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Piezomagnetic behavior of Fe-Al-B alloys

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Abstract

For the first time, the piezomagnetic behavior of polycrystalline Fe-Al-B alloys is accessed. Piezomagnetic factors of up to $4.0 \text{ kA}\cdot\text{m}^{-1}/\text{MPa}$ were reached for an interval of applied compressive stresses between 0 to -140 MPa. The experimental results together with a powerful multiscale and biphasic modeling allowed the general understanding of the magnetostrictive and piezomagnetic behaviors of these materials. The magnetic and mechanical localizations as well as homogeneous stresses were considered in the modeling and are associated to the intrinsic presence of the Fe_2B phase. The interplay of the magnetocrystalline anisotropy, initial susceptibility, saturation magnetostriction and texture were quantified by the model and compared to the experimental results. An improvement of the piezomagnetic factor to $15 \text{ kA}\cdot\text{m}^{-1}/\text{MPa}$ is predicted, for an alloy containing 20% of aluminum, by getting an adequate texture near $\langle 100 \rangle$ directions.

Keywords: iron-boron alloys, iron-aluminum alloys, piezomagnetism, magnetostriction, biphasic model, magnetomechanics

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1. Introduction

For devices that utilize magnetomechanical behavior for their operation, it is necessary to have materials that present large coupling between mechanical and some magnetic properties. Specifically, the variation of the magnetization M with respect to the applied uniaxial stress σ (*i.e.* $dM/d\sigma$) should be high for an appropriate sensitivity [1]. The coupling leads to the thermodynamic condition that $\mu_0 \cdot dM/d\sigma = d\lambda/dH$ [2]: λ is the magnetostriction "measured" in the magnetic measurement direction, H is the magnetic field and typical desirable $d\lambda/dH$ values are greater than 1.0 nm/A [1], which is equivalent to 0.83 kA.m⁻¹/MPa for $dM/d\sigma$.

Rare earth elements (Tb, Dy) are often used as secondary elements in alloys that are employed as magnetic sensors or actuators, because they exceptionally enhance the magnetomechanical properties of these materials. For example, Terfenol D presents very high magnetostriction values, typically 1000 ppm at room temperature [3, 4]. The sensitivity of the magnetostriction and magnetization to the application of mechanical stresses, called the magnetomechanical or piezomagnetic behavior is very high for these materials. The variation rate of deformation to the applied field measured at a compression of -9 MPa is 16 nm/A for a bias field of 22 kA/m [4]. Likewise, the sensitivity of the magnetization with respect to the applied stress estimated from the data in [4] is ~ 15 kA.m⁻¹/MPa for an absolute maximum compression value of -25 MPa and 120 kA/m bias field. However, due to the increase of price and limited availability of these rare earth elements, the development of rare earth free alloys is relevant.

26

27 Fe-Al alloys demonstrated to be interesting candidates [5]. Fe-Al binary
28 alloys are monophasic for aluminum contents lower than 17% (atomic) and
29 in this range only the α -phase occurs that is a disordered bcc phase with
30 A2 structure. α -phase of pure iron has saturation magnetization of 1.7×10^6
31 A/m and a anisotropy constant $K_1 = 4.8 \times 10^4 \text{ J.m}^{-3}$ (cubic symmetry).
32 Both properties decrease monotonically with increasing aluminum content
33 until 17%. In the range $17 < \% \text{ Al} < 30$ a partial ordering can take place de-
34 pending on the cooling rate of the material and the ordered Fe_3Al phase with
35 D03 structure can coexist with the α -phase [6, 7]. Fe_3Al phase is also fer-
36 romagnetic and has a saturation magnetization of $8.03 \times 10^5 \text{ A/m}$ and Curie
37 temperature $T_C = 440^\circ\text{C}$ [8]. The anisotropy constant K_1 decreases from
38 $2.3 \times 10^4 \text{ J.m}^{-3}$ for $\sim 17\%$ Al to zero for 22% Al. From 22% Al to 30% Al,
39 $K_1 < 0$ and has a minimum of $-0.9 \times 10^4 \text{ J.m}^{-3}$ for 26% Al [6]. K_1 is null for
40 30% of Al [6].

41

42 Recently an important increase of the Fe-Al alloys magnetostriction has
43 been observed due to the addition of boron [9]. The introduction of boron to
44 Fe-Al alloys leads to a two or three phases alloy depending on the aluminum
45 content. The boron added to Fe-Al alloys is not soluble in the cubic lattices,
46 but causes the formation of the Fe_2B phase [9]. The as cast microstructure of
47 Fe-Al-B alloys is a dendritic solidified structure in which the dendrites are the
48 Fe-Al cubic phases and the inter-dendritic region is a micro-constituent com-
49 posed of Fe-Al and Fe_2B phases. A subsequent annealing generates spheroid
50 shaped particles of the Fe_2B phase distributed through the Fe-Al matrix [10].

51 Fe₂B is also a ferromagnetic phase, which structural, magnetic and magne-
52 tostrictive properties are detailed in Appendix A [11, 12]. This material
53 exhibits a positive longitudinal magnetostriction and leads to an enhance-
54 ment of magnetic and magnetostrictive behavior of the composite formed by
55 Fe-Al matrix and Fe₂B phase.

56 The influence of boron content has been extensively discussed in a previ-
57 ous work [10]. In the present work, Fe-Al-B were chosen such that the boron
58 atomic fraction was kept constant. The study comprises of four alloys Fe-Al-
59 B alloys, three with fixed B content with composition in the range of 14 <
60 %Al < 22 (at.%) and one sample without aluminum. Magneto-mechanical
61 measurements and microstructure assessment are the tools used to quantify
62 Fe-Al-B alloys piezomagnetic performance. Multiscale modeling consider-
63 ing biphasic material allowed the understanding of the magnetostrictive and
64 piezomagnetic behavior.

65 **2. Expected role of Fe₂B phase in Fe-Al-B ternary alloy**

66 *2.1. Composite effect*

67 The influence of the Fe₂B phase inside the Fe-Al matrix can be understood
68 as a composite effect. Indeed the presence of two different phases creates a
69 local perturbation called demagnetizing field in magnetism and residual stress
70 in mechanics [13]. In this condition, the local fields are not generally the
71 same as the mean fields. Their calculation requires a mathematical operation
72 called localization. A medium composed of i phases of volume fraction f_i is
73 considered. The local magnetic field applied to the phase i is a complex
74 function of macroscopic field \vec{H} and the properties of the mean medium. In

75 the case of spheroidal inclusion [14], the field is demonstrated as homogeneous
 76 on each phase. Considering on the other hand a linear susceptibility of
 77 average medium χ_m , the local magnetic field in the phase i is given by:

$$\vec{H}_i = \vec{H} + \frac{1}{3 + 2\chi_m}(\vec{M} - \vec{M}_i) = \vec{H} + \vec{H}_i^d \quad (1)$$

78 where \vec{M} is the average magnetization, \vec{M}_i is the local magnetization.
 79 \vec{H}_i^d is the so called demagnetizing field acting on phase i . The extension to
 80 nonlinear behavior involves to use the sequent susceptibility for the definition
 81 of χ_m .

$$\chi_m = \|\vec{M}\|/\|\vec{H}\| \quad (2)$$

82 Averaging operations lead to

$$\vec{H} = \langle f_i \vec{H}_i \rangle \quad \text{and} \quad \vec{M} = \langle f_i \vec{M}_i \rangle \quad (3)$$

83 The solution of Eshelby's problem is the basis of the modelling of hetero-
 84 geneous media's behavior in mechanics. Its formulation has been extended
 85 by Hill considering a deformable matrix [15]. Equation 4 gives the stress field
 86 within the inclusion i submitted to a macroscopic stress σ . ϵ_i is the total
 87 strain tensor of the inclusion considered. ϵ is the average total strain tensor
 88 over the volume.

$$\sigma_i = \sigma + \mathbb{C}^*(\epsilon - \epsilon_i) = \sigma + \sigma_i^r \quad (4)$$

89 \mathbb{C}^* is the Hill's constraint tensor depending on the distribution and shape
 90 of inclusions and on the stiffness properties of materials. σ_i^r is the so called

91 residual stress tensor acting on inclusion i . If homogeneous isotropic elas-
 92 tic properties (Young's modulus E and Poisson's ratio ν) and additivity of
 93 deformation (total deformation = elastic deformation + magnetostrictive de-
 94 formation) are considered, equation 4 can be simplified in:

$$\boldsymbol{\sigma}_i = \boldsymbol{\sigma} + \frac{E(7 - 5\nu)}{15(1 - \nu^2)}(\boldsymbol{\epsilon}^\mu - \boldsymbol{\epsilon}_i^\mu) \quad (5)$$

95 where $\boldsymbol{\epsilon}_i^\mu$ and $\boldsymbol{\epsilon}^\mu$ denote the local and average magnetostriction strain
 96 tensor respectively.

97 Averaging operations lead to:

$$\boldsymbol{\sigma} = \langle f_i \boldsymbol{\sigma}_i \rangle \quad \text{and} \quad \boldsymbol{\epsilon}^\mu = \langle f_i \boldsymbol{\epsilon}_i^\mu \rangle \quad (6)$$

98 2.2. Application to α phase – Fe_2B phase composite

99 This approach is applied to α phase – Fe_2B phase microstructure with f_α
 100 and f_{Fe_2B} the volume fractions of α phase and Fe_2B phase respectively. The
 101 problem is next simplified in a 1D problem (all quantities measured along x
 102 axis for example), the average magnetic and magnetization fields verify:

$$H = f_\alpha H_\alpha + f_{Fe_2B} H_{Fe_2B} \quad (7)$$

and

$$M = f_\alpha M_\alpha + f_{Fe_2B} M_{Fe_2B} \quad (8)$$

103 The average uniaxial stress and longitudinal magnetostriction strain (λ)
 104 verify:

$$\sigma = f_\alpha \sigma_\alpha + f_{Fe_2B} \sigma_{Fe_2B} \quad (9)$$

$$\lambda = f_{\alpha}\lambda_{\alpha} + f_{Fe_2B}\lambda_{Fe_2B} \quad (10)$$

105 The magnetic field inside the α phase is given by:

$$H_{\alpha} = H + \frac{1}{3 + 2\chi_m}(M - M_{\alpha}) \quad (11)$$

106 Because Fe₂B phase is very soft (until magnetic rotation out of the easy
 107 axis - see Appendix A), $M_{Fe_2B} > M$ so that, due to averaging, $M_{\alpha} < M$.
 108 The magnetic field in the α phase is consequently higher than the average
 109 magnetic field ($H_{\alpha} > H$), enhancing both magnetization and magnetostric-
 110 tion.

111

112 The stress field inside the α phase is given by:

$$\sigma_{\alpha} = \sigma + \frac{E(7 - 5\nu)}{15(1 - \nu^2)}(\lambda - \lambda_{\alpha}) \quad (12)$$

113 Due to soft magnetic properties of Fe₂B phase, longitudinal magnetostric-
 114 tion is higher than average magnetostriction at low magnetization level ($\lambda_{Fe_2B} >$
 115 λ), so that, due to averaging, $\lambda_{\alpha} < \lambda$. The stress field in the α phase is
 116 consequently higher than the average stress field ($\sigma_{\alpha} > \sigma$). Considering an
 117 unloaded specimen ($\sigma=0$), a positive stress is created inside the matrix coun-
 118 terbalanced by a negative stress field in the Fe₂B phase. The longitudinal
 119 magnetostriction being positive for α -phase, the positive residual stress leads
 120 to enhanced magnetization and magnetostriction properties as well.

121

122 This simplified approach allows to explain in few words the composite
 123 effect that occurs for Fe-Al-B alloys. It must nevertheless be nuanced be-

124 cause of the non-linearity of both magnetic and magnetostrictive behaviors,
125 multiaxiality of stress and texture effects (isotropic distribution of phase is
126 assumed). An experimental approach is required to underline these limits.

127 **3. Experimental Methods**

128 The alloys were produced by arc melting in argon atmosphere and re-
129 melted in a high vacuum furnace inside an alumina tube of 25 mm of di-
130 ameter. The bars obtained had around 110 mm of length and 280 g (figure
131 1). Plates of thickness of 3 mm were cut from the center in the longitudinal
132 direction of the bars by electro erosion. The plates were annealed in inert at-
133 mosphere at 1100°C during 24 h and quenched in water. These plates are the
134 samples studied in this work and had their microstructure and piezomagnetic
135 behavior analyzed.

136 The microstructure of the samples were characterized by scanning elec-
137 tron microscopy: imaging by secondary electrons, composition by EDX and
138 crystallographic texture by EBSD. The samples preparation for SEM obser-
139 vation consists of mechanical and subsequent electro polishing. The EBSD
140 measurements were made in areas of just about 1.2 mm² in both sides of the
141 plates, at the positions where the strain gauges were glued. The anhysteretic
142 piezomagnetic behavior measurement set up acquires the magnetization (M)
143 and longitudinal magnetostriction (λ) under different levels of applied stress
144 varying the magnetic field. For each applied magnetic field, the sample is
145 demagnetized [16]. The active ranges of stresses and magnetic field are -140
146 $\leq \sigma \leq 50$ MPa and $0 < H < 17$ kA/m, respectively.

147



Figure 1: Typical Fe-Al-B alloy bar of 25 mm in diameter and 110 mm in length

148 The system consists in a sample plate positioned inside a primary cylin-
149 drical coil having 85 turns. Two soft ferrite U-yokes close the magnetic circuit
150 and one strain gauge is glued in each side of the plate to acquire the magne-
151 tostriction using also a Wheatstone bridge. To measure the magnetization,
152 a pick up coil of 50 turns is wound in the central region of the plates close
153 to the position of the strain gages. Hydraulic jaws of the tensile-compressive
154 machine used to apply the stress grab the system by the sample. The con-
155 centration of boron was determined by atomic absorption in a spectrometer
156 PerkinElmer, model Analyst 800, with an integrated system of graphite fur-
157 nace and flame. Samples of ~ 0.1 g were analyzed.

158 4. Experimental results and related modeling

159 4.1. Microstructure characterization and associated analytical modeling

160 Figure 2 shows the typical microstructure obtained for the annealed Fe-
161 Al-B alloys. In figure 2 (a) it is possible to verify that the fingerprint of
162 the dendritic solidified microstructure is still present even after the anneal-
163 ing. The Fe_2B lamellas localized in the interdendritic microconstituent (not
164 shown) in the as cast alloys consolidated into a bulk phase as shown in more
165 details in figures 2(b) and (c). The matrix is the Fe-Al binary alloy as dis-
166 cussed previously, but the Fe_2B phase did not form exactly the same spheroid
167 shaped particles as in the previous studies [10]. This may be related to the
168 difference in mass of the samples and thus to different thermal conditions.
169 The mass of the samples in the previous study were 4 g, while it was close
170 to 30 g in the present study.

171

172 The measured EDS aluminum content of the matrix of each of the three
173 alloys in atomic percentage and the respective standard deviation are 14.5
174 $\pm 0.2\%$, $20.2 \pm 0.5\%$ and $22 \pm 0.4\%$. From now on, we are going to label
175 the samples of these alloys as $A1 - 14Al$, $A2 - 20Al$ and $A3 - 22Al$ and the
176 one without aluminum ($x=0$) $A0 - FeB$. Chemical analyses demonstrate
177 that the mean quantity of boron in the samples is 1.6% and the aluminum
178 contents agree with EDS measurements.

179

180 The volume fraction of the phase Fe_2B was evaluated by image analyses
181 and also from EBSD results (associated to low confidence index - CI). The
182 values obtained are very close to 13% . This result is consistent with chemical

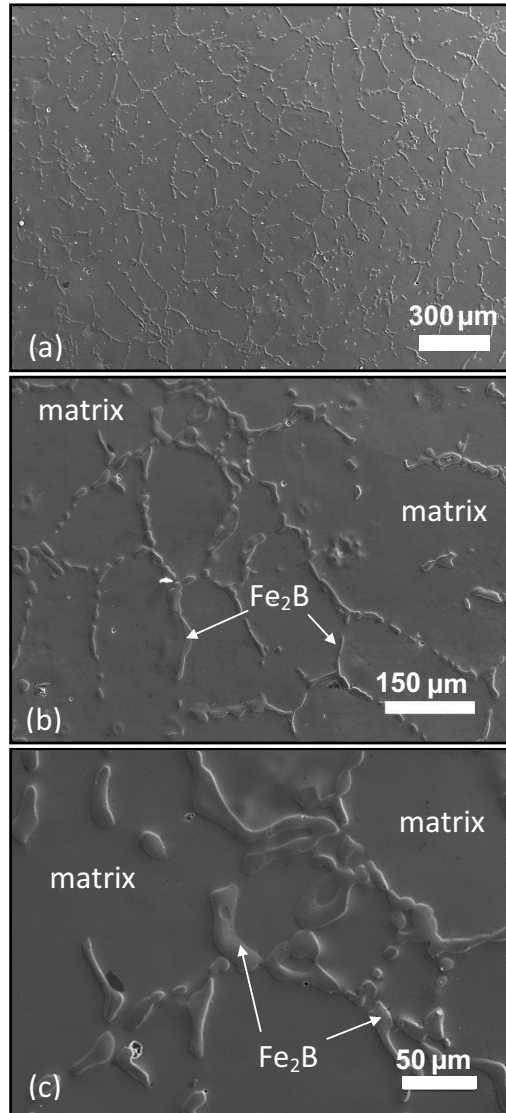


Figure 2: SEM secondary electrons images of the typical microstructure obtained for the Fe-Al-B and Fe-B annealed alloys.

184 Figure 3 shows a typical result obtained from EBSD measurement (A1 –
185 14Al sample - IPF along *X* direction), illustrating the large grain size and

186 indexing errors associated to the Fe_2B phase.

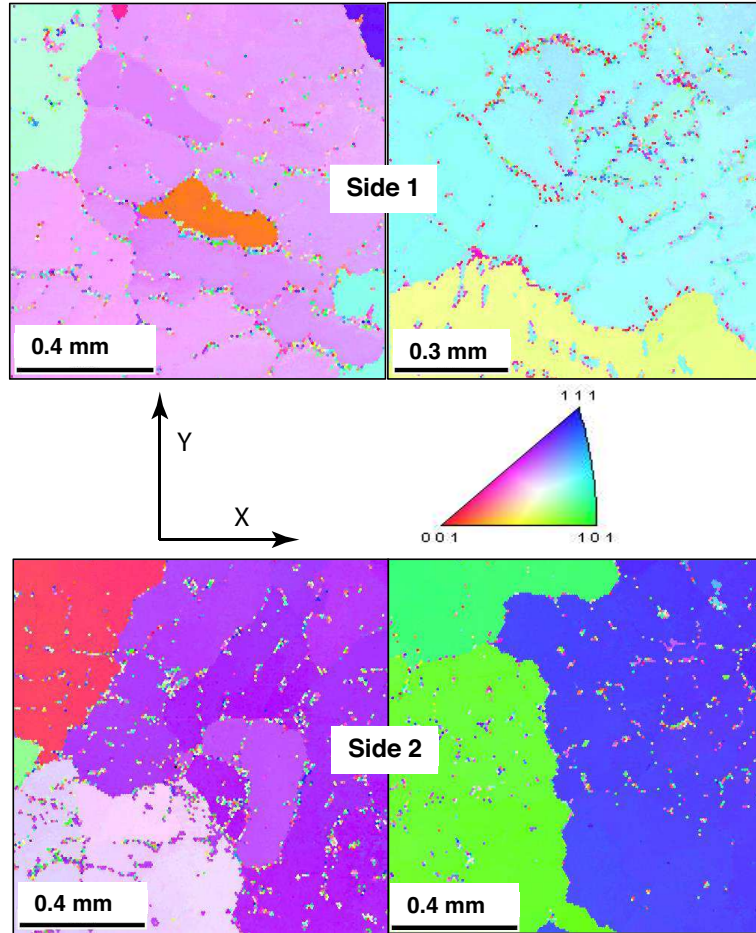


Figure 3: Inverse pole figures (IPF) obtained for the alloy $A1 - 14Al$ giving the crystallographic direction in the direction of measurement (X). Above are two areas analyzed in one side of the plate and below the other two areas of the other side.

187 EBSD data are composed of Euler angles (ϕ_1, θ, ϕ_2) giving the angular
188 position of a point i (x_i, y_i, z_i) inside the global frame (X, Y, Z) where X
189 indicates the direction of magnetic and mechanical loading. It is possible to
190 calculate the loading direction inside the crystal frame at each EBSD point

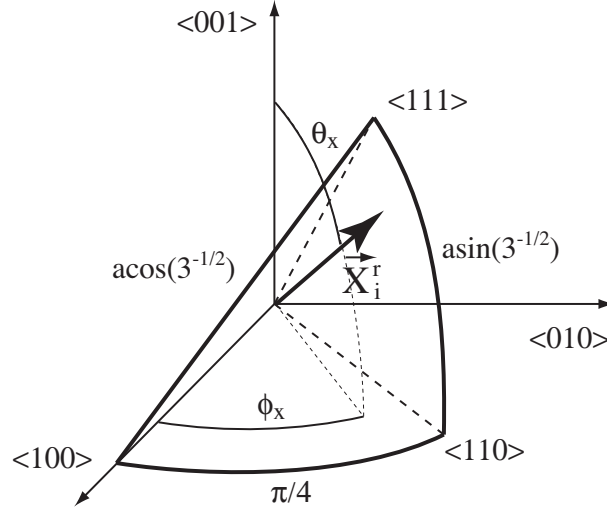


Figure 4: Loading direction \vec{X}_i^r placed inside the standard triangle defined $\langle 100 \rangle$, $\langle 110 \rangle$ and $\langle 111 \rangle$ crystallographic directions after permutation and associated spherical angles.

191 i (becoming \vec{X}_i) of dominant α -phase (Fe_2B phase is not considered). This
 192 direction can be expressed inside the standard triangle defined between $[100]$,
 193 $[110]$ and $[111]$ crystallographic directions (figure 4) thanks to appropriate
 194 permutation operations since the cubic symmetry is considered for the matrix
 195 [17]. The corresponding rotated vectors \vec{X}_i^r are averaged over the points i
 196 leading to an averaging direction $\langle \vec{X}^r \rangle$:

$$\langle \vec{X}^r \rangle = \frac{1}{N} \sum_{i=1}^N \vec{X}_i^r \quad (13)$$

197 The closest associated Miller index $\langle uvw \rangle$ to $\langle \vec{X}^r \rangle$ direction is
 198 finally estimated. The average orientation results are shown in Table 1 for
 199 all samples.

200 As explained in [18], the behavior of an isotropic polycrystal is necessar-

Table 1: Average orientation of the α phase calculated from EBSD results of Fe-Al-B and Fe-B samples.

Sample	$A0 - FeB$	$A1 - 14Al$	$A2 - 20Al$	$A3 - 22Al$
Average orientation	$\langle 421 \rangle$	$\langle 321 \rangle$	$\langle 420 \rangle$	$\langle 421 \rangle$

201 ily given by a loading along a specific direction inside the standard triangle.
 202 Because behaviors are not linear, this direction is not the average direction
 203 and may change with stress or magnetic field level. Nevertheless it is possi-
 204 ble, as first approximation, to consider that an isotropic behavior is roughly
 205 obtained when the loading is corresponding to the average direction of the
 206 standard triangle. In case of cubic symmetry, this direction is defined by
 207 spherical angles $(\phi_X, \theta_X) = (38.81^\circ, 77.54^\circ)$, that is close to $\langle 431 \rangle$ di-
 208 rection. Samples $A0 - FeB$, $A1 - 14Al$ and $A3 - 22Al$ may exhibit some
 209 barely isotropic behavior since their orientations are close to $\langle 431 \rangle$. Sam-
 210 ple $A2 - 20Al$ should exhibit on the contrary a dominant effect of $\langle 100 \rangle$
 211 direction.

212

213 A theoretical estimation of the magnetostriction without application of
 214 external stresses and using the average orientation results is possible after
 215 several approximations. Assumptions concern equiprobable initial distribu-
 216 tion of domains, homogeneous magnetic and stress fields within the sample.
 217 The approach to saturation is on the other hand considered as only depend-
 218 ing on anisotropy strength of the material. Indeed α phase could present a
 219 low magnetocrystalline constant K_1 that would lead to concomitant rotation

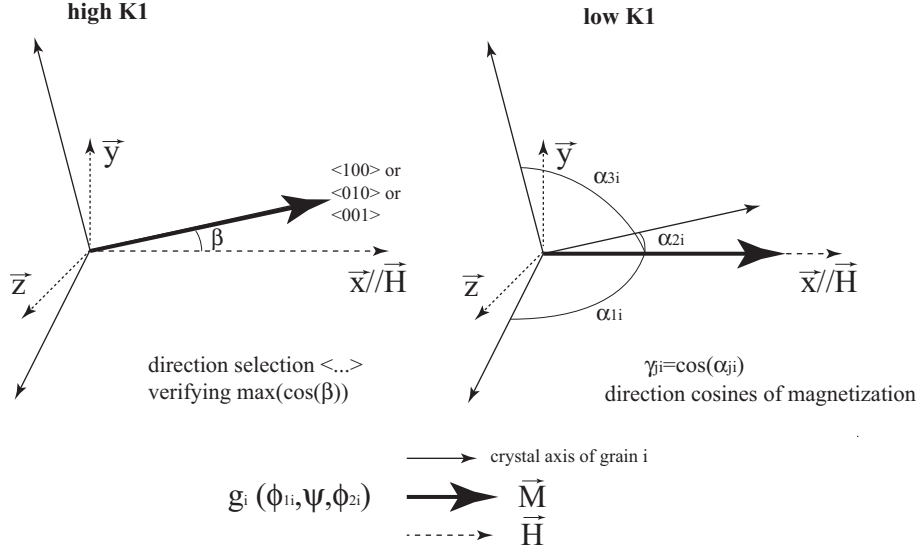


Figure 5: Possible positions of magnetization vector at a position i for high and low magnetocrystalline constant strengths.

220 and domain wall motion. For high K_1 values, the domain wall motion occurs
 221 before rotation and actually in the present case the rotation is supposed to
 222 be unachieved.

223

224 Figure 5 shows the two possible positions of magnetization vector at a
 225 given EBSD point i belonging to the α -phase matrix. These two situations
 226 correspond to two different magnetization states under applied field associ-
 227 ated with two possible magnetocrystalline constant strengths.

228 For both situations, two estimations of average longitudinal magnetostric-
 229 tion strain λ are made: $\lambda = \lambda_s$ the saturation magnetostriction if K_1 is "low";
 230 $\lambda = \lambda_{max}$ the maximal magnetostriction if K_1 is high. Each EBSD position i
 231 is defined by its Euler angles $(\phi_1^i, \theta^i, \phi_2^i)$ in the macroscopic frame (X, Y, Z)

232 as shown in figure 5. The magnetic field is applied along X axis. At satu-
 233 ration or considering a low K_1 situation, the magnetization at high field is
 234 considered to be aligned with the magnetic field direction. Direction cosines
 235 of the magnetization can be deduced because the direction X is known. For
 236 a "high" K_1 situation, the magnetic field is supposed not strong enough so
 237 that the magnetization direction remains in the direction of one of the three
 238 easy axes of the cubic symmetry. The selected direction is obtained by min-
 239 imizing the angle β , which gives the closest direction to the applied field
 240 direction X . In both cases, magnetostriction tensor is calculated at each po-
 241 sition i . Equations (14) and (15) give this tensor for low and high K_1 values,
 242 respectively.

$$\epsilon_{\mu}^i = \frac{3}{2} \begin{pmatrix} \lambda_{100}^{\alpha}(\gamma_1^2 - \frac{1}{3}) & \lambda_{111}^{\alpha}\gamma_1\gamma_2 & \lambda_{111}^{\alpha}\gamma_1\gamma_3 \\ \lambda_{111}^{\alpha}\gamma_1\gamma_2 & \lambda_{100}^{\alpha}(\gamma_2^2 - \frac{1}{3}) & \lambda_{111}^{\alpha}\gamma_2\gamma_3 \\ \lambda_{111}^{\alpha}\gamma_1\gamma_3 & \lambda_{111}^{\alpha}\gamma_2\gamma_3 & \lambda_{100}^{\alpha}(\gamma_3^2 - \frac{1}{3}) \end{pmatrix} \quad (14)$$

$$\epsilon_{\mu}^i = \begin{pmatrix} \lambda_{100}^{\alpha} & 0 & 0 \\ 0 & -\frac{\lambda_{100}^{\alpha}}{2} & 0 \\ 0 & 0 & -\frac{\lambda_{100}^{\alpha}}{2} \end{pmatrix} \quad (15)$$

243 λ_{100}^{α} and λ_{111}^{α} are the independent magnetostriction constants of the α
 244 phase cubic single crystal in the directions $\langle 100 \rangle$ and $\langle 111 \rangle$. γ_j for j
 245 =1, 2 and 3 are the direction cosines of the magnetization as illustrated in
 246 figure 5.

247 As discussed in Appendix A Fe_2B phase exhibits an easy magnetization
 248 plane that leads in a first approximation to an enhancement of both magnetic
 249 and magnetostrictive properties. This effect is isotropic since the distribu-

250 tion of Fe₂B phase is considered as isotropic. The averaging operations are
 251 made in the framework of homogeneous stress hypothesis: over the EBSD
 252 measurement points i first, then considering Fe₂B phase of volume fraction
 253 f , as indicated in equations (16) and (17).

$$\boldsymbol{\epsilon}_\mu^\alpha = \langle \boldsymbol{\epsilon}_\mu^i \rangle \quad (16)$$

$$\boldsymbol{\epsilon}_\mu = f \boldsymbol{\epsilon}_\mu^{Fe_2B} + (1 - f) \boldsymbol{\epsilon}_\mu^\alpha \quad (17)$$

The isotropic distribution of Fe₂B with high intensity uniaxial anisotropy implies that (see Appendix A for detailed calculations) :

$$\boldsymbol{\epsilon}_\mu^{Fe_2B} = \frac{1}{2} \begin{pmatrix} \lambda_{100}^{Fe_2B} & 0 & 0 \\ 0 & -\frac{\lambda_{100}^{Fe_2B}}{2} & 0 \\ 0 & 0 & -\frac{\lambda_{100}^{Fe_2B}}{2} \end{pmatrix} \quad (18)$$

254 leading to the final expression of average magnetostriction strain tensor
 255 $\boldsymbol{\epsilon}_\mu$. Therefore, an estimation of average magnetostriction strain $\boldsymbol{\epsilon}_\mu$ is obtained
 256 by using the expression of $\boldsymbol{\epsilon}_\mu^i$ from equation (14) in the definition of $\boldsymbol{\epsilon}_\mu^\alpha$ for
 257 low K_1 . For high K_1 the expression of $\boldsymbol{\epsilon}_\mu^i$ from equation (15) is used. The
 258 magnetostriction strain measured in the direction of applied field is finally
 259 given by:

$$\lambda = f \frac{\lambda_{100}^{Fe_2B}}{2} + (1 - f)^t \vec{X} \boldsymbol{\epsilon}_\mu^\alpha \vec{X} \quad (19)$$

260 Some hypotheses were made concerning the crystallographic ordering of
 261 the Fe-Al matrix. The A2 structure (disordered α phase) is chosen as the
 262 matrix of samples A1 – 14Al and A2 – 20Al. For the matrix of sample A3 –

263 22Al, a mix of A2 and D03 structures (ordered phase) with the proportion
264 1:1 is chosen. The physical constants of Fe₂B phase are $\lambda_{100}^{Fe_2B} = 20$ ppm [9].
265 The volume fraction of Fe₂B already previously presented is $f = 0.13$. Table
266 2 shows the values of the Fe-Al matrix physical constants ($\lambda_{100}^\alpha, \lambda_{111}^\alpha, K_1$ [6]),
267 as well as the theoretical and experimental values of the magnetostriction
268 measured in the direction of the applied magnetic field.

Table 2: Physical constants of the Fe-Al matrix [6] complemented by theoretical and experimental parallel magnetostriction measured in the direction of the applied magnetic field (shown in the last two columns).

Sample \ Param.	Structure	λ_{100}^α	λ_{111}^α	K_1	$\lambda^{calc.}$	$\lambda^{exp.}$
Unit	-	ppm	ppm	kJ.m^{-3}	ppm	ppm
A0 – FeB	A2	21	-21	48 (high)	8.6	5.8
A1 – 14Al	A2	80	-3	31.3 (high)	21.8	17.3
A2 – 20Al	A2	79	4	15.5 (high)	32	25
A3 – 22Al	A2/D03	76/84	7/26	11.5/0 (high/low)	33.5	34.2

269 A relatively good agreement is obtained between theoretical and exper-
270 imental magnetostriction, meaning that orientations are representative of
271 samples. We suppose that except considering a $\langle 100 \rangle$ oriented sample,
272 the Fe-Al-B alloys of the present work are not good candidate for actuation.
273 The piezomagnetic behavior (magnetization *vs* stress) may be more interest-
274 ing since the performance of such application would depend not only on the
275 magnetostriction magnitude but also on the sensitivity of magnetostriction

276 to magnetic field ($d\lambda/dH$).

277 4.2. Magnetic and piezomagnetic behavior

278 Figure 6 shows the measured curves M vs. H for fixed values of stress
279 σ for the four samples. The stress magnitude used for $A0 - FeB$ sample
280 is lower than for others samples due to its much lower yield stress. As ex-
281 pected, the magnetization of the sample $A3 - 22Al$ is smaller than $A2 - 20Al$
282 that is smaller than $A1 - 14Al$ and finally to $A0 - FeB$, due to the increase
283 of aluminum content, a nonmagnetic element. It must be noticed that the
284 occurrence of the Fe_3Al (D03) phase would cooperate also to decrease the
285 saturation magnetization in sample $A3 - 22Al$ [19].

286 The values of the initial susceptibility are about 1500, 800, 600 and 500 for
287 the correspondingly samples $A0 - FeB$, $A1 - 14Al$, $A2 - 20Al$ and $A3 - 22Al$.
288 This trend seems in contradiction with the decreasing of magnetocrystalline
289 constant K_1 with increasing aluminum content. It is on the contrary in ac-
290 cordance with a lower mobility of domain wall due to an increased pinning
291 effect. Indeed magnetostriction is strongly enhanced with aluminum content
292 that increases the magnetoelastic interactions (NB: the reduction of magne-
293 tostriction is a well known technique employed to increase the permeability
294 of materials - such for permalloys - [20]).

295 The M vs. σ curves at constant H for each alloy were built from the
296 data of figure 6 for six fixed values of magnetic field in the range of 1-15
297 $kA.m^{-1}$ and are depicted in figure 7. Subsequently, from these M vs. σ
298 curves the respective sensitivity $dM/d\sigma|_H$ is calculated and figure 8 displays
299 the associated plots for each sample at the same values of fixed applied field.

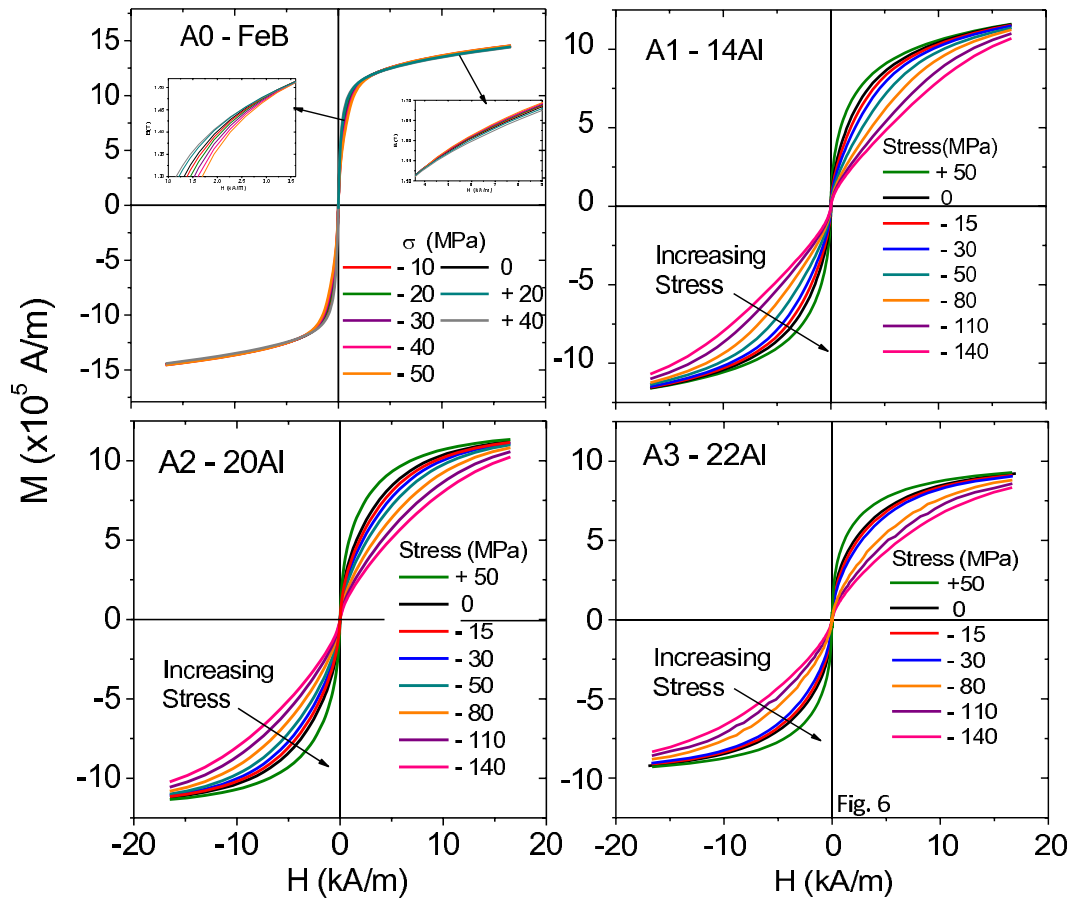


Figure 6: Magnetization as a function of the applied magnetic field for fixed values of stresses for all samples.

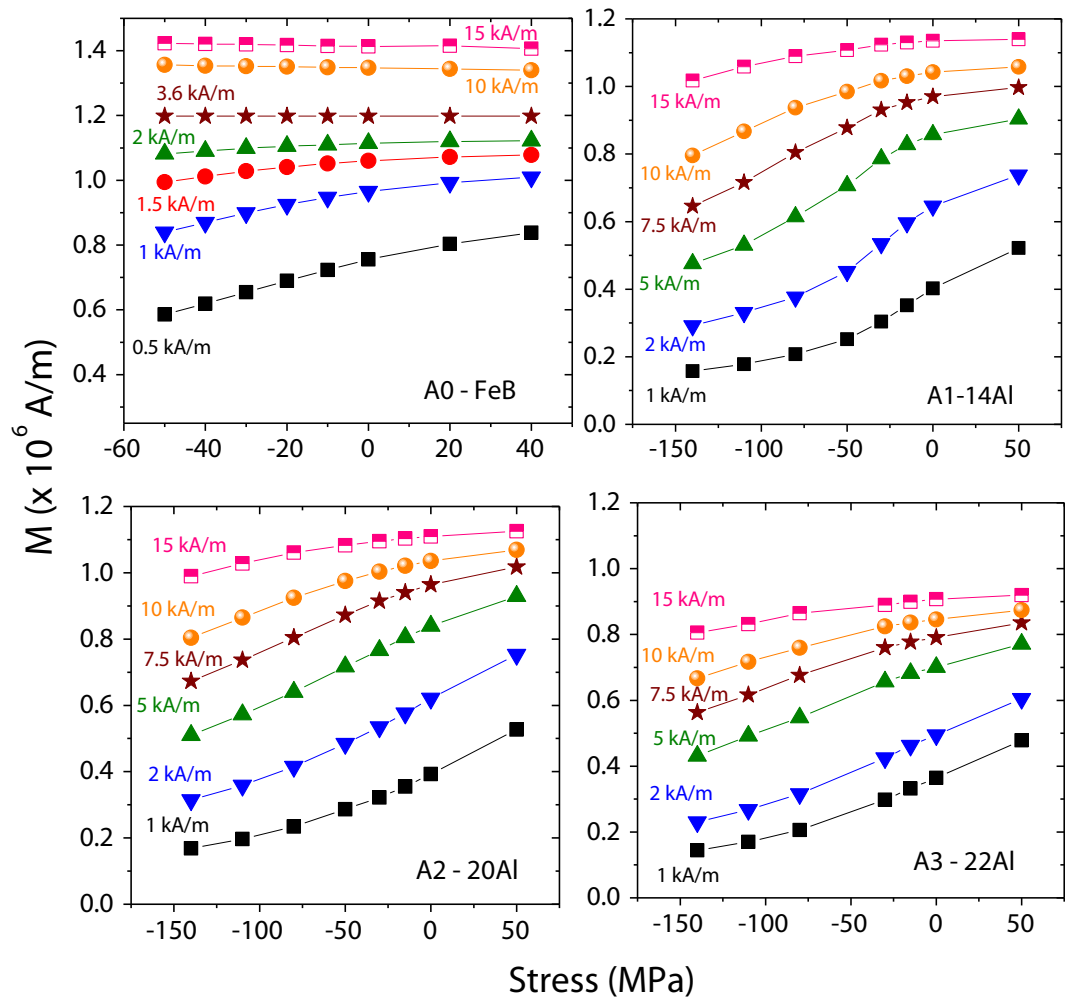


Figure 7: Magnetization as a function of applied stress σ for all samples at fixed values of magnetic field, ($1 \leq H \leq 15$ kA/m).

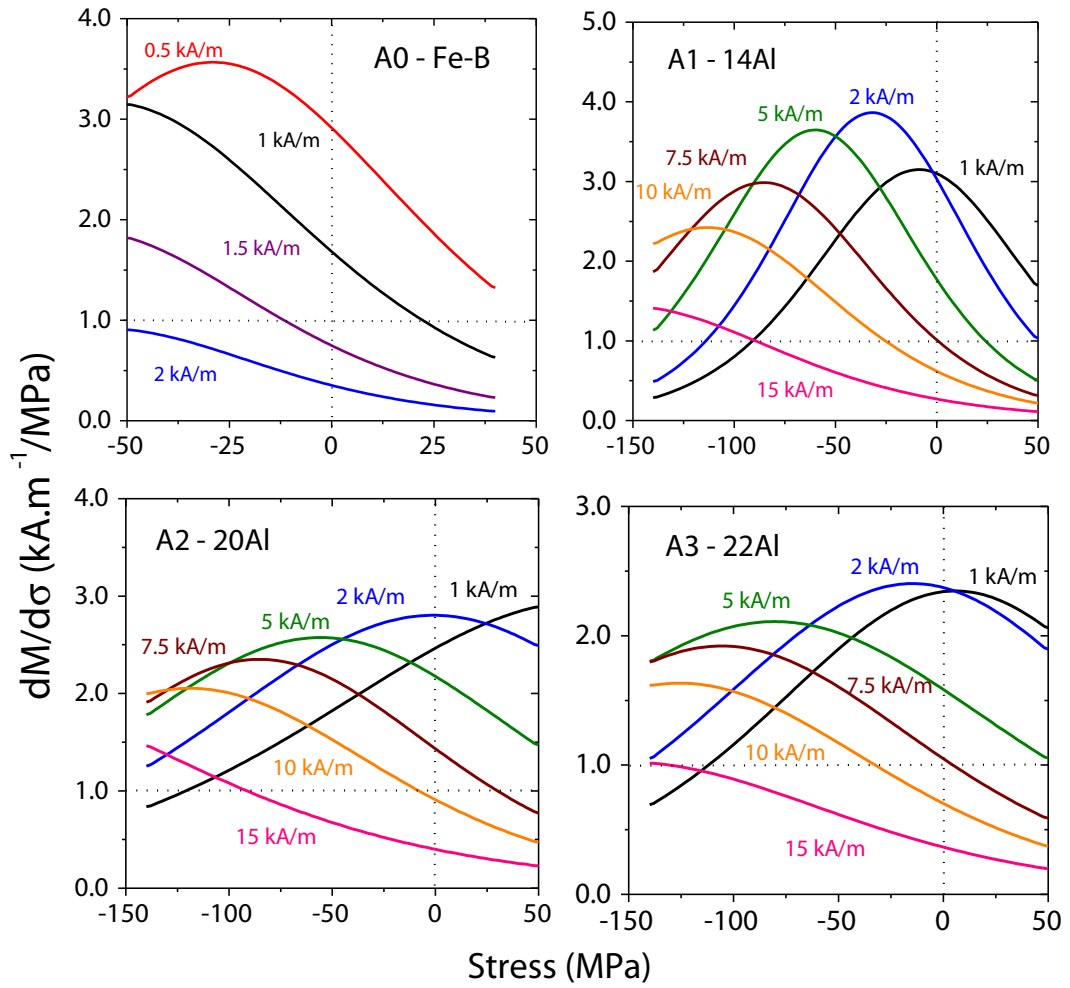


Figure 8: Piezomagnetic sensitivity $dM/d\sigma|_H$ vs. σ for all samples at fixed values of magnetic field, ($1 \leq H \leq 15$ kA/m).

300 By covering all the compression range from 0 to -140 MPa and with sen-
301 sitivity greater than $1.0 \text{ kA.m}^{-1}/\text{MPa}$, there are some different possibilities
302 for using the Fe-Al-B alloys as force sensors. Two of them are described
303 next. For a compression sensor that should work in constant bias field, for
304 all the three alloys, the best bias field is 5.0 kA.m^{-1} . Among them, sample
305 *A2-20Al* exhibits the largest sensitivity for the entire compression range, be-
306 tween 1.8 and $2.6 \text{ kA.m}^{-1}/\text{MPa}$, for a magnetization variation range of about
307 330 kA.m^{-1} . Although sample *A1-14Al* has a maximum sensitivity at -30
308 MPa, *i.e.* $3.9 \text{ kA.m}^{-1}/\text{MPa}$. Its sensitivity decreases to $0.5 \text{ kA.m}^{-1}/\text{MPa}$ at
309 -140 MPa.

310 A dispositive constructed for varying bias field would employ the interval of
311 $2.0 \leq H \leq 10 \text{ kA.m}^{-1}$ and in this case, the best alloy would be *A1-14Al*,
312 with sensitivity in the range of 2.2 to $3.9 \text{ kA.m}^{-1}/\text{MPa}$ for a magnetization
313 variation range of about 710 kA.m^{-1} . Although Terfenol-D has a very high
314 sensitivity of $\sim 15 \text{ kA.m}^{-1}/\text{MPa}$ [4], this material application is restricted to
315 a maximum compression of -25 MPa due to its brittleness. In addition, the
316 bias field is close to 120 kA.m^{-1} that is one order of magnitude higher than
317 the optimal bias field obtained for Fe-Al-B alloys.

318 Results are interesting and some applicability can arise. It is nevertheless not
319 fully understood why alloy *A1-14Al* exhibits a higher piezomagnetic sensi-
320 tivity peak than *A2-20Al* or *A3-22Al* although its magnetostriction level is
321 the lowest. Many parameters like saturation magnetization level, anisotropy
322 constant, magnetostriction constants, crystallographic texture, and interac-
323 tion between matrix and Fe_2B phase must be considered. On the other hand,
324 an increasing of piezomagnetic behavior may be surely reached by a better

325 choice of crystallographic texture for example. A modeling involving these
 326 parameters is presented hereafter.

327

328 4.3. Multidomain modeling

329 A two-scale reversible modeling of the magneto-mechanical behavior of
 330 each phase is proposed mixing the propositions of [18] and [21]. This model
 331 comes from a simplification of the so-called multiscale model [14] where only
 332 domain and grain scales are considered. Even if cubic symmetry is consid-
 333 ered, easy directions are not defined *a priori* allowing a statistical descrip-
 334 tion of domain distribution (N domains are considered) and avoiding energy
 335 minimization. At each domain ϕ of direction $\vec{\gamma}_\phi = \gamma_i \vec{e}_i$ corresponds a mag-
 336 netization vector $\vec{M}_\phi = M_s \vec{\gamma}_\phi$, and a magnetostriction tensor ϵ_ϕ^μ (previously
 337 defined by equation (14)). This single crystal is considered as submitted to
 338 a magnetic field H and/or uniaxial stress σ applied in direction \vec{X} (see table
 339 1) defined by angles ϕ_X and θ_X of the spherical frame (figure 4). It is consid-
 340 ered that the contribution to the free energy of a magnetic domain W_ϕ are
 341 the magnetostatic energy W_ϕ^H , the magnetocrystalline energy W_ϕ^K and the
 342 magnetoelastic energy W_ϕ^σ (20).

$$W_\phi^H = -\mu_0 \vec{H} \cdot \vec{M}_\phi \quad W_\phi^\sigma = -\boldsymbol{\sigma} : \boldsymbol{\epsilon}_\phi^\mu \quad W_\phi^K = K_1((\gamma_1\gamma_2)^2 + (\gamma_2\gamma_3)^2 + (\gamma_1\gamma_3)^2) \quad (20)$$

343 The volume fraction f_ϕ of a domain is calculated as function of the free
 344 energies using the statistical Boltzmann formula (21):

$$f_\phi = \frac{\exp(-A_s \cdot W_\phi)}{\int_\phi \exp(-A_s \cdot W_\phi) d\phi} \quad (21)$$

345 A_s is a parameter related to the initial susceptibility χ_0 of the magneti-
 346 zation curve:

$$A_s = \frac{3\chi_0}{\mu_0 M_s^2} \quad (22)$$

By employing f_ϕ it is possible to calculate the average magnetization $M(H, \sigma)$ and magnetostriction $\lambda(H, \sigma)$ in the direction of applied field/stress by using equations (14) and (23).

$$\vec{M} = \frac{1}{N} \int_\phi f_\phi \vec{M}_\phi d\phi \quad \epsilon^\mu = \frac{1}{N} \int_\phi f_\phi \epsilon_\phi^\mu d\phi \quad M = \vec{M} \cdot \vec{X} \quad \lambda_{//} = {}^t \vec{X} \cdot \epsilon^\mu \cdot \vec{X}. \quad (23)$$

347 Moreover the following conditions have been used for calculations:

- 348 • The possible directions $\vec{\gamma}_\phi$ are described through the mesh of a unit
 349 radius sphere. A N=34635 points mesh has been used in the present
 350 study.
- 351 • Both materials are modeled. Self consistent localization rules are given
 352 in section 2.1. Final results are given in term of average behavior.
 353 Effect of localization is quickly addressed hereafter.
- 354 • EBSD did not allow the measurement of Fe₂B phase orientations. An
 355 isotropic distribution of this second phase has been assumed. Uniaxial
 356 anisotropy condition allows to consider this phase, in the magnetic field
 357 range tested in this study, as a soft isotropic crystal ($K_1=0$), exhibiting

358 a saturation magnetization of $M_s'^{Fe_2B} = \frac{\pi}{4}M_s^{Fe_2B}$, a saturation magne-
 359 tostriction $\lambda_s'^{Fe_2B} = \frac{1}{2}\lambda_s^{Fe_2B}$ and a magnetic susceptibility $\chi_0^{Fe_2B} = 8000$
 360 (see Appendix A for detailed calculations).

361 • The material parameters used for the modeling of α phase strongly de-
 362 pend on aluminum content. K_1 , λ_{100}^α , λ_{111}^α have already been defined.
 363 The aluminum content allows a direct and theoretical estimation of M_s
 364 ($M_s^\alpha = (1 - at\%Al/100)M_s^{Fe}$). A mix (1:1) of A2 and DO3 structures
 365 has been considered for sample A3 – 22Al α phase (mixture rule of
 366 all physical constants including saturation magnetization). These pa-
 367 rameters are complemented by initial susceptibility χ_0^α which has been
 368 estimated for each phase independently to fit properly the experimen-
 369 tal magnetization measurements. Table 3 gathers the various physical
 370 constants used in the modeling.

371 • The magnetic parameters are complemented by Young’s modulus and
 372 Poisson’s ratio mechanical parameters used to express the mechanical
 373 localization. Young’s modulus and Poisson’s ratio of pure iron have
 374 been chosen (see Table 3).

375 • As already underlined, direction \vec{X} can be restricted to the standard
 376 triangle due to cubic symmetry. The average crystallographic direction
 377 is considered first (see table 1). Figure 9 shows the magnetostrictive
 378 behavior predicted by the model along the longitudinal direction when
 379 this direction is considered. A discrepancy is clearly observed between
 380 saturation values from experiments (table 2 - figure 10a) and model.
 381 This discrepancy can be explained by the fact the average behavior

382 is not obtained along the average direction. The loading direction has
383 consequently been optimized in regards to magnetostriction experimen-
384 tal values. Table 4 gathers the old and new spherical angles used in
385 the modeling of all samples. This optimization leads to small angle
386 variations (except for sample $A2 - 20Al$) or no angle variation (sample
387 $A3 - 22Al$), enough to improve the modeling results of magnetostrictive
388 behavior as illustrated in figure 9 to be compared to figure 10. This
389 direction is kept constant for all loading levels.

Table 3: Parameters used in the modeling of α and Fe_2B phases.

Phase \ Param.	K_1	λ_{100}	λ_{111}	M_s	χ_0	E	ν
Units	kJ.m^{-3}	ppm	ppm	10^5A.m^{-1}	-	GPa	-
$\alpha - A0 - FeB$	48	21	-21	17.1	1000	200	0.3
$\alpha - A1 - 14Al$	35.5	80	-3	14.7	700	200	0.3
$\alpha - A2 - 20Al$	15.5	79	4	13.7	500	200	0.3
$\alpha - A3 - 22Al$	5.25	80	16.5	10.7	400	200	0.3
Fe_2B	0	10	10	9.4	8000	200	0.3

390 4.4. Modeling results

391 Influence of aluminum content

392 Figures 11 to 13 show the result of modeling for magnetization curves under
393 stress, magnetization *vs* stress at different magnetic field levels (same set of
394 values than for experiments), and piezomagnetic sensitivity *vs* stress at the
395 same magnetic field levels. It can be observed that the model reproduces

Table 4: Spherical angles ($^\circ$) corresponding to loading direction \vec{X} : average directions (corresponding to Miller index reported in table 1) compared to optimized directions.

Sample	ϕ_X av.	θ_X av.	ϕ_X opt.	θ_X opt.
<i>A0 – FeB</i>	28	77	31	75
<i>A1 – 14Al</i>	36	73	38	70
<i>A2 – 20Al</i>	23	82	34	76
<i>A3 – 22Al</i>	28	76	28	76

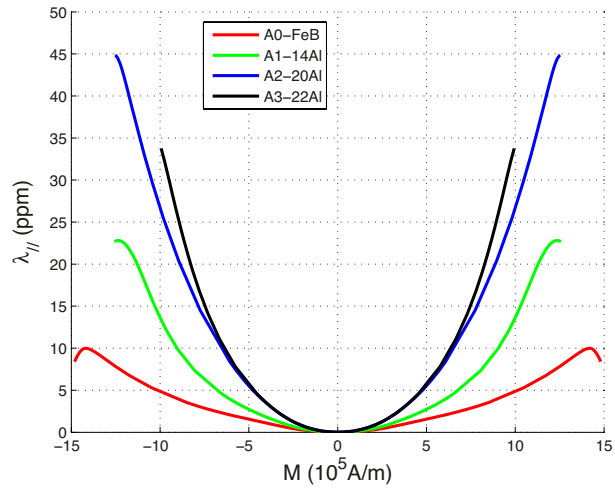


Figure 9: Initial modeling of magnetostriction in the direction of applied field.

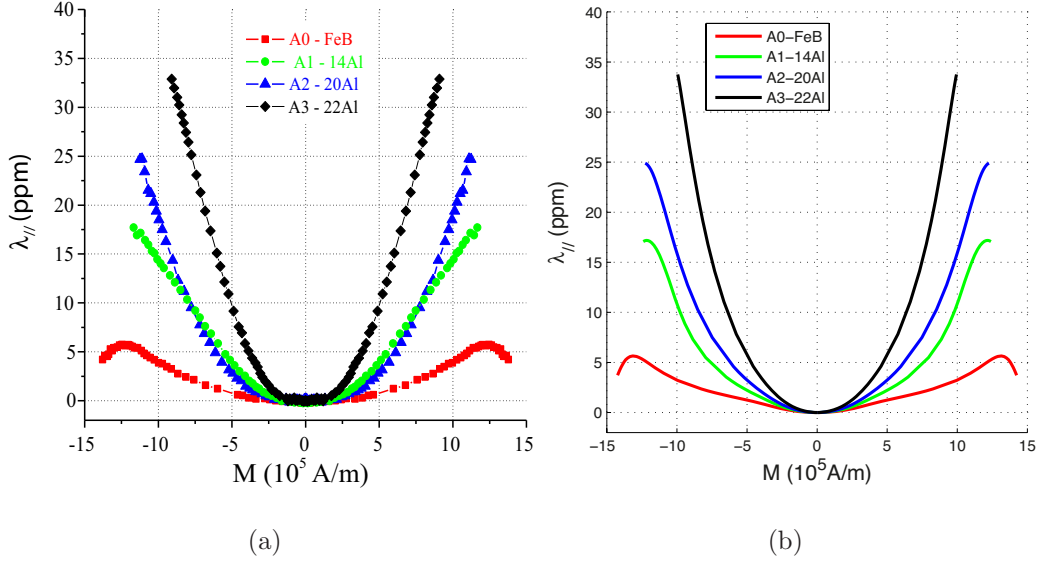


Figure 10: (a) Experimental measurement of the longitudinal magnetostriction of samples $A0$ to $A3$; (b) new modeling of magnetostriction after optimization of applied field direction taking account of behaviors non-linearity.

396 accurately all behaviors. Experimental and modeled values of piezomagnetic
 397 sensitivity are in accordance. The stress and magnetic levels where maximum
 398 piezomagnetic sensitivity is reached are in accordance too. It is nevertheless
 399 observed that $A2 - 20Al$ and $A3 - 22Al$ samples lead to the highest sensi-
 400 tivity in contradiction with experiments where the maximum sensitivity was
 401 reached for $A1 - 14Al$ sample. Such result could be improved by a better
 402 choice of initial susceptibility values for phases. Indeed due to a miss of
 403 experimental data the initial susceptibility of Fe_2B phase remains unknown,
 404 like initial susceptibility of α phase for all samples. This choice is crucial
 405 since initial susceptibility enter in the expression of A_s parameter that de-
 406 fines the sensitivity to field and to stress through their energy expression.
 407 Moreover the calculated values of samples $A0$ and $A1$ that exhibit a $A2$ α

408 phase are much closer to experimental results than samples *A2* and *A3*. It
409 seems clear that due to uncertainty on ordered Fe_3Al phase quantity and to
410 its influence on domain wall mobility, initial susceptibility of alloys *A2* and
411 *A3* matrix is not a confident parameter.

412

413 Effect of Fe_2B phase

414 Figure 14 illustrates the modeled magnetic and magnetostrictive behaviors
415 of both phases and average medium for *A1 – 14Al* sample. This figure il-
416 lustrates the localization phenomenon. Due to the high initial susceptibility
417 of Fe_2B phase, the composite initial susceptibility is higher than the suscep-
418 tibility of the pure α phase. On the contrary, the high uniaxial anisotropy
419 and low saturation magnetization of Fe_2B phase lead to a global decrease
420 of magnetic performances of the composite at magnetic field higher than 2
421 kA/m comparing to pure α -phase. Due to the low magnitude of Fe_2B phase
422 magnetostriction, the magnetostriction magnitude of the composite is usu-
423 ally lower than the magnetostriction magnitude of pure α phase. This point
424 seems in contradiction with results obtained in [9] and [10]. The variations
425 that have been observed were probably related to variations in the samples
426 texture.

427 Figure 15a and 15b allow the comparison of modeled piezomagnetic sen-
428 sitivity of *A1 – 14Al* α phase only, *A1 – 14Al* sample considering localization,
429 and *A1 – 14Al* sample considering homogeneous stress and field for two dif-
430 ferent magnetic field levels (1 kA/m and 2 kA/m). These figures show on the
431 one hand that localization always improve the sensitivity comparing to the
432 homogeneous fields condition. This enhancement effect due to localization

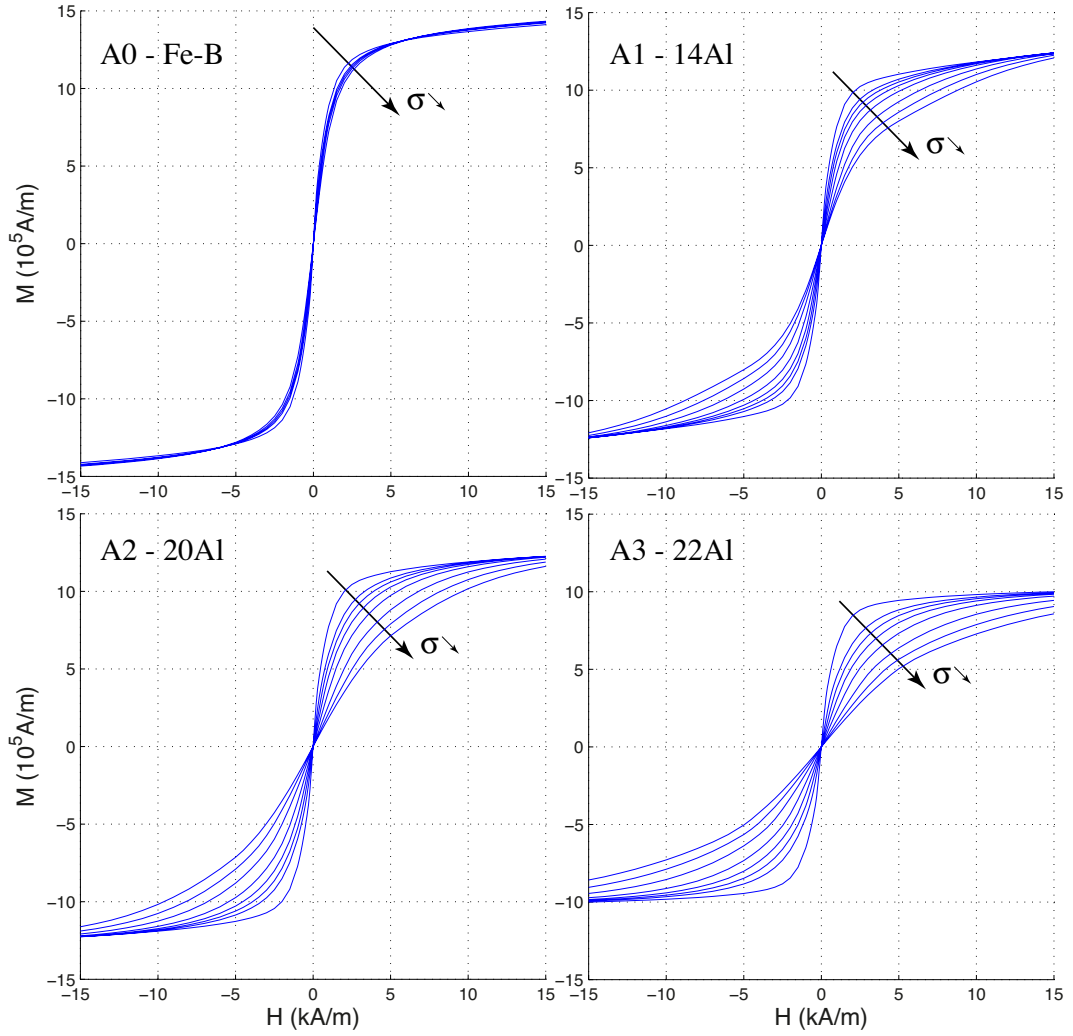


Figure 11: Modeling results: magnetization as a function of the applied magnetic field for fixed values of stresses for all samples ($\sigma \in [50, 0, -15, -30, -50]$ MPa for $A0 - FeB$ sample; $\sigma \in [50, 0, -15, -30, -50, -80, -110, -140]$ MPa for $A1$ to $A3$ samples).

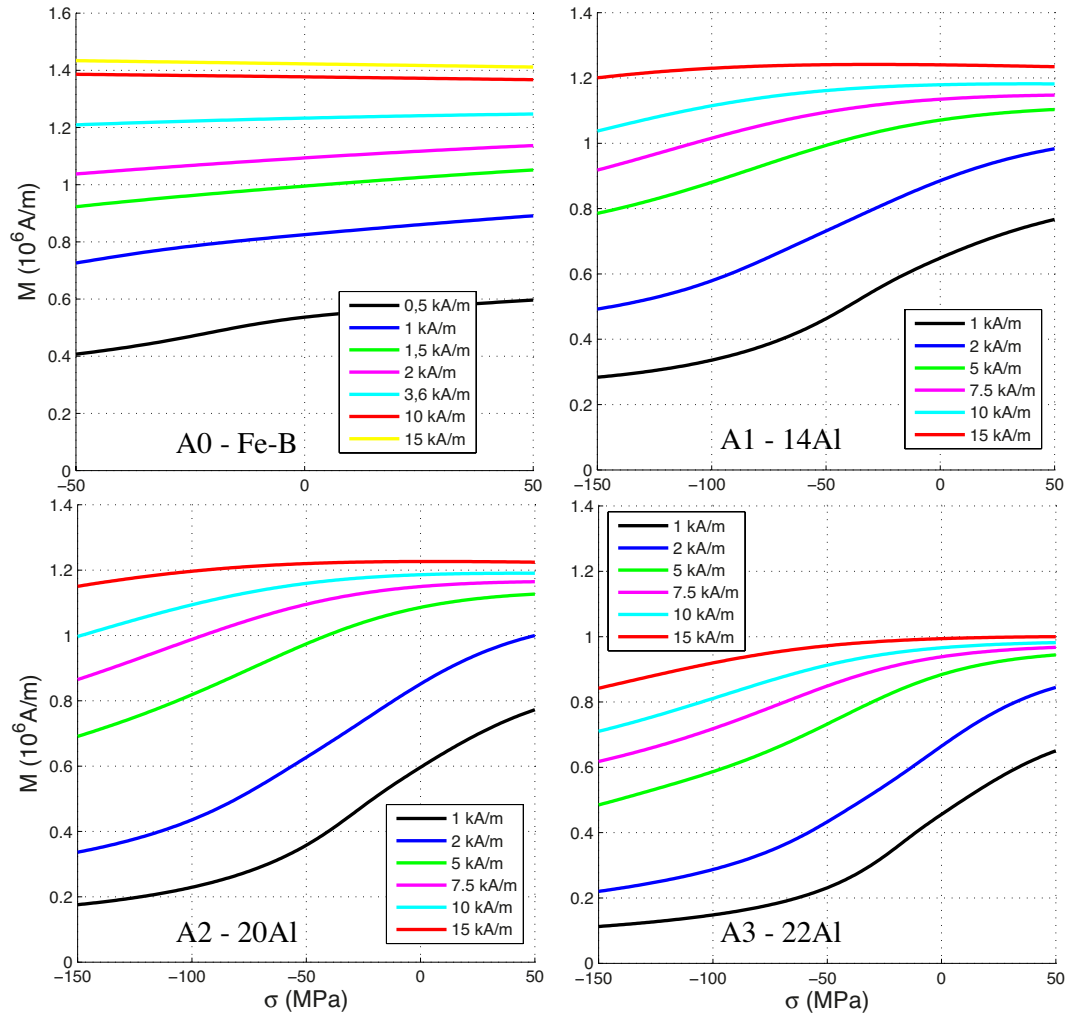


Figure 12: Modeling results: magnetization as a function of applied stress σ for the all samples at fixed values of magnetic field, ($1 \leq H \leq 15$ kA/m).

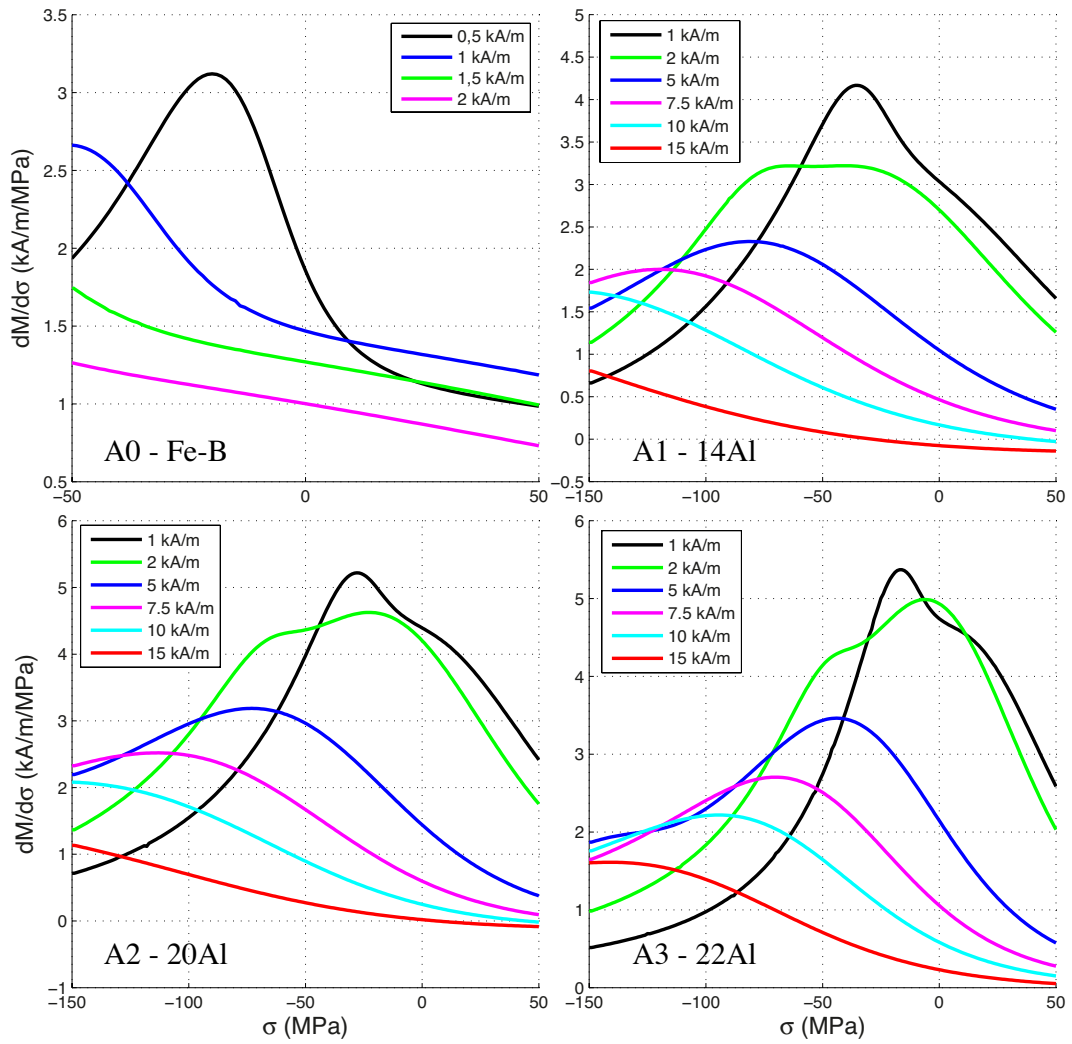


Figure 13: Modeling results: piezomagnetic sensitivity $dM/d\sigma|_H$ vs. σ for all samples at fixed values of magnetic field, ($1 \leq H \leq 15$ kA/m).

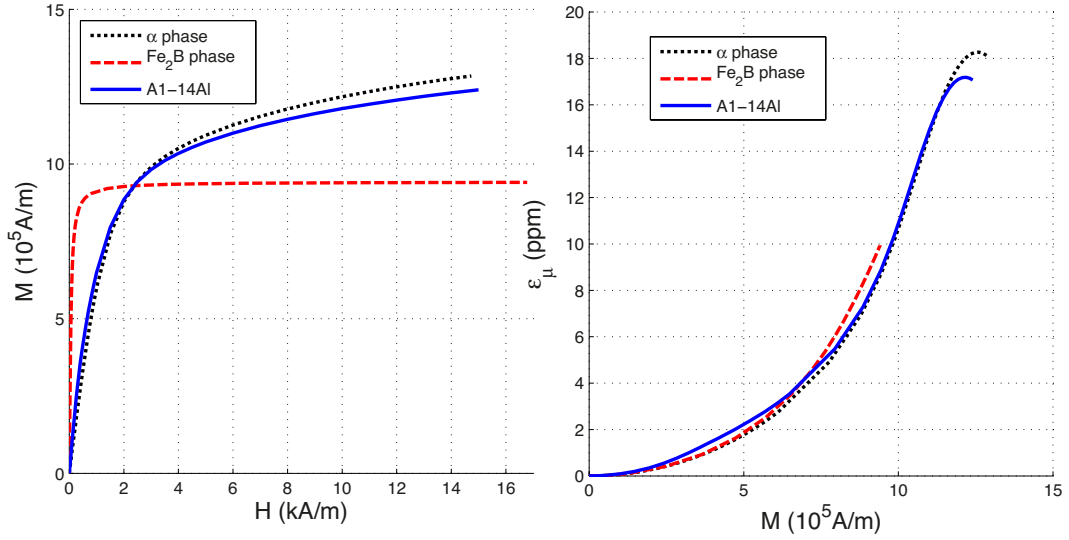


Figure 14: Modeled magnetic and magnetostrictive behaviors of both phases and average medium for $A1 - 14Al$ sample.

433 phenomenon explains the recent interest of scientific community for compos-
 434 ite alloys. These figures show on the other hand, comparing to pure α phase,
 435 that improvement is strongly magnetic field dependent. Indeed, at 2kA/m
 436 magnetic field level, Fe_2B phase saturates. The pure α phase exhibits now a
 437 higher sensitivity than the composite. This effect combined with the reduc-
 438 tion of susceptibility of α phase with increasing aluminum content explain
 439 the existence of an optimum combination of aluminum/boron content and
 440 magnetic field level.

441 Towards an optimization of piezomagnetic sensitivity

442 Improvement of piezomagnetic sensitivity is theoretically possible by choos-
 443 ing the appropriate aluminum/ boron content and texture for the material
 444 matrix. The model can help to reach this goal. Since sensitivity is related to
 445 variation of magnetostriction with field, the basic idea would be to choose the

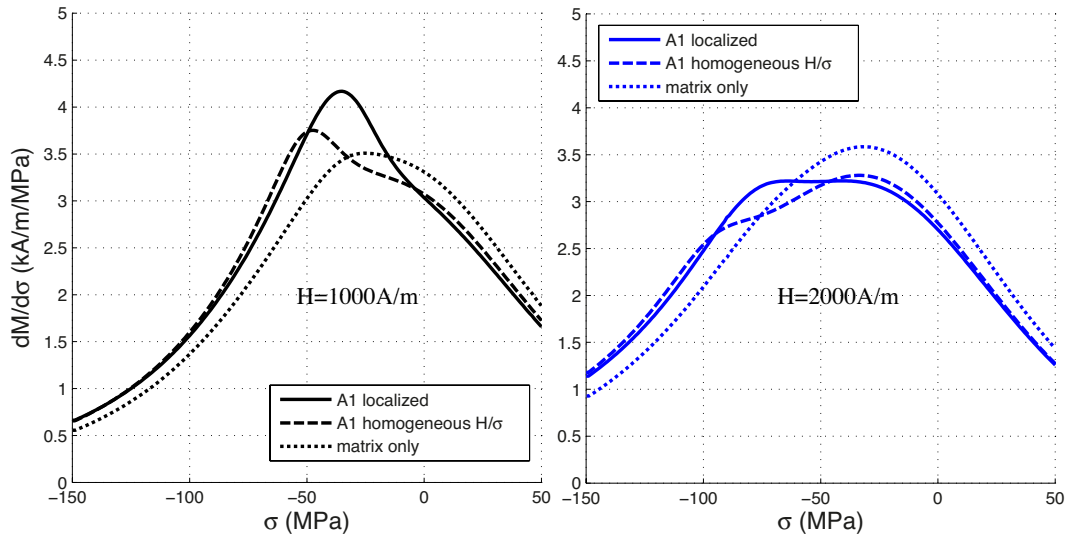


Figure 15: Modeled piezomagnetic sensitivity at two magnetic field level of: $A1 - 14Al \alpha$ phase only, $A1 - 14Al$ sample considering localization, and $A1 - 14Al$ sample considering homogeneous stress and field.

446 composition leading to highest piezomagnetic effect and the crystallographic
 447 direction exhibiting the highest magnetostriction strain. By adjusting the
 448 field, it would be possible to create an improved Fe-Al-B alloys for sensor
 449 application. Alloy with 20%Al content is considered. Magnetostriction is
 450 high along the $\langle 100 \rangle$ direction. We select this direction and model the
 451 piezomagnetic response in figure 16 for various magnetic field levels in a range
 452 of -100MPa to 50MPa. Maximal sensitivity level is about 15 kA/m/MPa.
 453 This value is 3 times higher than the value obtained for $A2 - 20Al$ sample
 454 exhibiting a less favorable orientation. Development of directional solidified
 455 samples seems relevant for a future application of these materials.

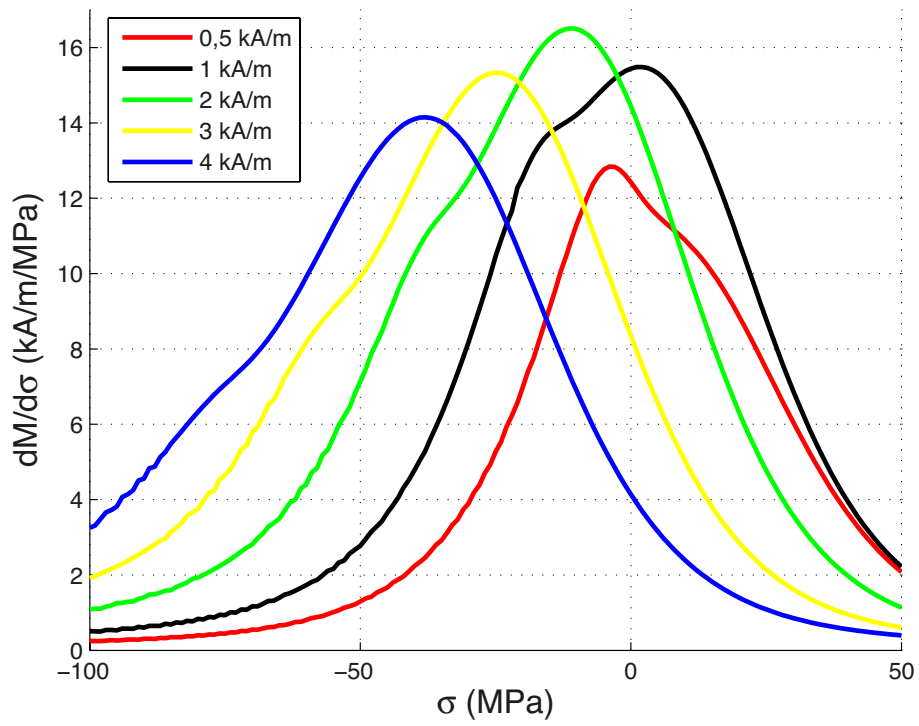


Figure 16: Modeled piezomagnetic sensitivity of ideal $\langle 100 \rangle$ oriented $(Fe - 20\%Al)_{98.4}B_{1.6}$ alloy at different magnetic field levels.

456 5. conclusion

457 Fe-Al-B alloy with 14% of aluminum and 1.6% of boron has a maximum
458 sensitivity value for sensor performance of $4.0 \text{ kA}\cdot\text{m}^{-1}/\text{MPa}$ at -35 MPa for 2
459 kA/m. For the interval from 0 to -100 MPa the minimum sensitivity is about
460 $1.5 \text{ kA}\cdot\text{m}^{-1}/\text{MPa}$. Although the sensitivity is smaller compared to Terfenol
461 D, the range of applied stresses is much higher and the applied field is much
462 smaller [4]. The microstructure of Fe-Al alloys with low quantities of boron
463 addition contains the Fe_2B phase. This phase does not degrade the sensing
464 behavior of these materials, because Fe_2B has a high magnitude of the sat-
465 uration magnetization [11]. Moreover, it enhances the sensing behavior due
466 to both strong magnetic and mechanical localization effects by the increase
467 of the initial susceptibility of the 14% of aluminum alloy at fields lower than
468 $2.0 \text{ kA}/\text{m}$. Despite magnetostriction is strongly enhanced by an increase in
469 aluminum content and magnetocrystalline anisotropy is lowered, the piezo-
470 magnetic behavior is not strongly improved. This can be explained by a lower
471 permeability associated with enhanced magneto mechanical pinning effect.
472 This effect is added to the fact that higher Al contents alloys have also low
473 saturation magnetization magnitudes. An improvement of the piezomagnetic
474 factor to $15 \text{ kA}\cdot\text{m}^{-1}/\text{MPa}$ is predicted, for the alloy with 20% of aluminum,
475 by getting an adequate texture near $\langle 100 \rangle$ directions. This would also
476 favor the actuation behavior of the material. Discrepancies between experi-
477 ments and modeling are due to many modeling approximations at different
478 levels, representativeness of bulk texture from surface EBSD measurements,
479 magnetostatic surface effects that are significant for high grain size materials.
480 Moreover, for alloys with aluminum contents higher than 17% the existence

481 of ordering complicates the analyses. More studies are needed to better un-
482 derstand the effect of Fe₂B on the piezomagnetic behavior of these alloys.

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529 **Appendix A. Magnetic and magnetostrictive behavior of Fe₂B phase**

530 The Fe₂B phase is a tetragonal phase with CuAl₂ (C-16) prototype struc-
531 ture. Even if it is not a well known material, it is possible to find some partial
532 informations about its physical constants, magnetic and magnetostrictive be-
533 havior in references [9, 11, 12] It is a ferromagnetic material with $T_C^{Fe_2B} =$
534 742°C. The anisotropy constant (uniaxial anisotropy) is high reaching $K_1 =$
535 $4.27 \times 10^5 \text{ J}\cdot\text{m}^{-3}$ at room temperature and goes to zero at 251°C [12]. Due
536 to uniaxial anisotropy Fe₂B exhibits an easy (001) plane. The magnetization
537 behavior of isotropic polycrystalline Fe₂B is reported in figure A.17a exhibit-
538 ing a saturation magnetization of $1.2 \times 10^6 \text{ A/m}$ [11]. The magnetostrictive
539 behavior of isotropic polycrystalline Fe₂B is reported in figure A.17b [9] (the
540 demagnetizing field due to form effect - cubic sample - has not been removed
541 explaining the high magnetic field level required to create a significant mag-
542 netostriction strain). The saturation magnetostriction is estimated to be 20
543 ppm. The magnetostriction is supposed isotropic at the grain scale in the
544 paper. It is a strong hypothesis than cannot be verified.

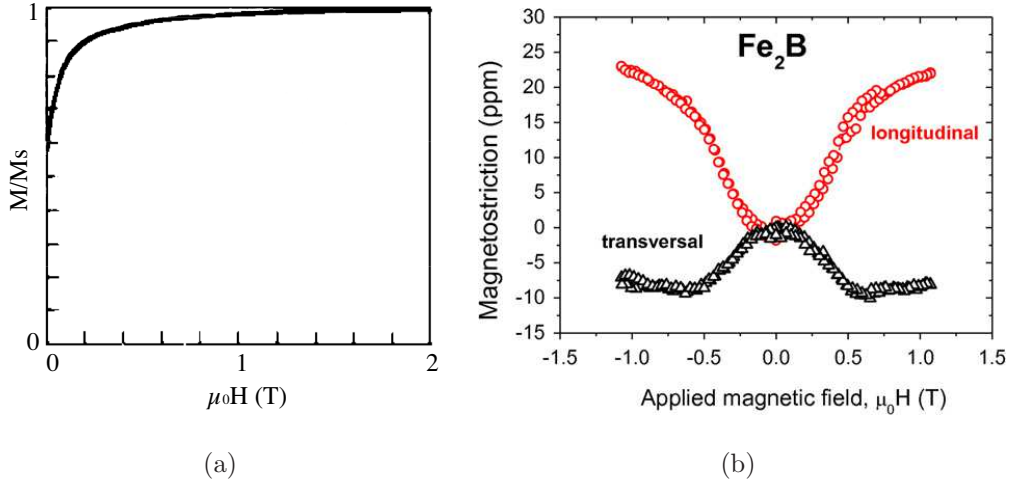


Figure A.17: (a) Magnetization curve of Fe_2B as reported in [11]; Magnetostriction behavior of Fe_2B as reported in [9].

545 Coene *et al.* [11] reported in figure A.17a a very sharp increase of M at
 546 low field. This can be understood by a simple magnetization rotation model
 547 in the easy plane.

548 *Appendix A.1. Theoretical estimation of magnetization behavior*

An isotropic distribution of Fe_2B grains is considered. This means that quadratic \vec{c} -axis is regularly distributed in a unit radius sphere. \vec{c} -axis is given using spherical parameters α_1 and β by:

$$\vec{c} = \begin{pmatrix} \cos\alpha_1 \\ \cos\beta\sin\alpha_1 \\ \sin\beta\sin\alpha_1 \end{pmatrix} \quad (\text{A.1})$$

549 The magnetization vectors are for the same reason regularly distributed in-
 550 side the easy plane of particles. This point is illustrated in figure A.18a: the

551 average magnetization is null.

552

553 The in-plane magneto-crystalline anisotropy is probably very low in accor-
554 dance with experimental data. A weak magnetic field H applied in direction
555 \vec{x} is consequently enough to select the most favorable direction in the easy
556 plane with respect to the magnetic field direction. The selected vector is
557 denoted \vec{u} . It belongs to the (\vec{c} -axis, \vec{x}) plane so that:

$$\vec{u} = \begin{pmatrix} \cos(\pi/2 - \alpha_1) \\ -\cos\beta\sin(\pi/2 - \alpha_1) \\ \sin\beta\sin(\pi/2 - \alpha_1) \end{pmatrix} = \begin{pmatrix} \sin\alpha_1 \\ -\cos\beta\cos\alpha_1 \\ -\sin\beta\cos\alpha_1 \end{pmatrix} \quad (\text{A.2})$$

558 This point is illustrated in figure A.18b. At the saturation \vec{u} rotates
559 progressively in direction to \vec{x} . The average magnetization is the saturation
560 magnetization.

561 For all conditions, calculating an average \vec{u} direction denoted $\langle \vec{u} \rangle$ leads
562 to calculate the associated magnetization supposing that each \vec{u} defines one
563 magnetization direction.

564 • without magnetic field, it is obvious that the average magnetization is
565 null.

566 • under a moderate magnetic field, average $\langle \vec{u} \rangle$ direction can be cal-
567 culated following:

$$\langle \vec{u} \rangle = \frac{1}{2\pi} \int_0^{2\pi} \int_0^{\pi/2} \vec{u} \sin\alpha_1 d\alpha_1 d\beta = \frac{1}{2\pi} \int_0^{2\pi} \int_0^{\pi/2} \sin^2\alpha_1 d\alpha_1 d\beta \vec{x} \quad (\text{A.3})$$

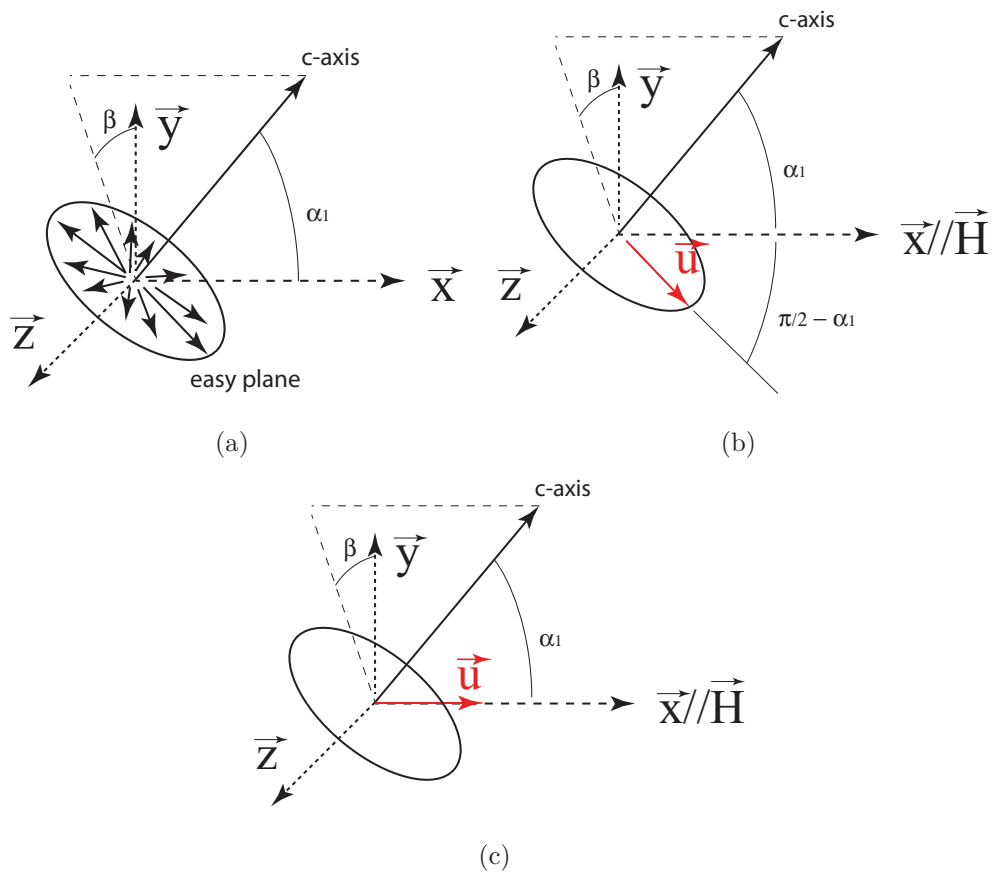


Figure A.18: Illustration of magnetization distribution at different magnetization levels: (a) without magnetic field, multiple directions are regularly distributed inside the easy plane; (b) under low magnetic field, one unique direction inside easy plane is selected before rotation; (c) high magnetic field leads to a magnetization rotation.

we obtain:

$$\langle \vec{u} \rangle = \pi/4 \vec{x} \quad (\text{A.4})$$

The magnetization is given by:

$$\frac{M}{M_s} = \frac{\pi}{4} \approx 0.785 \quad (\text{A.5})$$

568

This result is in accordance with the results of Coene *et al.* [11].

- at saturation, the following result is obviously

$$\langle \vec{u} \rangle = \vec{u} = \vec{x} \quad \text{and} \quad \frac{M}{M_s} = 1 \quad (\text{A.6})$$

569 This magnetization can be achieved with a field high enough to over-
570 come the uniaxial anisotropy (about 2T following Coene's results [11])

571 *Appendix A.2. Theoretical estimation of magnetostriction*

572 A theoretical estimation of magnetostriction is possible following the pro-
573 cedure used for the magnetization, using the descriptions in figure A.18. At
574 zero applied field, the magnetization vectors are regularly distributed inside
575 the easy plane of particles, the average magnetostriction is null. As discussed
576 above, a weak magnetic field H applied in direction \vec{x} is enough to select the
577 most favorable direction in the plane with respect to the magnetic field di-
578 rection. The selected vector is \vec{u} given by equation (A.3). The components
579 of \vec{u} are the direction cosines γ_i of the magnetization. They are used in the
580 definition of the magnetostriction tensor for isotropic conditions (as supposed
581 for Fe₂B):

$$\epsilon_{\mu}^{\vec{u}} = \frac{3}{2} \lambda_s \begin{pmatrix} (\gamma_1^2 - \frac{1}{3}) & \gamma_1 \gamma_2 & \gamma_1 \gamma_3 \\ \gamma_1 \gamma_2 & (\gamma_2^2 - \frac{1}{3}) & \gamma_2 \gamma_3 \\ \gamma_1 \gamma_3 & \gamma_2 \gamma_3 & (\gamma_3^2 - \frac{1}{3}) \end{pmatrix} \quad (\text{A.7})$$

582 An average magnetostriction tensor is calculated now so that:

$$\langle \epsilon_{\mu}^{\vec{u}} \rangle = \frac{1}{2\pi} \int_0^{2\pi} \int_0^{\pi/2} \epsilon_{\mu}^{\vec{u}} \sin \alpha_1 d\alpha_1 d\beta \quad (\text{A.8})$$

583 After few calculations the following result is obtained:

$$\langle \epsilon_{\mu}^{\vec{u}} \rangle = \begin{pmatrix} \frac{\lambda_s}{2} & 0 & 0 \\ 0 & -\frac{\lambda_s}{4} & 0 \\ 0 & 0 & -\frac{\lambda_s}{4} \end{pmatrix} \quad (\text{A.9})$$

584 so that the longitudinal magnetostriction reaches $\lambda_s/2$ at very low field
 585 for a magnetization of about $0.785 M_s$.

586 At the saturation, \vec{u} rotates progressively in direction to \vec{x} . The average
 587 magnetostriction is the saturation magnetostriction λ_s itself. This is reached
 588 for a very high magnetic field level.

589 *Appendix A.3. Consequence in term of modeling for present paper*

590 The magnetic field level used in the experiments presented in the paper
 591 is not enough to begin the rotation mechanism. For simplicity reasons, the
 592 in easy plane rotation mechanism has only been considered for the modeling
 593 of magnetic and magnetostrictive behaviors of the Fe₂B phase (see section
 594 4.3). Fe₂B phase has been considered as an isotropic very soft phase ($K_1=0$;
 595 $\chi_0=8000$) with apparent saturation magnetization $M_s^{Fe_2B}=(\pi/4)M_s^{Fe_2B}=9 \times$
 596 10^{-5}A.m^{-1} and apparent saturation magnetostriction $\lambda_s^{Fe_2B} = \frac{1}{2}\lambda_s^{Fe_2B}=10$
 597 ppm. Figures A.19a and A.19b show the obtained modeled magnetization
 598 and magnetostrictive behaviors in the magnetic field range of the work pre-
 599 sented in the paper. These modeling are in accordance the few available
 600 experimental data for this material.

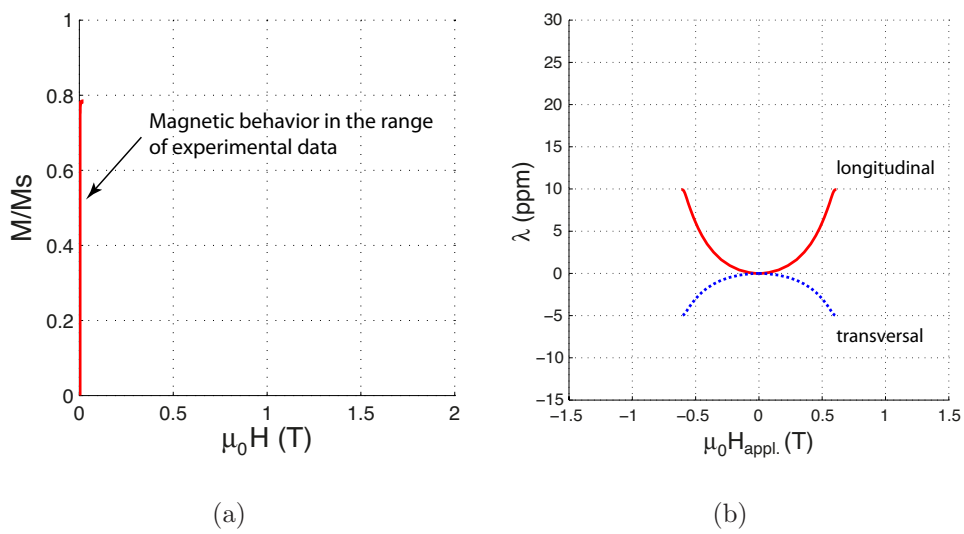


Figure A.19: Modeled magnetization and magnetostrictive behavior of Fe₂B phase in the magnetic field range of the work presented in the paper - to be compared to experimental results reported in figures A.17a and A.17b (a cubic form effect is considered to define the applied magnetic field in figure (b)).