are cubic Laves phase materials where the easy axis is parallel to the [111] direction. Clark [3] as well as Hathaway and Cullen [4] suggested a microscopic model for this magnetostriction as demonstrated graphically in figure 1. Figure 2 shows as an example the in TbFe₂ measured magnetostriction into the [111] direction at different temperatures [1]. At room temperature a value of nearly +2000 ppm is achieved however at external fields of 800 kA/m (~ 10 kOe). This field is too high for technical
applications. In order to reduce the magnetocrystalline anisotropy of Tb about 70% Dy is added which leads to a material which is under the trade name Terfenol D (Tb$_{0.3}$Dy$_{0.7}$)Fe$_{1.92}$ industrially available. Terfenol D is generally used in a pre-stressed state as well as aligned material. Figure 3 shows room temperature magnetostriction measurements on Terfenol D. With this material a linear magnetostriction of about 1000 ppm is available, however at fields of 5 kOe. H.S.Lim et al [5] report about a plastic bonded Terfenol D which has the advantage that the material can also be used in dynamic applications due to the reduced electrical resistivity. Table 1 compares single crystal magnetostriction values of Fe, Ni and Terfenol D.

**Table 1.** Single crystal magnetostriction values of Fe, Ni and Terfenol D.

| Material     | $\lambda_{(100)}$ ppm | $\lambda_{(111)}$ ppm |
|--------------|-----------------------|-----------------------|
| Fe           | -20                   | -21                   |
| Ni           | -46                   | -24                   |
| Terfenol D   | 90                    | 1640                  |

**Figure 1.** Model of the changed distance between the rare earth atoms (Tb) due to the coupling between the 4f clouds located at “1” and “2” [3,4].

**Figure 2.** Magnetostriction of a TbFe$_2$ single crystal measured in the [111] direction at different temperatures [1].

**Figure 3.** Magnetostriction of industrial Terfenol D at room temperature.

**Figure 4.** Magnetostriction of a SmFe$_2$ single crystal measured in the [111] direction at room temperature [6].
Even Terfenol D is an excellent high magnetostrictive material (see figure 3), the technical application is limited due to the brittleness of the material as well as due to the high costs of the heavy rare earth elements. Another option is SmFe$_2$ which is also a [111] cubic Laves phase which exhibit a high but negative magnetostriction at room temperature, RT, see figure 4. The easy axis of magnetization is parallel to the [1 1 0] direction at low temperatures, however it changes to the [111] direction at temperatures above 195 K.

SmFe$_2$ shows a typical cubic anisotropy with anisotropy constants $K_1=5.3 \times 10^6$ and $K_2=1.9 \times 10^6$ erg/cm$^3$ at 300 K, however a magnetoelastic part has also to be considered [6]. Magnetostriction constants were determined to be $\lambda_{111} = -2010$ ppm, $\lambda_{100} = -130$ ppm and $\lambda_5 = -1258$ ppm at 300 K. The problem of producing this material is that due to the high vapor pressure of Sm combined with the peritectic point of the formation of SmFe$_2$ in the binary phase diagram it is rather difficult to achieve single phase material. However even just after a melting process (which leads to a “dirty” sample) a rather high magnetostriction of 500 ppm (see figure 5) or more in a polycrystalline state can be achieved. However in the as-cast state, the sample was not homogeneous. The chemical composition of the alloy determined by electron probe microanalysis (EPMA) EDX technique showed that the as cast sample exhibits different compositions at different spots such as: SmFe$_{2.3}$, SmFe$_{1.25}$ and SmFe$_{1.5}$. When this material is submitted to a heat treatment at 700°C for 132 h under vacuum with a Ta-foil as getter material, XRD analysis showed that the sample became single phase SmFe$_2$. This single phase SmFe$_2$ presented a crystallite size of 21nm and a lattice constant of 7.426648 Å corresponding to the space group Fd3m. The magnetostriction could be improved significantly – see figure 6.

![Figure 5](image1.png) **Figure 5.** Magnetostriction of a polycrystalline, as-cast “dirty” SmFe$_2$ measured at room temperature.

![Figure 6](image2.png) **Figure 6.** Magnetostriction of a annealed, polycrystalline SmFe$_2$ measured at room temperature.

Also in SmFe$_2$ the magnetocrystalline anisotropy is rather high, which makes it difficult to saturate this material. However producing this material (as well as TbFe$_2$) in a nanocrystalline state might be a way to reduce the anisotropy. This route was already proved on thin film TbFeCo multilayers with excellent soft magnetic properties [7].

2.1. Fe-Ga (Galfenol)

Generally 3d elements (such as Fe, Co or Ni) exhibit a rather low magnetostriction (see table 1), which comes from the fact that in these elements the orbital moment is considered to be quenched. However due to the much lower price and the excellent mechanical properties, especially Fe-based alloys are always of technical interest. Compared to highly anisotropic and corrosive rare earths Fe-based alloys Fe-Ga alloys have the advantage that they are generally magnetically soft and chemically rather stable. In the last years the group of Clark et al investigated Fe-Ga and Fe-Ga-Al based alloys [8,9]. It was found that by substituting between 15 and 28 %Ga (or Al) into the Fe lattice causes a significant increase of the magnetostriction [8,9,10]. A similar but smaller effect was found in Fe$_{1-x}$Al$_x$ [9,10]. In the mean time the effect of substituting many different nonmagnetic elements was investigated [11]. Figure 7 shows $3/2 \lambda_{100}$ of the Fe$_{1-x}$Ga$_x$ system which shows values up to 450 ppm [12].
The fundamental question here was, which mechanism drives the development of a high magnetostriction in these materials. The most important points influencing magnetostriction, beside the involved elements, is the structure as well as the local symmetry at different lattice sides. Most of the Fe-X (X = Al, Si, Ge, Ga) phase diagrams look very similar at the Fe-rich side, exhibiting first a disordered A2 structure which changes at higher X-concentrations to ordered B2 and DO3 structures. Clark et al. have explained the main origin of high magnetostriction in 3d-based Fe-Ga alloys is due to the change of magnetoelastic energy because of the Ga substitution causing a lattice softening. The magnetostriction increase is explained by an increase of the magnetoelastic coupling constant due to short range ordering between the Ga–Ga atoms in bcc $\alpha$-iron (A2 structure) with randomly substituted gallium atoms [13]. Measuring the elastic constants in Fe-Ga and calculating the magnetoelastic energy constants, delivers indeed a peak at 19 at. % Ga [12].

Recently, we investigated the local Fe-Ga atomic structure in highly magnetostrictive $\alpha$-Fe$_{80}$Ga$_{20}$ melt-spun ribbons [14] and splat cooled material [15] using Extended X-ray Absorption Fine Structure (EXAFS) analysis at the Fe and Ga K-edges. A careful XRD showed that this ribbon crystallize in the A2 phase. In this work the presence of small Ga clusters could be excluded by EXAFS and XANES, since no first shell Ga-Ga bonds were detected. However, EXAFS analysis of the second coordination shell around Ga clearly provides evidence for the presence of one highly strained (+4%) Ga-Ga pair and five Ga-Fe pairs among six crystallographically equivalent $<100>$ atomic pairs. This conclusion supports recent total energy calculations which assign the large magnetostriction in these alloys to the strain caused by the rotation of the magnetization in the vicinity of such defects.

Even polycrystalline material exhibits much higher magnetostriction then pure Fe. Figure 8 shows as an example magnetostriction measurements performed on polycrystalline Fe-Ga samples [16]. It is clear that for a polycrystalline isotropic material the achievable magnetostriction is smaller than $\lambda_{100}$ (assuming $\lambda_{100} > |\lambda_{111}|$) according to the well-known formula

$$\lambda_s = \frac{1}{5} \left( 2 \lambda_{100} + 3 \lambda_{111} \right)$$

(1)

Fe-Ga based alloys have additionally the advantage that they are magnetically soft, which was demonstrated by hysteresis measurements on ring shaped Fe-Ga samples [17] and is also shown in...
figure 9. Note that in this case an external field of about 16 kA/m (= 200 Oe) is sufficient to saturate the material.

Figure 8. RT magnetostriction measurements on polycrystalline Fe-Ga samples.

Figure 9. Hysteresys loops measured at different frequencies on annealed polycrystalline Fe81Ga19 at RT [17].

2.2. Ni-Ga Alloys

Keeping in view that addition of non-magnetic V, Cr, Al and Ga in Fe increases the magnetostriction of iron as reported by Hall [18] and Clark et al [8,10,19] one has to ask if the same idea will not work in Ni. Therefore two samples of polycrystalline Ni-Ga with the composition Ni98Ga2 and Ni95Ga7 were investigated. Figure 10 shows the magnetostriction ($\lambda_{par}$) of Ni98Ga2 and Ni95Ga7 and pure Nickel in the as cast state. It can be seen that after addition of 2 % Ga the magnetostriction of Ni slightly increases from -35 ppm to -39 ppm. However with a higher amount of ~7 at% Ga the magnetostriction value decreases to -10 ppm. The addition of Ga in Ni does not significantly enhance the magnetostriction and for higher Ga values the magnetostriction decreases. This shows that the magnetostriction of the fcc Ni behaves completely different by substituting with the nonmagnetic element Ga.

Figure 10. Longitudinal magnetostriction of pure, polycrystalline Ni, Ni98Ga2 and Ni95Ga7 at room temperature.
2.3. Fe-Al alloys

The phase diagram of Fe-Al in the Fe-rich region is similar to that of the Fe-Ga system. As in Fe-Ga alloys, the lattice constant increases due to the non-magnetic Al substitution. As can be seen in figure 7, the magnetostriction constant $\lambda_{100}$ increases with Al substitution up to around 20 at. % Al and reaches a maximum around 150 ppm. The magnetostriction behavior of this system is similar to that of Fe-Ga alloys (see figure 7). In polycrystalline Fe$_{1-x}$Al$_x$ alloys, we reached a longitudinal magnetostriction of 60 ppm for the sample containing 19 at % Al [20].

The effect of different substituting elements is summarized in table 2. It is evident that the substitution of Ga causes the largest magnetostriction enhancement in the [100] direction. The reason for this is very probable both a structural and electronic one.

| Compound | at % in Fe | $\lambda_{100}$ ($10^6$) | $\lambda_{111}$ ($10^6$) |
|----------|------------|----------------------|----------------------|
| 0"**      | 22         | -21                  |
| 13% Ga**  | 153        | -16                  |
| 17% Ga**  | 207        | -                    |
| 16% Al*   | 86         | -2                   |
| 5% Si**   | 27         | -7                   |
| 15.6% Cr* | 51         | -6                   |
| 15.6% V*  | 43         | -10                  |

2.4. CoFe$_2$O$_4$

Co-ferrite is a ferromagnetic oxide which is non-metallic. It exhibits from all 3d-element based spinel oxides the highest magnetostriction. The value of the achievable magnetostriction depends strongly on the exact stoichiometry as well as on the production methods. Because Co-ferrite is an oxide it is produced by chemical and mechanical methods (such as sol-gel, ball milling etc) which leads finally to a powder material. In order to get bulk samples a pressing and annealing process has to be used. An improvement of the magnetostriction of polycrystalline cobalt ferrite can be obtained by magnetic annealing due to the effect of induced uniaxial anisotropy [22]. On the other hand, it is well known that the magnetic and magnetoelastic properties depend on the processing parameters [23, 24]. Nlebedim et al. [24] investigated the influence of powder compaction pressure (87 MPa ≤ P ≤ 127 MPa) and annealing conditions on the magnetostriction of CoFe$_2$O$_4$ and found the highest magnetostriction value of -200 ppm for the sample pressed at 127 MPa and annealed at 1350 °C for 24 h.

Later A.Muhamad et al investigated the influence of compaction pressure for P > 127 MPa , up to 276 MPA and found a large improvement if the pressed samples are additionally submitted to a field annealing [25]. For the sample pressed at 127 MPa and field annealed, the magnetostriction value was -400 ppm. Figure 12 shows complete measurements of all possible directions between the strain gauge and the hard (easy) axis after field annealing, for example, performed on a sample after a powder compaction pressure of 239 MPa. The open symbols represent a strain gauge placed parallel to the direction of the magnetic field annealing, and filled symbols are results by placing a strain gauge perpendicular to the magnetic field annealing direction. The magnitude of the maximum magnetostriction found by measuring longitudinal magnetostriction, applying the external magnetic field perpendicular to the direction of magnetic field annealing (hard axis), is similar to the transverse and perpendicular magnetostrictions measured by applying the field along the easy axis. Comparing with single crystal data of field annealed cobalt ferrite [26] one gets the impression that $\lambda_{\text{trans}}$ and $\lambda_{\text{perp}}$ are mainly determined by $\lambda_{110}$, whereas $\lambda_{\text{long}}$ is given by $\lambda_{100}$. So the different single crystal parameter...
contributions to the “polycrystalline” behaviour are significantly changed by the field-annealing process.

![Graph showing magnetostriction curves measured after magnetic annealing on a sample pressed at 239 MPa.](image)

**Figure 12.** (Colour online) Magnetostriction curves measured after magnetic annealing on a sample pressed at 239 MPa

3. **Summary**

Materials with high magnetostriction are summarised. The highest magnetostriction was found in $\text{TbFe}_2$ and $\text{SmFe}_2$, however Terfenol D has the advantage of a reduced magnetocrystalline anisotropy. Rare earth free alloys such as Fe-Ga and Fe-Al exhibit a lower magnetostriction however they are magnetically soft, which means they offer a high magnetostriction at low external fields. Co-ferrite is an oxide material with magnetostriction values between 100 and 400 ppm depending on the production method.

4. **References**

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